GENERATION AND ACCELERATION OF ULTRASHORT ELECTRON BUNCHES

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ABSTRACT

Creating a simple and efficient electron accelerator is a great challenge for many physicists and engineers around the world. Many applications ranging from television and computer screens to highly sophisticated electron microscopes utilise an electron beam in some way.

This work presents a novel and simple method for the generation and acceleration of short electron bunches that can be further developed into an efficient electron accelerator. High-power femtosecond laser pulses have been used to excite surface plasmons in 50-nm silver and gold films. Nonlinear excitation results in the emission of an electron bunch through a combination of multiphoton excitation at laser power up to a few GW cm$^{-1}$ and laser-induced tunnel emission at laser powers above this threshold. The energies of photoelectrons are found to extend as high as 0.4 keV. The electron emission occurs on the femtosecond time-scale. On average each electron bunch contains approximately 40 fC of charge corresponding to $2.4 \times 10^4$ electrons per bunch. Calculations show that the high energies are due to ponderomotive acceleration in an evanescent electromagnetic field that extends from the metal film out into the vacuum. Analysis of the initial conditions of the accelerating process reveals that the electrons can gain additional energy depending on the phase of the evanescent field during the electron liberation process. It is shown that the experimental arrangement can be further simplified if the cathode consists of an array of small metallic protrusions. The theory and experiment suggests that it is possible to accelerate electrons to even higher energies in the new arrangement. This opens the opportunity of employing the new acceleration scheme into various time-resolved techniques.
CHAPTER 1

INTRODUCTION

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CHAPTER 1

INTRODUCTION

1. 1. PREFACE

The research described in this work was started after I was advised to read the N. Scherer group paper. [1] At the beginning it seemed to be a simple and easy idea for generating short electron pulses. As the experiments progressed, the whole picture of the generation of an ultrashort electron bunch become more complicated and at the same time more intriguing. It was evident that a novel scheme of electron acceleration was being observed and this has not been described before. Therefore, a theory of the observed acceleration process was developed and compared with experimental data. After close examination of the energy spectra of ultrashort electron bunches in light of the theory, it was concluded that an other, more fundamental electron-electromagnetic-wave interaction was being observed. Therefore, the description of the electron emission in the presence of intense ultrashort laser pulses was attempted. Simultaneously, attempts were undertaken to further simplify the experimental arrangement, obtain higher acceleration rates and demonstrate possible applications of the experimental idea. There have been many successful experiments together with those that did not work at all. Therefore, this work is more a report of the steps of discovery and investigation of generation and acceleration of ultrashort electron-pulse rather than the systematic description of an experiment.

1. 2. MOTIVATION

Electron beams are used in many experimental techniques as a powerful tool to investigate properties of matter. Continuous electron beams are used in many areas of contemporary physics to provide sub-micron imaging of a solid surface or to investigate its atomic structure in various schemes of electron microscopy. Copper or tungsten electron cathodes are used commonly in electron microscopes to observe features on a surface on a nanometer scale or investigate the crystallographic
structure of a solid. Due to the nature of commonly used cathodes, usually a continuous, high density beam of electrons is obtained as a result of a thermionic or field emission in the presence of high voltage applied to the target. To obtain electron pulses, an originally continuous beam needs to be modulated externally.

Another method of generating an electron beam is the photoelectric effect. An electromagnetic wave that interacts with a solid target can excite electrons out of it. Recent advances in femtosecond laser technology provide an opportunity for generating short electron pulses and afterwards a possibility of constructing a time-resolved experiment where one of the beams could be a femtosecond electron beam. The main challenges involving standard methods of generating electrons with laser pulses are the possibility of ion emission if the damage threshold of the material is exceeded, low electron energy and a wide angle of emission.

This thesis proposes a novel and simple method for the generation and acceleration of ultrashort electron pulses. The experiments presented here describe the electron interaction with intense ultrashort laser pulses at a metal surface and the basic theory of the electron interaction with an evanescent optical field. The inspiration of this work was a paper published by Norbert Scherer and co-workers [1] where new pomp-probe techniques were reviewed. In this work it was demonstrated that it is possible to detect a time-resolved signal of photoemitted electrons when a femtosecond laser beam was incident on a metal surface at the surface-plasmon (SP) resonance angle. The signal was attributed to a surface plasmon assisted multiphoton ionisation process. This showed the possibility of creating a time-resolved electron spectrometer that can serve as a surface sensitive probe that uses short electron pulses. It will be shown that a simple multiphoton emission model does not fully describe the electron emission process in the presence of an intense and ultrashort laser pulse. As a result of our investigation, a novel technique of accelerating electrons in the optical field was developed, and the analysis of electron excitation and emission in the presence of an intense ultrashort laser pulse was attempted. It is evident that short electron bunches were generated and that these can be used as a tool to probe chemical reactions at the metal surface.

The uniqueness of the experiments and theory presented in this work lies in the novelty of the electron acceleration scheme combined with the simplicity of the
experimental arrangement. It was proved that it is possible to accelerate electrons in a fast oscillating field. The experimental arrangement was simple and involved only a standard glass prism with a thin metal film evaporated on one side. Single 800 nm laser beam was used to excite electrons out of the metal layer and subsequently accelerate them. It was shown that the optical electric field could be enhanced in the presence of SPs at the metal/vacuum interface. Further methods of enhancing the electric field at the metal surface were proposed. The electron emission spectra were analysed and the analytical derivation of the equation of motion of a charged particle in the evanescent field was solved. It was proven that the accelerating force in this process arises from changes of intensity of the laser beam across the travel path of an electron and this proves that the force has a ponderomotive character. The theoretical considerations were confirmed by numerical simulations of the electron motion in an evanescent field and subsequently compared with experimental results. Furthermore, another accelerating force was identified in the evanescent-wave acceleration process that arises from the initial conditions of the electron-evanescent field interaction. These results suggested that further detailed analysis of the electron emission from a solid in the presence of an intense ultrashort laser pulse is necessary.

1.3. OUTLINE OF WORK

The structure of the work is as follows. The two first Chapters (Chapter 2 and 3) are an introduction to the experiments presented later. In Chapter 2 all experimental techniques and methods are described in detail. The sample preparation method, experimental arrangements, and acquisition techniques are presented. Laser systems used throughout the experiment are described. Furthermore, the method and the precision of the time-of-flight technique is explained. In Chapter 3, the basics of the theory of SP are reviewed. The emphasis is put on the electric field enhancement resulting from SP excitation at the metal/vacuum interface. Chapter 4 analyses the first experiments that could not be explained using the current electron emission theory in the presence of SP at the metal surface and leads to the development of the evanescent field acceleration theory. It argues that the commonly accepted theory of electron emission in the presence of SPs via the multiphoton ionisation process is inadequate. Additionally, the analysis of the electron dynamics inside a thin metal
film in the presence of an intense ultrashort laser is outlined. Subsequent chapters explain the experimental results presented in Chapter 4. Chapter 5 presents an analytical solution of the equation of averaged motion of an electron in the optical evanescent field. The final energy of an averaged motion of an electron is found and identified as the ponderomotive energy. It is shown that the final electron energy depends on the intensity of the incident laser beam, the frequency of the driving field and the length of the interaction with the evanescent field that is defined by the optical pulse duration. Chapter 6 presents the experimental results of electron emission spectra from flat silver and gold films. The spectra are analysed as a function of intensity of the incident beam as well as the pulse duration and confirm the analytical theory presented previously. Additionally, a numerical model of the electron motion in an evanescent field is presented. This model suggests that initial conditions of the electron motion play an important role in the acceleration process and argues that there is another force that contributes to the overall acceleration. Chapter 7 analyses the electron liberation process in the presence of the intense and ultrashort laser pulses. The analytical considerations identify the additional accelerating force that arises from the value and direction of the vector potential of the optical field at the moment of the electron liberation. It is evident that further detailed analysis of the electron emission process is necessary in the presence of intense and ultrashort laser pulses. Chapter 8 describes a method of further enhancement of the SPs’ electric field. The electric field enhancement at rough metal surfaces is discussed. The results of the electron emission from rough metal surfaces are presented and analysed.

CHAPTER 2

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CHAPTER 2

OVERVIEW OF EXPERIMENTAL TECHNIQUES

In this chapter all of the experimental techniques are described. The methods used for sample preparation are given in detail. The methods used for thin metal film evaporation, the preparation of rough metal films through baking the sample and the nano-pyramid deposition and the deposition of a molecular monolayer are also explained. The schematic drawings and pictures of the experimental arrangement are presented and examined. The electron detection systems from the simple self-build array detector to time-of-flight spectrometer are described. Finally the details of laser systems used throughout the whole work are given and the operating principles of femtosecond laser systems are reviewed.

2. 1. SAMPLE PREPARATION

2. 1. 1. FLAT METAL SURFACES

Two different sized right angle, BK7 prisms were used in experiments involving flat metal surfaces. The side lengths of the prisms were 15 mm and 40 mm respectively. The hypotenuse faces of the prisms were cleaned in the standard way [1]: the sample was washed with Triton x-100 detergent and boiled for 5 minutes in ethanol or methanol to remove any possible contamination. To remove the final residue, the sample was ultrasonicated in distilled water for at least 15 minutes. Next, the sample was removed from the water, left to dry and mounted in the evaporation chamber.

The distance between the target and the evaporation source was 30 cm. 99.99% pure silver or gold (Goodfellow Cambridge Ltd.) was used throughout. Before placing in the vacuum chamber the effusive source was rinsed in methanol to remove possible contamination. A standard tungsten filament was used to evaporate gold whereas a molybdenum boat was used for silver evaporation. The chamber pressure for the whole process was maintained below \(10^{-5}\) Torr. The deposition rate was
maintained at 0.2 nm/s. After the evaporation process the sample was left for half an hour in the vacuum chamber to settle. Immediately afterwards it was transferred to one of the spectrometers and mounted as described below.

To monitor the morphology of the metal film the surface was examined using an Atomic Force Microscope (AFM). The AFM images were produced using an etched Si nanoprobe in tapping mode with spring constant of approximately 0.2 N/m. A typical AFM image (300 nm x 300 nm) is shown in FIG. 2.1.

![AFM image of the clean gold surface](image)

**FIG. 2.1.** AFM image of the clean gold surface; image was taken in the tapping operation mode; the scan range is 300 nm by 300 nm and the maximum height of the surface features is 10 nm; the tip oscillation amplitude was 0.05 V.

### 2.1.2. ROUGHENING OF THE METAL FILM

In order to create a rough metal surface, flat metal films were heated on the substrate up to a set temperature below the melting point. This process reorganised the flat metal film and helped to spontaneously form small droplets of metal on the glass substrate. The metal films were then heated in air at a rate of 5 C/min up to a temperature of 300°C. The melting point of gold and silver is 1064.18 and 961.78°C respectively. Afterwards the sample was cooled down slowly overnight in the oven chamber to minimise the possible heat strain effects in the glass. The resulting layer was transparent and the size of the formed droplets was difficult to determine with
optical microscopy suggesting that the size of metal particles was less than the wavelength of visible light. The conductivity of the formed particle layer was very small and further surface characterisation using scanning electron microscopy was therefore not feasible. Furthermore the prism sample was not suitable for AFM imaging.

2. 1. 3. DEPOSITION OF MOLECULAR MONOLAYERS

Self-assembling molecular films were deposited onto flat metal surfaces using the immersion method [2]. A few crystals of the hexadecanethiol (Aldrich) were dropped separately into 100 ml of ethanol and diluted. Once the crystals did not dissolve any longer the solution was thought to be saturated. Flat metal films were immersed into the solution and left overnight. Afterwards the samples were removed from the solution rinsed with methanol and left to dry. A typical AFM image (300 nm x 300 nm) of a gold surface coated with hexadecanethiol is presented in FIG. 2.2.

![AFM image](image.png)

**FIG. 2.2.** The AFM image of flat gold surface covered with a monolayer of hexadecanethiol molecules; scans were taken in the tapping operation mode, maximum height of surface features is 10 nm, the tip oscillation amplitude was 0.1 V
2.1.4. NANO-PYRAMID ARRAY DEPOSITION

The nanolithography method used to deposit a nano-pyramid array was first developed by J.H. Hulteen et al. [3]. A 30% solution of 300±5 nm polystyrene nanospheres (Polymer Laboratories Ltd) were diluted (9:1 by volume) in a solution of methanol and surfactant Triton X-100 (1:400 by volume). The surfactant was used to assist the solution in wetting the substrate. Indium tin oxide (ITO) coated glass (Flabeg Holdings GmbH) was used to provide the conductive substrate for the nano-pyramid array. The resistivity of the oxide film was 15 Ω per square. The nanosphere/methanol solution was spin coated onto a cleaned substrate at 3600 rpm and transferred to the evaporation chamber.

FIG. 2.3. The schematic drawing of the evaporation chamber with the apertures used to collimate the evaporated atom beam indicated; the sample (target) was mounted onto a cold finger cooled with liquid nitrogen (LN₂).

The schematic diagram of the evaporation chamber is presented in FIG. 2.3. The sample was mounted on a cold finger 30 cm above the effusive source. Three 12 cm diameter apertures were spaced regularly between the source and the sample, which aided collimation of the particle stream. Before the evaporation, the sample was
cooled with liquid nitrogen to prevent migration of metal particles during the sedimentation process. A 40 nm of silver at a rate of 0.2 nm/s was deposited at a pressure of $10^{-5}$ Torr or below. The deposition rate and thickness of the evaporated metal was monitored with a standard quartz film thickness monitor. After deposition the sample was left in the chamber for about half an hour to let it return to the ambient temperature and allow reorganisation of the evaporated film in vacuum. [4, 5] The polystyrene nanospheres were removed from the substrate by ultrasonicating the sample for about 2.5 minutes in CH$_2$Cl$_2$ until they dissolved. Afterwards the samples were mounted onto one of the spectrometers.

The morphology of the samples was examined using an Atomic Force Microscope. The ITO sample (originally 20 mm by 30 mm size) was broken into small pieces to enable its convenient mounting onto a standard AFM stud. All images were produced in air in the contact mode. A standard Si probe was used during the experiments. The probe height was 10-15 µm, thickness 2 µm and the typical force constant was 0.2 N/m with a resonant frequency of 13 Hz. A typical AFM image of a nanopiramid layer (11 µm x 11 µm) is presented in FIG. 2.4.

**FIG. 2.4.** AFM image of nano-pyramid array, scan range 11 µm by 11 µm, scan acquired in contact mode; to improve resolution the scan was taken using 8 substeps.
2. 2. TYPE OF MEASUREMENTS

2. 2. 1. ELECTRON EMISSION DEPENDENCE ON INCIDENCE ANGLE OF LASER BEAM

The optimum angle of the electron emission was found by measuring the total emitted photocurrent under repetitive irradiation by a femtosecond laser beam for a series of angles of incidence. Before performing the experiment the Maxwell equations for TM-polarised light at the thin film boundaries were solved. From the given complex refractive indices of metal and glass [6], the optimum thickness of the metal film was found. The resonance angle for attenuated total internal reflection of an optimum metal film thickness was calculated as well the optical field enhancement.

![Schematic diagram of the electric circuit used in the measurement of the dependence of laser beam incidence angle on electron emission intensity. The measurements were carried out either in air or vacuum. The pivot axis of the sample-collecting electrode arrangement is indicated.](image)

Samples, prepared in the standard way (see section 2.1), were placed on the rotational stage with an angular resolution of one second. The rotational axis of the stage was chosen to be in the plane of the metal film or the NP layer as shown in FIG. 2.5. The collecting electrode was either a thin tungsten tip sharply pointed with the apex diameter of a few micrometers or a flat copper plate. The electrode was electrically isolated and mounted on a rotational stage at a distance of between 3 cm to 2 mm from the sample. During the experiment the metal or ITO layer was grounded while the electrode was connected to the lock-in amplifier. The lock-in detection was chosen to improve the signal-to-noise ratio. The chopping frequency of the laser beam (the reference signal to the lock-in amplifier) was about 1.5 kHz, so
chosen to avoid possible interference with other high frequency light sources in the laboratory. Measurements were performed either in air or in vacuum.

2.2.2. SPATIAL CHARACTERISATION OF AN ELECTRON BEAM

The angular distributions of the photoelectrons, both in the plane of incidence of the laser and perpendicular to it, were measured with an array detector. The photograph of an array detector prototype is presented in FIG. 2.6.

![Array Detector Prototype](image)

**FIG. 2.6.** The top view of the 2.5 cm diameter array detector with the prism in the holder, whole assembly is placed in the interior of custom build vacuum chamber; all electrical connections are made via standard vacuum feedthrough.

Ten copper strip electrodes were placed uniformly along half of a circumference of a circle of radius 2.5 and 10 cm as shown in the FIG. 2.7. The sample prepared in the standard way (as described in section 2.1) was placed in the centre of the detector with the metal film facing the electrodes with the circle central axis in the plane of the film, see FIG. 2.8. The sample with the detector was placed inside the vacuum chamber. The peak power density of the 800 nm laser beam at the metal surface was 0.4 GW/cm$^2$ at 150 fs laser pulse duration and 250 kHz repetition rate. The focused beam entered the chamber through a standard vacuum window, hit the back of the prism and was focused on the metal layer. The laser beam was adjusted to coincide with the central axis of the detector array, see FIG. 2.7. Before the chamber was
evacuated the resonance angle was found by rotating the sample-detector arrangement along its axis to visually locate the SP resonance angle. The correct SP resonance angle was confirmed by observing the image of the reflected beam where a dark sharp vertical line appeared across it. All experiments were carried out at the pressure of $10^{-6}$ Torr or less. Once the vacuum chamber was evacuated, the maximum electron emission angle was found by adjusting the laser beam position using one of the mirrors. This final adjustment was less than 1 degree. During the measurements the metal layer was grounded and one of the electrodes was connected to the lock-in amplifier and DC power supply. All other electrodes were grounded. The voltage was varied from +20 V to –20 V. Each measurement took approximately 2 minutes, with data acquired every 0.3 s. As a final result, the average value for one data set for one electrode was taken to obtain the best signal-to-noise ratio.

![Diagram](image)

**FIG. 2.7.** Top (A) and side (B) view of two different arrangements of an array detector and the sample; (A) was used to measure the horizontal distribution of emitted electrons and (B) to measure the vertical one; for simplicity electrical connections are not shown.

### 2.2.3. DEPENDENCE OF AN ELECTRON BEAM ON LASER INTENSITY

A lock-in amplifier was used to measure the total photocurrent as a function of the incident laser intensity. The typical experimental arrangement is presented in FIG. 2.8. The sample was mounted inside a vacuum chamber with a 2.5 cm diameter copper disc as an collecting electrode placed 3 cm away, parallel to the metal layer. During all experiments the pressure was less then $10^{-6}$ Torr. The metal layer was grounded during the entire experiment. The collecting electrode was electrically insulated from the vacuum chamber and connected to the lock-in amplifier and DC power supply. Shielded coaxial cables or twisted pairs were used to minimise
interference effects from the surroundings. The voltage was varied between +30 V to –20 V. The laser beam entered the chamber through a standard vacuum window, reached the back of the prism and struck the metal layer at its resonance angle. The rough estimation of this angle was found in the same manner as above and afterwards adjusted precisely using one of the mirrors. The intensity of the laser beam was adjusted using a motorised half-wave retardation plate and polarising cube. The half wave retardation plate was rotated at low speed (0.3 motor setting) where the rotation axis was coaxial with the laser beam direction. This provided a continuous change of the angle of polarisation of the incident beam. The polarising cube allowed the transmission of TM polarised light only, therefore the laser light intensity at the sample was adjusted continuously. Later, the laser beam was focused with a 50 cm lens and the sample was placed at its focal point. Maximum intensity of the laser beam at the metal layer was approximately 50 GW/cm² at 150 fs laser pulse duration and 250 kHz repetition rate. A Si photodiode was used to monitor the laser beam intensity. Outputs of the photodiode and lock-in amplifier were connected to a computer to collect the data. For a given voltage each data set was acquired for at least 30 minutes. This resulted in approximately 230 scans through the whole intensity range. Consequently each scan was averaged to improve the signal to noise ratio.

**FIG. 2.8.** Schematic diagram of the experiment to measure the average photocurrent as a function of the intensity; for simplicity the electrical connections are not shown.
2.2.4. TIME-OF-FLIGHT MEASUREMENTS

The Time-of-Flight (TOF) technique was employed to acquire the energy spectrum of the electron bunch. Three different TOF spectrometers were used. Each TOF spectrometer was equipped with a sample holder, stainless steel vacuum flight tube and a multichannel array plate (MCP) as a fast electron detector.

2.2.4.1. MULTICHANNEL ARRAY PLATE

The active device of the MCP (AOF-TOF-18, Galileo Corporation) is an array of silicon microtubes that act as an electron amplifier. The diameter of each tube is 5 µm and centre-to-centre spacing is 6 µm and the resulting diameter of the MCP is 18 mm (as indicated in FIG. 2.9). This yields a 2.52 cm² active detection area for the device. In the electron-detecting mode the entrance grid is placed directly in front of the microchannel array at ground potential. The typical transmission of the entrance grid is 90%. Once the electron from the sample reaches the grid it feels a 600 V attractive potential and is accelerated toward the detector array. When the initial electron strikes the microtube it cause the emission of secondary electrons inside it. An accelerating potential of 2400V is applied across each microtube that causes avalanche emission and acceleration of secondary electrons, resulting in the MCP gain. The gain of the device used in experiments presented here is $10^7$ at 2400 V. The typical pulse width at FWHM is 690 ps. The schematic drawing of the MCP layout at the maximum voltage values that can be applied to it are shown in FIG. 2.9.

![FIG. 2.9. Schematic drawing of MCP layout, not up to scale; maximum values of voltages that can be applied across the device are indicated.](image-url)
2.2.4.2. TOF ACQUISITION METHOD

Time-of-Flight spectra were measured using three different TOF spectrometers. One of the experimental arrangements is shown in FIG. 2.10. In each case the sample was mounted on the outside of the vacuum chamber with the metal film or a nano-pyramid layer directed towards the interior, permitting the electrons to travel through a hole toward the detection system. In experiments where an external voltage was applied, a 175 µm square opening mesh copper grid was fitted 2 mm in front of the sample. The detection system was placed at a distance of 20 cm, 32 cm or 55.5 cm away from the sample. The laser beam entered the prism through one of the faces opposite the hypotenuse in order to excite SPs in the metal film. To determine the SP resonance angle, first the normal reflection from the first prism interface was found. Next, the mirror on the translation stage was moved and rotated to allow the beam to reach the metal layer at the SP resonance angle that was found in previous experiments (see sections 2.2.1 and 2.2.2). The image of the reflected beam from the metal film was observed. At the SP resonance angle the whole area of the image appeared to be darker when the incident beam was collimated, or a sharp dark line appeared across a bright image when the beam was focused. When the glass slab with nano-pyramids was mounted, the beam reached the back of the sample at the same angle as the SP generation angle in order to preserve the same laser spot size and profile as well as to prevent photoelectron generation at the MCP grid excited by the laser pulse. The triggering photodiode was placed at exactly the same optical path length from the beam splitter as the sample. The laser power was adjusted using either a combination of a half wave retardation plate and a polarising cube or a set of reflective neutral density, 1 mm thick filters. When no external voltage was applied, the sample was grounded to the vacuum chamber all the time with the shortest possible connection to avoid stray capacitance in the circuit. When the external voltage was applied, the sample was electrically insulated form the vacuum chamber and a negative bias was applied to it with respect to the ground. The MCP entrance grid was grounded at all times. In addition, electrical connections from the triggering diode and MCP to the oscilloscope were kept the same length.
FIG. 2.10. The photograph of the TOF set-up; 1: finely adjustable mirror mounted on the translation stage, 2: the detection Ge photodiode with set of ND filters in front to avoid saturation, 3: set of $\lambda/2$ retardation plate and a polarising cube to adjust the intensity of the incidence beam at the prism, 4: the length of the TOF tube, 5: MCP mounted on the TOF tube; the laser beam path is indicated; the reflection from the first prism interface is taken as a reference beam to trigger the oscilloscope, note that the position of the photodiode need to be changed.

The data were acquired using a 1GHz, 5GSample/s TDS 630C Tektronix oscilloscope triggered with a germanium photodiode. Self-built LabVIEW software was used to acquire the data. When no laser light was present at the sample the baseline was collected and later subtracted from the TOF data to eliminate the possible electric interference effects of the surroundings. Additionally, data were averaged with the oscilloscope up to 100 times to improve the signal-to-noise ratio. A schematic diagram of the TOF set-up is presented in FIG. 2.11.
FIG. 2.11. Schematic diagram of the time-of-flight set up; the lens was removed when a collimated beam was used in experiments; the lens was removable from the set-up to provide either collimated or focused beam; all electrical connections were made with standard coaxial cables; the cable leading to the MCP was additionally shielded with a metal sleeve or ferrite rings were mounted to reduce the high frequency noise.

2. 2. 4. 3. DATA CONVERSION FROM TIME TO ENERGY

The original data were acquired in the time domain. Later, they were converted to the energy domain. It was assumed that all electrons start their acceleration process at a metal surface that is perfectly parallel to the MCP entrance grid. The electrons emerge out of the metal layer with zero initial kinetic energy and are accelerated only in the evanescent field and in the external electric field if it is applied. The region of acceleration in the evanescent field is small compared to the electron’s flight path, therefore it is assumed that the electrons travel through TOF tube at a steady velocity.

The number of emitted electrons per unit time $dt$ is $n(t)$. The total number of electrons in one bunch is a sum of all electrons emitted during the acquisition time: [7]

$$N(t) = \int_0^\infty n(t) \, dt. \quad (2.1)$$

Similarly in the energy domain, the number of electrons whose energy falls within an energy unit $dE$ is $n(E)$. Therefore, the total number of electrons $N(t)$
expressed by equation (2.1) is also a sum of all electrons with energy defined within $dE$ interval over energies form zero to infinity:

$$N(E) = \int_{0}^{\infty} n(E)dE.$$  \hspace{1cm} (2.2)

Consider an electron with an initial velocity $v_0$ due to acceleration in the evanescent electric field. The equation of motion of this electron is:

$$z = v_0 t,$$  \hspace{1cm} (2.3)

where $v_0 = L/t$ is the initial velocity that is acquired from the interaction with the optical field and $L$ is the flight path:

$$E = \frac{1}{2} m_0 \left( \frac{L}{t} \right)^2.$$  \hspace{1cm} (2.4)

From the above equation the Jacobian can be found. Now substituting the Jacobian into equation (2.1) one finds:

$$N = \frac{\sqrt{2m_0}}{4} L \int_{0}^{\infty} \frac{1}{\sqrt{E^3}} n(t) dE.$$  \hspace{1cm} (2.5)

During experiments, every emission spectrum that was obtained in the time domain as seen on the oscilloscope was recalculated in terms of energy using equations (2.4) and (2.5). This procedure resulted in a non-uniform data point distribution. Therefore, data were always averaged using the binning method into bins of equal energy. This resulted in selective averaging and led to less densely spread data points. The typical size of one bin was 0.1 eV.

**2. 2. 4. 4. RESOLUTION OF TOF SPECTROMETER**

The resolution of the TOF spectrometer in the electron detection mode is determined by the duration of the time of flight, the divergence of the electron beam caused by the space charge effects and the temporal resolution of the detecting device. The relative energy resolution was found as a partial differentiation of equation (2.4): [8]
\[ \frac{\Delta E}{E} = 2 \frac{\Delta t}{t} + 2 \frac{\Delta L}{L}, \]  

where \( \Delta t \) is the precision at which the signal is measured and \( \Delta L \) the precision at which the electron path is measured.

The temporal resolution of the experiment is determined by whichever was greater the MCP resolution or the oscilloscope resolution. As quoted above the MCP resolution is \( \Delta t_{\text{MCP}} = 690\,ps \), oscilloscope resolution is \( \Delta t_{\text{osc}} = 1\,ns \). Therefore the resolution of the oscilloscope determines the time resolution of the TOF experiment. The resolution due to the length dispersion is explained in FIG. 2.12. \( \Delta L \) for the 20 cm TOF tube is 20 nm. However, the TOF tube was measured with a precision of 0.5 mm. Therefore the second value is used in the estimation of the precision of experiments.

![Diagram of electron interaction area](image)

\[ \Delta L = L_i - L = \sqrt{L^2 + 0.25\phi^2} - L. \]

FIG. 2.12. The path difference of the electron that path of flight coincidences with the main axes of the TOF spectrometer \( L \) and the one that reaches the edge of the detection area of MCP \( L_i \), \( \phi \) is the diameter of the MCP active area.

The accuracy of the measurement in the TOF technique depends on the energy of the measured electrons. Assume 20 cm TOF tube the accuracy at 1 eV is 4 meV but at 400 eV it is only 17.6 eV. However, when the 55.5 cm TOF tube was used the electron energy was resolved with precision of 6.4 eV at 400 eV.

2. 3. DESCRIPTION OF LASER SYSTEMS USED

Three femtosecond laser systems were used in the experiments described in this thesis. Each laser system was a Ti:sapphire based regenerative amplifier system used in a different configurations. The choice of the laser was dictated by the laser power.
and pulse duration it could deliver and its availability. Many good reviews of a femtosecond laser systems are readily accessible, some of them are [9-11]. In the following only summarises of the parameters of the three laser systems used in the experiments are described in the subsequent Chapters.

Table 2.1 presents the parameters of the femtosecond laser systems. In the first stage of an experiment a commercial model from Coherent was used. This system produced circa 150 fs pulses. The highest repetition rate at which the system could operate was 250 kHz, however these pulses were the longest and the lowest energy of all the possible settings. A 25W CW argon ion laser was used to pump the oscillator (10W) and regenerative amplifier (15W). The regenerative amplifier was of a multipass (25 to 30 roundtrips) design and did not require a stretcher. A grating and a mirror combination was used to compress the pulses to 150 fs.

<table>
<thead>
<tr>
<th>Laser:</th>
<th>Mira/Reg A</th>
<th>TOPS BNF</th>
<th>TOPS Alfa</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pump laser</td>
<td>CW 25W Ar ion</td>
<td>CW 5W Nd:YVO₄</td>
<td>CW 5W Nd:YVO₄</td>
</tr>
<tr>
<td></td>
<td>10W to oscillator</td>
<td>12W Q-switched YLF</td>
<td>12W Q-switched YLF</td>
</tr>
<tr>
<td></td>
<td>15W to amplifier</td>
<td></td>
<td>20W Q-switched YLF</td>
</tr>
<tr>
<td>Oscillator</td>
<td>100 fs</td>
<td>85 fs</td>
<td>20 fs</td>
</tr>
<tr>
<td>1st stage</td>
<td>150 fs</td>
<td>100 fs</td>
<td>40 fs</td>
</tr>
<tr>
<td>amplifier</td>
<td>8 µJ</td>
<td>800 µJ</td>
<td>450 µJ</td>
</tr>
<tr>
<td></td>
<td>250 kHz</td>
<td>1 kHz</td>
<td>1 kHz</td>
</tr>
<tr>
<td></td>
<td>800 nm</td>
<td>785 nm</td>
<td>800 nm</td>
</tr>
<tr>
<td>2nd stage</td>
<td>none</td>
<td>none</td>
<td>40 fs</td>
</tr>
<tr>
<td>amplifier</td>
<td>none</td>
<td>none</td>
<td>3 mJ</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>1 kHz</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>810 nm</td>
</tr>
</tbody>
</table>

**Table 2.1.** The comparison of parameters of laser systems used in the experiments described in this work; TOPS is the Terahertz to Optical Pulse Source laser facility at Strathclyde University; the first generation TOPS laser was obtained from British Nuclear Fuels (BNF) and later custom developed to the Alfa laser facility.
The second system was a 1 kHz tabletop laser originally built for British Nuclear Fuels (BNF) by BMI. It comprised a Coherent Mira oscillator pumped by a 5W diode pump Nd:YVO$_4$ laser called Verdi. The Verdi provided better beam stability but lower power. However, the beam was afterward amplified in the regenerative amplifier to 800 mJ, 100 fs duration pulse. The amplifier was pumped by a Q-switched a 12W YLF laser. Better stability of the oscillator resulted in better signal-to-noise ratio of the output beam.

Lastly, the TOPS (Terahertz to Optical Pulsed Source) Alfa laser was the most sophisticated of all the three systems. The oscillator was again Verdi pumped and used chirped mirrors to allow compact design and high stability for periods longer than 12 hours. The oscillator produced 20 fs pulses and was used to seed 10 Hz and 1 kHz amplifier systems. However, only the 1 kHz system was used in this work. After the oscillator pulses were stretched to 200 fs. Each seed pulse was picked up by a Pockels cell before the next one arrived and was trapped in the cavity and amplified. In the first stage amplification 12W YLF Q-switched laser was used to pump a Ti:sapphire crystal and the pulse was amplified to 450 mJ. Afterwards, the pulse was sent to the second stage amplifier where it travelled twice through the Ti:sapphire crystal pumped by the 20W Q-switched YLF laser. There the pulse was amplified further to 3 J in a double pass amplifier. A grating pair (10x15 cm) compressor was used to compress the pulse to 40 fs. It is important to point out that the TOPS Alfa system was used during its initial operation period and typical starting-up problems were experienced. During most of the experiments only a first stage amplification was used and at all times the output beam was astigmatic.

In this chapter all the experimental techniques have been described in detail and the measurements methods listed. The operation basics of the equipment used in the experiments is presented and the experimental details reviewed. In the following chapters data regarding specific experiments will be discussed and reference will be included where required to this chapter.
2.4. REFERENCES


CHAPTER 3

OVERVIEW OF SURFACE PLASMON THEORY

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CHAPTER 3

OVERVIEW OF SURFACE PLASMON THEORY

In this chapter a review of the surface plasmon theory will be presented. The surface plasmon wave equation will be introduced and the concept of an evanescent optical field discussed. The SP dispersion relations in a two-boundary system will be presented and analysed using Drude theory. The possible experimental configurations that allow surface plasmon excitation will be introduced and compared. The emphasis will be put on the electric field enhancement factor in the presence of SPs at the metal surface.

3.1. SURFACE PLASMONS IN THREE LAYER SYSTEMS

Surface plasmons (SPs) are the mixture of electromagnetic (EM) waves and electrical charge oscillations at the metal interface. The excitation of SPs involves the coupling of electromagnetic (EM) waves to a surface electric dipole. SPs have been the subject of many reviews. [1-5]. Here only a short summary of the generation process of SPs and their properties will be given as an introduction to discussion and experiments presented later.

Consider a plane, monochromatic EM wave impinging at the arbitrary angle $\theta$ on a metal film of a thickness $d$, that is sandwiched between two different semi-infinite dielectric media (see FIG. 3.1). Following Drude theory [6], each medium can be characterised with the dielectric function $\varepsilon_n(\omega)$, $\quad n=1,2,3$ that depends on the frequency of the driving EM field $\omega$. As Fresnel formulated, at each interface the EM wave will be partially reflected and partially transmitted. This leads to the combination of partial waves in a system, as indicated in FIG. 3.1. All waves in the system are a sum of individual partial $(i)$ waves propagating in the given direction $j$ in each medium $n$: 

26
\[
\vec{E}(r,t) = \sum_j \vec{E}_{0,j}(k_{n,j}, \omega) \exp[ i k_{n,j} \cdot r ] \exp[-i \omega t],
\]

(3.1)

where \( \vec{E}_{0,j}(k_{n,j}, \omega) \) is the EM wave amplitude that is dependent on the direction of propagation \( j \) and the frequency of the driving field \( \omega \) in medium \( n \). When two interfaces are perfectly parallel then there are only two or less directions of propagation in each medium that depend on the incidence angle \( \theta \) as shown in FIG. 3.1. Each Cartesian component of the wave vector of the EM wave can be evaluated at the boundary. This leads to: [3, 4]

\[
k_{n,x} = k_{n+1,x}, \quad k_{n,y} = k_{n+1,y}, \quad n = 1, 2, 3,
\]

(3.2)

\[
k_{n,x}^2 + k_{n,y}^2 + k_{n,z}^2 = \frac{c^2}{\omega^2} \epsilon_n(\omega),
\]

(3.3)

where the \( k_{n,x}, k_{n,z} \) are the Cartesian components of the wavevector \( k \).

FIG. 3.1. The schematic diagram showing a plane, monochromatic wave impinging at the double-interface system at the incidence angle \( \theta \), the two outermost media are semi-infinite whereas the thickness of the middle one is \( d \); each medium is non-magnetic and characterised with the frequency dependant dielectric function \( \epsilon_n(\omega) \), \( n = 1, 2, 3 \); the amplitudes of the total incoming, reflected, transmitted waves within the thin film waves are denoted as \( E_{in} \), \( E_R \), \( E_T \), \( E_A \), \( E_B \) respectively; the Cartesian coordinates used throughout the calculations are indicated.
A SP wave can only be excited in a system where a non-zero component of the EM wave exists that is perpendicular \( (E_z) \) to the interface \( (x) \) at which the EM wave is incident. This can be realised when the transverse magnetic (TM) EM is incident at this interface. The electric field vector of the TM wave is in the plane perpendicular to the interfaces of the system. The amplitude of the electric field vector of the TM EM wave is:

\[
\overline{E}_{0,i}(r) = (E_{x,i}, 0, E_{z,i})
\]

and its wavevector is:

\[
\overline{k}_i(r) = (k_{x,i}, 0, k_{z,i})
\]

At each interface the electric and magnetic field vectors of the EM wave are continuous and therefore need to fulfil the boundary conditions [5]. Consider the interface at the point \( z = b \). The close proximity to the boundary at its left side can be noted as \(-b\) and the close proximity at its right side can be expressed as \(+b\). Following this formalism, each spatial coordinate of each component of the electromagnetic field, denoted in general as \( \Psi(r) \), needs to fulfil one of the following equations. Note that time dependent factor \( \exp[-i\omega t] \) can be neglected as it does not change in all three media. The component parallel to the surface:

\[
\Psi_{x,-b}(b) = \Psi_{x,+b}(b) \\
\Psi'_{x,-b}(b) = \Psi'_{x,+b}(b)
\]

and the component perpendicular to the surface:

\[
\cos(\theta_b)\Psi_{z,-b}(b) = \cos(\theta_b)\Psi_{z,+b}(b) \\
\cos(\theta_b)\Psi'_{z,-b}(b) = \cos(\theta_b)\Psi'_{z,+b}(b)
\]

where \( \Psi_n \) is evaluated at a boundary, where \( n = x, z \), and defined in FIG. 3.1, \( \theta_b, \theta_{+b} \) are the angles of incidence and refraction at the boundary respectively.

The solution of the above equations in a double interface system defines all electromagnetic modes that are possible to excite in the system. The existence of
particular modes and their exact localisation will depend on the value of the dielectric functions of media considered and the angle of incidence $\theta$ of the EM wave. All surface modes that are possible to excite in the double interface system are discussed in detail elsewhere [5], here the focus is on the SP modes only.

3.2. SPS AS AN EM WAVE, DISPERSION RELATIONS

A SP is a freely propagating EM wave along the surface of a metal. The maximum amplitude of a SP wave is at the surface and it decays exponentially away from it, therefore it is called an evanescent wave.

![Evanescent wave diagram](image)

**FIG. 3.2.** The perspective view of the calculated real part of the electric field vector as a function of position in a glass/metal/vacuum system; the EM wave is impinging at the first interface at the SP resonance angle $\theta_{SP}$, SPs are excited in the intermediate metal layer, this results in a strong evanescent field at the metal/vacuum interface.

The SP’s electric field can be defined as:

$$
\vec{E}_{SP}(\vec{k},t) = \begin{cases} 
E_0 \exp[i(k_z x - \omega t)] \exp[k_z z], & z \geq 0 \\
E_0 \exp[i(k_z x - \omega t)] \exp[-k_z z], & z < 0
\end{cases}
$$

(3.8)
with \( k_x, k_z \) - the SPs wavevectors in the x and z direction respectively. The \( k_z \) is always purely imaginary causing the exponential decay of the \( E_z \) component of the \( E_{SP} \) vector.

One of the solutions of the boundary conditions (equations (3.6) and (3.7)) are the dispersion relations. The dispersion relations define the dependence of the SPs angular frequency \( \omega \) on the their wavevector in the plane of the metal film \( k_x \). The component of the wavevector in the plane of the film \( k_x \) can be calculated from the relation: [4, 5]

\[
k_{SP,x}^2 = \left( \frac{\omega}{c} \right)^2 \frac{\varepsilon_x \varepsilon_{n+1}}{\varepsilon_n + \varepsilon_{n+1}}, \quad n = 1, 2, 3, \tag{3.9}
\]

where index \( n \) denotes the appropriate medium where the wavevector is evaluated. The component perpendicular to the interface can be calculated using equations (3.3) and (3.9). At the SP resonance angle \( \theta_{SP} \) \( k_z \) is a pure imaginary number. The solution to equation (3.9) depends on the exact values of the dielectric functions of the media used and in general is complex. The detailed analysis of all the possible combinations of media is discussed elsewhere. [5] Here only a graphical interpretation will be presented.

FIG. 3.3. The schematic diagram of the dispersion relations in the double interface system: glass/metal/vacuum; A is a dispersion relation of light in vacuum; B is a dispersion relation of light in metal film; C is the SPs dispersion relation; the guidelines show the only allowed value of the wavevector \( k_x \) at which efficient coupling can occur of the SPs modes to the EM wave in a metal film.
FIG. 3.3 presents the dispersion relations in a double interface system where a metal film, characterised by the dielectric function \( \varepsilon_2 = \text{Re}(\varepsilon_2) + i \text{Im}(\varepsilon_2) \), is sandwiched between two different dielectrics. The dielectric function of the two outer media are \( \varepsilon_1 = \text{Re}(\varepsilon_1) \) and \( \varepsilon_3 = \text{Re}(\varepsilon_3) \) respectively. Medium 3 was chosen to be vacuum \( \varepsilon_3 = 1 \).

Line A in FIG. 3.3 is the dispersion relation of light propagating freely in a vacuum \( \omega_{\text{light}} = c k_x \sin(\theta) \). All EM modes for which \( \omega > \omega_{\text{light}} \) are radiative, e.g. both components of the wave vector \( k_x \) and \( k_z \) are real quantities. Line B represents the dispersion relation of light propagating in the metal film \( \omega_{\text{metal}} = \frac{c}{\sqrt{\varepsilon_{\text{metal}}}} k_x \sin(\theta) \).

Curve C is the SPs dispersion relation. Since \( \omega_{\text{SP}} < \omega_{\text{light}} \) therefore SPs are the non-radiative modes. The SPs modes can couple efficiently to the EM wave in the metal film whenever \( k_{z,\text{SP}} = k_{z,\text{metal}} \). In the system analysed in FIG. 3.3 there exists only one value of the SPs wavevector that allows the efficient coupling (pointed out with the guidelines). Therefore, following equation (3.3) there exists only one unique angle of the incident EM wave and the resonance angle \( \theta \), at which the SP modes can be excited. It is the attenuated total internal reflection angle.

### 3.3. ATTENUATED TOTAL INTERNAL REFLECTION

There are two commonly used experimental configurations where the SPs modes are excited: the Otto [1] and Kretschmann configurations [4] (see FIG. 3.4). Both of the configurations employ the attenuated total internal reflection (ATIR) method to efficiently couple the EM wave to SP at the metal surface.

The Kretschmann configuration is more favourable experimentally as it allows for the convenient detection of the SPs generated photoemission. A thin metal layer laid down on a prism face as described in Chapter 2. Since the SPs wave is localised at the metal/vacuum interface (see FIG. 3.4, part A) the electrons are emitted out of the metal film toward vacuum. The electron detection device can be conveniently placed at a distance in front of the metal film. On the contrary, the Otto configuration involves an arrangement where a flat metal surface is placed at close proximity to the glass prism (see FIG. 3.4 part B). In this arrangement a clean, well defined metal surface can be prepared prior to experiment. The technique of cleaning the metal surface was well
established in classic photoemission experiments. However in the Otto configuration the SPs excitation occurs inside the arrangement in a small air gap between the prism and the metal block. Therefore the observation of the possible electron emission is difficult.

**FIG. 3.4.** Methods of exciting SPs: A the Kretschmann configuration, the thin metal layer is deposited onto the hypotenuse side of the right angle glass prism; B the Otto configuration, the thin air gap is created between right angle glass prism and a bulk slab of metal; the interface at which the SPs are excited is indicated in the appropriate insets; in both cases the most efficient coupling between photons and electrons occurs when the EM wave is incident at the metal layer at the resonance angle \( \theta \); the amplitude of the electric field vector decays exponentially away from the surface at which the SPs are localised.

The TM polarised EM wave that is incident at the metal surface, at an angle larger then the critical one, is expected to be totally internally reflected. However, in the Kretschmann or Otto configurations there exists a certain resonance \( \theta \) where the EM wave is attenuated. At the SPs resonance angle, the EM wave wavevector can couple efficiently to the SPs one in metal layer (see FIG. 3.3). The dependence of the reflection coefficient on the angle of incidence is a function of the dielectric functions of the 3 media used, the thickness \( d \) of the metal layer and the incident wavelength.

If a TM polarised EM wave is defined by equation (3.1) then the reflection and transition coefficients of the \( z \) component of the \( E \) vector can be calculated by inserting equation (3.1) for each individual partial wave into the boundary conditions (equations (3.6) and (3.7)) for the double interface system with the appropriate, media
dependent amplitudes (as defined in FIG. 3.1). Then the reflection coefficient of the \( z \) component of the EM wave is defined as: [6]

\[
R = \left| \frac{E_{R,z}}{E_{in,z}} \right|^2,
\]

and the transmission coefficient is defined as: [6]

\[
T = \left| \frac{E_{T,z}}{E_{in,z}} \right|^2,
\]

where the amplitudes of the EM waves are defined in equation (3.4).

The variation in the reflection coefficient with angle of incidence \( \theta \) in the Kretschmann geometry is presented in FIG. 3.5 for a range of silver film thickness \( d \) (\( d = 10 - 70 \) nm).

**FIG. 3.5.** Calculated reflection coefficient as a function of the incidence angle of the EM wave for the Kretschmann configuration; different curves correspond to different thickness of the silver film as indicated, \( \theta_{crit} \) denotes the critical angle for the configuration; the values of the dielectric functions of the media used at 800 nm were taken from [7].
The reflection coefficient at the SP resonance angle is sensitive to the thickness of the metal layer as well as the exact value of the dielectric functions of all three media at the frequency of the driving optical field. Therefore, the coupling efficiency depends on the choice of the appropriate materials in a given experimental configuration. The most commonly used materials are silica (prism) and either silver or gold metal films. These arrangements combine cost effectiveness and good coupling efficiency, since in this configuration the SP resonance angle is close to 45° and the right angle prism provides nearly normal incidence of the laser beam at one of the sides adjacent to hypotenuse of the prism (see FIG. 3.4). In this way the loss of incident power due to reflections at this interface is minimised.

3.4. ELECTRIC FIELD ENHANCEMENT

One of the most significant properties of SPs is the electric field enhancement at the active interface. The electric field associated with the SPs has high density at the interface. This leads to the relative enhancement of the SPs field at the active interface. [3, 8] The maximum of the enhancement coincidences with the SPs resonance angle and can be expressed as (see FIG. 3.1):

$$\gamma = \frac{E_{r,z}}{E_{m,z}}.$$  \hfill (3.12)

It can be calculated solving the boundary conditions (equation (3.7)) for all partial waves in the double interface system.

FIG. 3.6 shows the reflection coefficient and the field enhancement factor as a function of the angle of incidence of the EM wave in the Kretschmann configuration. The maximum value of the field enhancement function and the minimum value of the reflection coefficient in the same arrangement occur at slightly different angles. This is an indication that the field is enhanced due to SPs generation at the metal surface. The difference between the two resonance angles depends on the imaginary part of the metal dielectric function and the smaller $\text{Im}\left[\varepsilon_{\text{metal}}\right]$ the smaller the difference between the two angles. This discrepancy can be attributed to the fact that the phase of the polarisation phase of the induced electric dipoles in metal lags behind the phase of the driving EM field. [5, 8-10] Moreover, for a perfect conductor the enhancement is expected to be
infinitely large since the induced charge oscillations would not be damped. [5] In real systems it is always a finite value.

FIG. 3.6. The calculated reflection (dashed line) and field enhancement (solid line) coefficients with silver film as functions of the incidence angle; where $\lambda = 800$ nm in the Kretschmann configuration; the maximum of the field enhancement and the minimum of the reflection coefficient do not coincidence with each other.

The magnitude of the field enhancement function depends also on the ratio of values of the dielectric functions of the two outer media in the Kretschmann geometry. It can be calculated from equation (3.7), which shows that the higher the $\frac{\varepsilon_1}{\varepsilon_3}$ ratio is the higher field enhancement can be obtained. The value of $\varepsilon_3$ remains fixed as it is always chosen to be vacuum. However, $\gamma$ can be further maximized by choosing an appropriately large value of the dielectric function of the first medium. In each individual arrangement the SP resonance angle has a unique value.

The electric field enhancement at the appropriate $\theta_{SP}$ as a function of the position perpendicular to the surface was calculated for several different arrangements of the three media in the Kretschmann geometry and is shown in FIG. 3.7. The zinc sulphide dielectric function is 2.314 at 800 nm compare to 1.51 for silica glass. [7] This leads to about 1.5 times higher enhancement coefficient at the appropriate resonance angle.
However, from the economical point of view the modification of the Kretschmann configuration is not favourable. The silica glass right angle prism is a standard optical component that can be purchased at low cost and short delivery time from a standard optics supplier. On the other hand ZnS is a more specialised material, therefore it is not easily accessible and is delivered by specialised suppliers at high cost. Therefore, without compromising much of the performance of the experiment the silica glass prism was used in the experiments presented in this work.

![Graph showing the electric field vector as a function of position for different prism-material/metal arrangements.](image)

**FIG. 3.7.** A calculations of the absolute value of the electric field vector \( (E_z) \) in the direction normal to the metal surface as a function of the position; the curves represent different prism-material/metal arrangements (as indicated) at the appropriate SPs resonance angle \( \theta_{SP} \); all calculations were performed for an exciting wavelength of 800 nm.

Further enhancement of the evanescent electrical field can be achieved by a change in the morphology of the metal film. It is well known that in the proximity of sharp objects the electric field is greatly enhanced. This way of enhancing the electric field was used in the emission cathodes of the electron microscopes. [11] Moreover, a significant optical field enhancement produced at rough metal surfaces is the basis of Surface enhanced Raman Spectroscopy (SERS) [12-14]. The creation of a rough metal surface or an array of a sharp metal tips (~50 nm) would therefore be expected to lead to
the significant enhancement of the surface plasmons and to the efficient production of photoelectrons. This subject will be discussed in more detail in Chapter 8.

In this Chapter the basic theory of SPs was introduced. The wave equation of the SPs waves was introduced. The SPs dispersion relation that describes the dependance of the angular frequency $\omega_{SP}$ on the SP wavevector $k_{SP}$ was discussed. The experimental configurations were proposed that provide an efficient coupling of the SPs to the free space EM wave. The enhancement of the electric field due to SPs generation at the metal surface was discussed, that can lead to electron emission from the metal surface. In the next Chapter the electron dynamics in the metal film in the presence of the ultrashort laser pulse irradiation will be presented. In this perspective the first experiments that lead to the conclusions about electron emission in the presence SP enhanced field will be analysed.

3.5. REFERENCES


CHAPTER 4

WHY NOT A SIMPLE SURFACE PLASMON-ASSISTED MULTIPHOTON PROCESS?

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CHAPTER 4

WHY NOT A SIMPLE SURFACE PLASMON-ASSISTED MULTIPHOTON PROCESS?

This chapter presents the experiments which enabled the observation of efficient electron acceleration in the evanescent field created by an intense laser pulse. First, it will be shown that the photoemission of electrons occurs in the presence of the surface plasmon enhanced electric field. Next, a short discussion of some aspects of the multiphoton electron emission in the presence of the SPs will be presented. The results of the experiments are compared to previous studies of photoelectron emission. Finally, the thermalisation processes of excited carriers in the metal films will be outlined to explain the unexpected directionality of the electron beam.

4.1. DEPENDENCE ON THE ELECTRON EMISSION ON THE INCIDENCE ANGLE OF THE LASER BEAM

As was pointed out in the previous chapter, there is a unique angle at which the light wavevector can couple with that of the surface plasmons (SPs) in the Kretschmann configuration. Hence, in the same way the electron emission, if due to the SP excitation, should show a similar dependence in the incident laser angle. FIG. 4.1 shows the theoretically calculated optical field enhancement due to SP generation and measured normalised photocurrent as a function of the laser beam incidence angle for transverse magnetic (TM) polarisation. No electron emission was detected for a transverse electric (TE) polarised laser beam of the same intensity at any angle of incidence.
FIG. 4.1. The calculated electric field enhancement (lines) in the ideal Kretschmann configuration for silver (dotted line) and gold (solid line) metal films and the total photocurrent emission as a function of the incidence angle for silver (triangles) and gold (circles) layers in the same configuration. The thickness of the metal films was about 50nm. All measurements were taken in air with a collimated beam of 0.4 GW/cm$^2$ intensity. For ease of presentation both datasets were normalised; the measured peak current was about 0.7 nA and the noise level was in the order of 1 pA.

The resonance angle of the maximum field enhancement strongly depends on the complex dielectric function $\varepsilon(\omega)$ of the metal for a given wavelength. However, $\varepsilon(\omega)$ depends not only on the kind of material but is determined by its structure too. Therefore, it is expected to be different for a crystalline bulk material and a polycrystalline thin film. [1] During numerical calculations, bulk values of the dielectric functions at 800 nm for gold and silver were taken. [2] Additionally, the error of the film thickness monitor used in the evaporation process was about 10% at the deposition rate of 0.2 nm/s. This explains the slightly different maximum angles and the narrower peak of the calculated field enhancement function compared to the electron emission intensity data.
4. 2. EMISSION INTENSITY

To establish the basic parameters of the photoelectron beam, a lock-in detection system was used to measure the average total photocurrent. The experimental set up is shown in FIG. 2.5. The electrode and the prism were placed inside the vacuum chamber. The laser beam at maximum power entered the vacuum chamber through a standard vacuum window and the metal coated side of the prism at the SP resonance angle. A copper plate was placed at a close distance to the prism facing its metal-coated side and connected to the lock-in detector in order to measure the average photocurrent. The Cu disk allowed the detection of electrons emitted in a 46.2° angle. The average electron current, the number of electrons per bunch and the emission efficiency is presented in Table 4.1 The Mira/Reg A laser system (see Table 2.1 in Chapter 2) was used to obtain the data below.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Energy per laser pulse</td>
<td>1 µJ</td>
</tr>
<tr>
<td>Laser pulse width</td>
<td>150 fs</td>
</tr>
<tr>
<td>Photon energy</td>
<td>2.4 x 10^{-19} J</td>
</tr>
<tr>
<td>Repetition rate</td>
<td>250 kHz</td>
</tr>
<tr>
<td>Number of photon per pulse $n_{ph}$</td>
<td>4.03 x 10^{12}</td>
</tr>
<tr>
<td>Average photoelectron current I</td>
<td>1 nA</td>
</tr>
<tr>
<td>Number of electron per pulse $n_e$</td>
<td>24969</td>
</tr>
<tr>
<td>Electric charge per pulse</td>
<td>4 x 10^{-15} C</td>
</tr>
<tr>
<td>Emission efficiency ($n_e/n_{ph}$)</td>
<td>6.02 x 10^{-9}</td>
</tr>
</tbody>
</table>

Table 4.1. Calculated parameters if the laser and photoelectron beam

4. 3. MULTIPHOTON EMISSION

The SP-assisted electron emission has been observed by a number of groups and has been attributed to the multiphoton (MP) photoelectric effect. [3-5]. Following the Fowler-DuBridge theory [6, 7], later extended by Betchel [8], it is possible to show that the MP emission is a superposition of all electrons emitted in each order of the MP process:
Each partial current $J_n$ can be defined as:

$$J_n = a_n A \left[ \frac{e}{h \nu} (1 - R) \right]^n T^2 F(X_n) I^n \equiv \sigma_n I^n$$

with the Fowler function $F(X_n)$ defined as:

$$F(X_n) = \begin{cases} 
\sum_{n=1}^{\infty} (-1)^{n+1} \frac{e^{X_n}}{n^2}, & X_n < 0 \\
\frac{\pi^2}{6} + \frac{X_n^2}{2} - \sum_{n=1}^{\infty} (-1)^{n+1} \frac{e^{-nX_n}}{n^2}, & X_n \geq 0 
\end{cases}$$

with:

$$X_n = \frac{nh \nu - \phi}{k_B T}$$

where $I^n$ is the EM wave’s incident intensity with the order of the MP process denoted as $n$ and $\sigma_n$ its cross section coefficient. $a_n$ is the experimentally determined $n$-photon ionisation constant, $A = 120 (A/cmK)$ is the Richardson constant, $k_B$ the Boltzmann constant, $T$ is the mean temperature of a metal surface, $R$ the reflectivity of the sample and $\phi$ its work function.

If $n=0$ then the equation (4.2) becomes the Richardson expression and defines thermionic emission [9]. The $n$-th partial current $J_n$ simply represents the photocurrent due to the $n$-photon photoemission process. This interpretation is valid only for the terms where the argument $X_n$ is above the photoionisation threshold, i.e., $(nh \nu - \phi) \geq 0$. When this requirement is not fulfilled and for finite temperatures of the emitter, these terms represent the thermally assisted $n$-photon photoemission. Usually this contribution is small unless the temperature of the emitter exceeds several thousand degrees [10].
4.4. POWER DEPENDENCE

The values of work functions for silver and gold were tabulated before and they are 4.5 eV and 5.5 eV respectively. [3, 11] A simple calculation shows that by exciting both materials with a 800 nm laser beam (e.g. 1.55 eV) one would expect a third or fourth order power dependence of the total photocurrent emission (see equation (4.2)). Indeed such a dependence has been reported for the SPs assisted multiphoton emission from silver and gold flat films excited with a beam of a power density less than 100 MW cm$^{-1}$ and wavelength 625 nm (i.e. 1.9 eV) laser beam. [4, 5, 12]

![Graph showing the dependence of the measured photocurrent versus incident peak power for emission from gold film. The data were measured with a DC bias applied between the prism and the detector as it is indicated and with the incident power density ranging from ~5 to ~50 GW/cm$^2$. The lines show fourth, seventh and ninth-order power dependencies respectively.](image)

**FIG. 4.2.** Dependence of the measured photocurrent versus incident peak power for emission from gold film; the data were measured with a DC bias applied between the prism and the detector as it is indicated and with the incident power density ranging from ~5 to ~50 GW/cm$^2$. The lines show fourth, seventh and ninth-order power dependencies respectively.

FIG. 4.2 shows the measured total photoelectron current as a function of the incident laser power for a flat gold film. The sample was prepared using the method described in Chapter 2, Section 2.1.1. The measurement method is described in Chapter 2, Section 2.2.3. The Mira/Reg A laser system was used to obtain the data (see Table 2.1 in Chapter 2). For zero bias voltage between the prism and the collector, the current increases approximately as the fourth power of the incident
laser intensity before saturating at the highest intensities (50 GW/cm$^2$). However, when a reverse bias is applied, the current is reduced and the power dependence is of a higher order.

At least two emission processes are observable here. The first one, manifesting itself with the fast increase of the photocurrent as a function of the incident laser power, could be attributed to the MP process.\cite{4, 12, 13} The second one, manifesting itself in the saturation region at high laser intensities, can be an indication of the tunnelling process\cite{13}. For the purpose of this chapter the emphasis will be put on the first process. The tunnelling process, its presence and importance in the experiments described here, will be explained later.

The change in the power dependence of the photocurrent with DC bias is surprising. 20 V applied across a distance of 2 cm gives an electric field of the order of 10 V/cm. Such a small change in the electric field strength is expected only to pull emitted electrons backwards to the sample. It is too small to cause any changes to the potential barrier at the metal/vacuum interface. Therefore, the only observable effect would be the reduction of the total number of emitted electrons. The increase of the slope of the power dependence is not easy to explain. The Fowler-DuBridge theory does not predict the change of the order of the MP process with variable bias. The order of this process is dependent only on the exciting EM wave intensity. Additionally, the probability of emission decreases with the increasing order of the multiphoton process, which makes the ninth order process at such low laser intensity very surprising (see FIG. 4.2, the intensity dependence with –30V applied DC bias).

As one of the effects of applying of the reverse DC bias is to selectively energy filter electrons therefore a simple spectrometer was constructed. Therefore, it is important to analyse the energy spectra of the photoelectrons. This is done in the following section.

4.5. ENERGY SPECTRA

The distribution of the electrons initial energy does not support the Fowler-DuBridge theory either. Suppose the emission was due to the MP process, then the energy of emitted electrons would need to be well defined and consist of a number of bands at intervals corresponding to the energy of an exciting $\lambda = 800$ nm wave e.g.
1.55 eV. However, the spectrum of the initial energy of the electrons was found to be continuous and broad with the maximum at about 0.5 eV and the energy spread to up to 0.4 keV (see FIG. 4.3). The spectra presented here were excited with the TOPS Alfa system (see Table 2.1).

**FIG. 4.3.** (a): Time-of-Flight spectra as recorded by an oscilloscope, (b): The electron-energy distribution spectra of photoelectrons emitted from a gold layer in the Kretschmann geometry; the inset shows the energy spectra on a linear scale, note no peaks in the energy spectra are present that could suggest the MP process; the laser beam was collimated and its intensity varied from a – d: 0.6, 1.8, 11.1, 26.7 GW/cm² respectively.
The detailed analysis of the spectrum is not within the scope of this Chapter and will be presented in the following chapters.

4. 6. SPATIAL DISTRIBUTION OF THE ELECTRON BEAM

Not only is the energy distribution of the emitted electrons unexpected but also the directionality of the electron beam is not that expected from a multiphoton ionisation mechanism modulated by a SP wave.

Because of the boundary conditions, the SP electric field must be perpendicular to the film and its wavevector must be in the plane of the film. When SPs are formed, they will acquire momentum directed in the plane of the film. It has been observed in the low laser-intensity regime, that SPs have a 13-μm mean free path in the metal film [14]. Thus the electron’s momentum excited out of the metal layer by a MP process, would need to resemble that of the SPs. Thus, one would expect the emitted electron beam to reflect the initial electron velocity originating from the SPs’ momentum. (see FIG. 4.4).

![FIG. 4.4. Schematic drawing of the wavevector directions in the Kretschmann configuration; the laser beam wavevectors are denoted as: $k_{in}$ incoming beam, $k_{out}$, the reflected beam; $k_{SP}$ is the SPs wavevector in the plane of the metal film; $E_{evanescent}$ is the evanescent field vector.](image)

The experiment was performed with the Mira/Reg A laser system (see Table 2.1 Chapter 2). The experimental arrangement was described in Chapter 2, Section 2.2.2. The results are presented in FIG. 4.5. The distribution in the plane of polarisation of the incident light was found to be directional with the majority of photoelectrons
emitted perpendicularly to the metal surface (graph (a) in FIG. 4.5). It fits reasonably well to a $\cos^2 \theta$ function and is independent of the applied bias (within the signal-to-noise ratio). In contrast, the out-of-plane distribution is much more uniform and a preferred direction of photoelectron emission cannot be distinguished (graph (b) in FIG. 4.5).

**FIG. 4.5.** A: In-plane, B: out of plane photoelectron distribution for a set of voltages, 0V (triangles), -5V (bright dots), -10V (dark dots); the Kretschmann arrangement for a silver layer was used and the laser beam was focused and its intensity was set to 50 GW/cm$^2$; the maximum detected photocurrent was 2 nA and the noise level was about 20 pA; on a radial axis the emission intensity is plotted; the angular axis represents the position of the detecting electrode; the solid line shows the fit to a $\cos^2 \theta$ function.

The above results suggest that the analysis of electron dynamics following intense laser pulse excitation of a metal film is essential to understand the emission process described in this work.
4. 7. ELECTRON DYNAMICS IN METALS

There have been many studies of the electron dynamics in metals in the presence of an ultrashort laser pulse. [15-18] The excitation by a short laser pulse (in an order of picosecond or less) produces a non-equilibrium distribution, which leads to a significant temperature rise of the electrons with respect to lattice ions.

A commonly used method to describe the dynamics between electrons and the lattice is the two-temperature model (TTM). If the distribution of electrons in a metal is spatially homogenous (the sample is much thinner then the optical absorption depth) then the electron distribution function can be described as: [19]

$$\frac{\partial n}{\partial t} = \left( \frac{\partial n}{\partial t} \right)_{e-e} + \left( \frac{\partial n}{\partial t} \right)_{e-ph},$$

(4.5)

where e-e denotes electron-electron scattering and e-ph electron-phonon dynamics.

Following the ultrafast laser excitation electrons are rapidly excited to high energies and its internal temperature substantially exceeds the ionic lattice temperature that remains relatively cold. After some time (described by electron-phonon coupling constant), through scattering at the lattice ions and impurities, the energy from the electron cloud is transferred to the ionic lattice and thermal equilibrium is established. The typical electron-phonon coupling times in gold and silver are presented in Table 1.

If the electron emission occurs such that the time scale is shorter than the energy relaxation time then no energy will be transferred from the electron cloud to the ionic lattice. This means that the ionic lattice remains relatively cold during the whole emission process and does not contribute to it. This leads to the conclusion that the thermionic emission process should not be expected in the experiments described in this work (see equation (4.2) for $n = 0$).
The electron dynamics that follows the excitation of the ultrashort laser pulse is more complicated than the excitation with the continuous or a nanosecond laser. The electron relaxation time is a non-linear process and depends on the energy to which the electron is excited. [27] At the first instance only a small number of electrons are excited to the high energies relative to the lattice temperature. Later during the internal thermalisation process (electron-electron scattering rate $\tau_{e-e}$) the access energy is transferred to unperturbed electrons. [22] The tabulated momentum relaxation times $\tau_{e-e}$ are presented in Table 4.2.

One can conclude that in metals the energy relaxation time is of the order of a few hundred femtoseconds and the momentum relaxation time is about 40 fs or faster at higher energies. Therefore when electrons are photoemitted outside of the metal

<table>
<thead>
<tr>
<th>Method used</th>
<th>Momentum relaxation lifetime (fs) $\tau_{e-e}$</th>
<th>Energy relaxation lifetime (fs) $\tau_{e-ph}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Time resolved study</td>
<td>48 [14]</td>
<td>710 [16]</td>
</tr>
<tr>
<td></td>
<td>10 at 4000K [20]</td>
<td>830 [16]</td>
</tr>
<tr>
<td></td>
<td></td>
<td>350 [21]</td>
</tr>
<tr>
<td></td>
<td></td>
<td>800 [23]</td>
</tr>
<tr>
<td></td>
<td></td>
<td>900 [22]</td>
</tr>
<tr>
<td></td>
<td></td>
<td>~1000 [24]</td>
</tr>
<tr>
<td></td>
<td></td>
<td>100 [20]</td>
</tr>
<tr>
<td></td>
<td></td>
<td>500 [21]</td>
</tr>
<tr>
<td>Second harmonic studies</td>
<td>40±7 [25]</td>
<td></td>
</tr>
<tr>
<td>Time resolved study using SEM</td>
<td>40 [26]</td>
<td></td>
</tr>
<tr>
<td>Electron dc conductivity measurements</td>
<td></td>
<td>30 [12]</td>
</tr>
</tbody>
</table>

Table 4.2. Table summarising the momentum and energy relaxation times of silver and gold reported in literature; only room temperature values are presented unless indicated; the measurement techniques used to obtained data and the reference sources are stated.
film, they should emerge in a random direction. However, this is inconsistent with the observed directionality of the electron beam and the energy spectrum of electrons. Therefore there must be force pushing the electrons away from the metal layer.

In this chapter a general characteristic of an electron beam emitted via the SP assisted process was presented. It was shown that the emission is sensitive to the angle of incidence of the exciting laser beam and follows the SPs angular dependence. The unexpected high order dependence of the electron emission on the laser power was presented and anisotropic spatial distribution of emitted electrons was demonstrated. The results were discussed with respect to the Fowler-DuBridge emission process, which was the commonly accepted emission mechanism in the presence of SP enhanced electric field. The conclusions were drawn that the MP theory cannot explain the observed high order electron emission nor the directionality of the electron beam. The results point out that the electrons interaction with the laser light at the metal surface in the presence of the surface plasmons cannot by explained only on the basis of the Fowler-DuBridge theory.

4. 8. REFERENCES


CHAPTER 5

THE CONCEPT OF EVANESCENT WAVE ACCELERATION

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CHAPTER 5

THE CONCEPT OF EVANESCENT WAVE ACCELERATION

In the previous chapter it was seen that electrons emitted from a thin (50 nm) gold film using the Kretschmann configuration was excited with 150 fs 800 nm laser pulses had an energy and angular distribution inconsistent with a simple surface plasmon assisted multiphoton ionisation model. In this chapter a mechanism involving electron acceleration with a ponderomotive force will be postulated. Electro acceleration techniques, by which high energy electrons beams are generated, are reviewed. The interaction of an electron with a high gradient optical field will be analysed. These considerations are then extended to investigate the possibility of electron acceleration in an evanescent field.

5.1. INTERACTION OF ELECTRONS WITH STRONG OPTICAL FIELDS

There is ever-growing interest to develop more efficient (with different spectral properties) particle accelerator schemes, to study various aspects of fundamental particle interactions with matter. The various electron accelerators offer short wavelength, high energies and a small particle mass. The interaction of an electromagnetic (EM) field with free electrons offers the possibility of creating high gradient accelerators of small bunches of electrons.

The concept of an electron interaction with the EM wave was initially postulated in 1933 by Kapitza and Dirac.[1] This early work discussed the reflection of a propagating electron beam from the standing EM wave within the framework of classical mechanics. However, it was over 30 years later when this interaction was understood in more detail. [2-4] The main reason for such a delay was the lack of strong, coherent EM fields. Only the development of high intensity pulsed desktop lasers made this effect strong enough to observe it in an appropriate experiment.
A large portion of laser driven electron acceleration research has been focused on plasma based schemes, such as a plasma beat-wave accelerator (PBWA), a laser wakefield accelerator (LWFA) and their various derivatives.

**FIG. 5.1.** Schematic of the plasma beat-wave acceleration of electrons.

A plasma beat-wave accelerator was first described in 1979 by Tajima and Dawson. [5] It is based on a coherent excitation of plasma charge oscillations by two laser beams. The operation principle of PBWA is presented in FIG. 5.1. Two long laser pulses of frequencies $\omega_1$ and $\omega_2$ are sent through the plasma. Subsequently the plasma is modulated by the laser beat frequency $\Delta \omega = |\omega_1 - \omega_2|$. The plasma ions and electrons follow the dynamics of the beat wave. They start transverse oscillations that are the result of the interaction with the laser beat wave and the space charge effects. The oscillations establish the plasma wakefield. The plasma wakefield can propagate at the velocity close to the speed of light and can trap plasma electrons. The trapped electrons pick up the momentum from the plasma wave and can be forward accelerated.

**FIG. 5.2.** Schematic of laser wakefield acceleration
In the laser wakefield acceleration only a single, short high intensity laser pulse interacts with the plasma.\cite{5, 6} In this experiment presented in FIG. 5.2 schematically, a single, high intensity ($10^9$ GW/cm$^2$), short (1 ps) laser pulse modulates the plasma. When the laser pulse enters the underdense plasma ($\lambda_\text{laser} / \lambda_\text{p} \ll 1$) the force called the ponderomotive force that arises from the variable intensity across the laser pulse length expels electrons away from the centre of the laser pulse. The space charge builds up and pushes electrons away from the high density regions. This establishes the longitudinal wave in the plasma called the wakefield, which the electric field vector points in the propagation direction. The wakefield is driven most efficiently when the laser pulse length $L = c \tau_L$ is approximately the plasma wavelength $\lambda_\text{p} = 2\pi c / \omega_\text{p}$, where $\omega_\text{p}$ is the plasma frequency.\cite{7} The electrons that stay in-phase with the longitudinal plasma field can “ride” it and be accelerated to relativistic energies.

There are various experimental schemes devised to intensify the plasma wakefield amplitude and accelerate electrons more efficiently. There are experiments incorporating one, two or more laser beams that interact with plasma. Multiple laser pulse are often used to maximise the accelerating force. A substantial review on this topic is presented by Esarey and Sprangle in\cite{8}. The typical gradient of the accelerating field obtained in the plasma-based schemes is about 100 GV/m.\cite{9} The main downfall of these methods is the requirement for the high uniformity plasma, laser beams at a set wavelength to match the plasma frequency $\omega_\text{p}$ and more than one intense laser source.

To avoid plasma stability based problems, schemes that accelerate electrons in vacuum were proposed.\cite{10} Since the EM field is a transverse electric wave the direct acceleration of an electron in the direction of the EM wave propagation is not possible. In the scheme proposed by Essarey et al\cite{10} the acceleration is achieved by interaction of two crossed laser beams with a co-axially propagating electron beam. The schematic of this arrangement is shown in FIG. 5.3. Two laser beams of the same frequency cross each other at the angle $2\theta$. The phase of the beams is adjusted such that the transverse electric field vectors add at the axis and the axial vectors cancel. Therefore, the accelerating electric field is a sum of only the transverse
components of the electric field vectors of the two individual beams. Properly phased electrons injected along the axis of the laser beam arrangement can be accelerated by the net axial component of the laser field. The acceleration gradients obtained in this type of experiment is of the order of $10^{10}$ eV/m for laser beam intensities of the order of $10^{12}$ TW/cm$^2$. Other laser acceleration schemes involve electron acceleration in the focus of a laser beam [11, 12] or by creation of a laser vacuum beat wave. [13]

FIG. 5.3. The laser vacuum acceleration scheme with two crossed laser beams; the wavevectors $k_1, k_2$ and the electric field vectors $E_1, E_2$ of the two laser beams are indicated; the accelerating field $E_{acceleration}$ is the sum of transverse components of the electric field vectors of the two beams and chosen to be co-linear with the propagating electron beam.

Both plasma- or vacuum-based laser acceleration schemes require complex experimental arrangements such as the generation of high quality plasmas or the interaction of multiple laser beams. The laser wavelength has to match precisely the plasma wavelength, or the laser and electron beams need to be properly phased and precisely coincident in the interaction region to efficiently accelerate electrons. This leads to complicated experimental schemes that require more then one high intensity laser beam and a relativistic electron source. Contrary to experiments described above, the method to be described here requires much less complicated experimental scheme with one laser beam and a simple cathode.
In the experiments presented in this thesis, one laser beam of moderate intensity (typically $10^9 \text{ Wcm}^{-2}$) interacts with a thin metal film. It was shown in Chapter 3 when the EM field interacts with electrons in the thin metal film, it can form coherent oscillations at the surface, known as surface plasmons (SP) waves. It is possible that electrons could be excited out of the metal layer and experience acceleration in the SPs optical field, which extends out of the metal layer toward the vacuum. It is known that the SP electric field vector is always perpendicular to the metal surface and its amplitude decays exponentially away from it. Therefore the ponderomotive force that arises from the intensity changes across the optical beam could cause the acceleration of electrons in the direction perpendicular to the metal layer. Then, the acceleration gradient in experiments presented in this work would be of the order of 400 V/m. However, it is possible to show that there are simple methods that allow the creation of higher acceleration gradients and the possibility of achieving higher acceleration efficiency.

5.2. OUTLINE OF EVANESCENT WAVE ACCELERATION PROCESS

The evanescent wave acceleration process can be described in a simple way. Consider an electron that emerges out of the metal layer in the presence of an evanescent field. At the first instance it will feel the electric field that will either pull it away from the metal layer or push it back toward the metal depending on the phase of the electric field vector at the time of an electron liberation, see FIG. 5.4. Within half of the optical cycle (1.2 fs at 800 nm) the sign of the electric field vector at the surface will reverse, changing the initial condition of an acceleration process. It is significant that the laser pulse reaches the metal surface with a random phase and that there are approximately 500 optical cycles in a 100 fs laser pulse at 800 nm. In the first instance it will be assumed that electrons are liberated out of the metal layer at any point during the optical cycle. This assumption will be revised later in Chapter 7 when the emission processes are discussed.
FIG. 5.4. At the time of emission from the metal film a liberated electron can be either pushed backward toward the metal film (A) or pushed away form it (B) depending on the phase of the optical field at $t_0$.

When an electron is liberated during the time when the electric field points toward the metal film (case A in FIG. 5.4) it will be pushed back toward to the metal. The electric field value inside the metal film is nearly zero (see FIG. 3.7) therefore the electron cannot be accelerated in the evanescent field. However, if the electron is liberated when the electric field vector points away from the metal surface (case B in FIG. 5.4) it will be pushed away form the film first. After 1.2 fs when the E vector changes its sign the electron will be dragged back toward the surface. However, the electric field amplitude decays exponentially with the distance away from the surface. Therefore after each push forward the electron will experience an effectively smaller electric field than during the previous step. The net result of such a motion will be the forward acceleration away from the origin.
5.3. SURFACE ELECTRIC FIELD

To understand the acceleration mechanism better, approximate calculations will be presented to indicate the magnitude of the physical processes involved in the acceleration.

![Graph showing the dependence of the surface electric field ion on the laser light intensity in the presence of SP.]

**FIG. 5.5.** The dependence of the surface electric field ion on the laser light intensity in the presence of SP.

The absolute value of the electric field at the metal surface $E_0$ is related to the laser beam intensity $I$ and the energy per laser pulse $U_{\text{pulse}}$ [14] by:

$$I = \frac{U_{\text{pulse}}}{V} = \frac{1}{2} \varepsilon_0 \left| E_0 \right|^2 \left[ \frac{J}{m^3} \right].$$  \hspace{1cm} (5.1)

The volume $V$ of the laser pulse is defined as the product of its spatial length and cross-section of the beam, $V = c \tau \cdot \pi \left( \frac{d}{2} \right)^2$, where $\tau$ is the duration of the pulse and $d$ the beam diameter. Therefore, the magnitude of the surface electric field may be given by:
It is possible to calculate that in the Kretschmann geometry the evanescent optical field decays over a characteristic length of about $\lambda_{ev} = 1 \ \mu m$. The calculations are presented in FIG. 3.7. Consider, the typical values of energy per pulse as $U_{pulse} = 1 \ \text{mJ}$, the pulse duration: $\tau = 100 \ \text{fs}$ and beam diameter $d = 0.1 \ \text{mm}$. Thus the surface electric field calculated from equation (5.2) is $|E_0| = 9.8 \cdot 10^9 \ \text{Vm}^{-1}$. However, SPs at the metal/vacuum interface enhance the surface electric field by around a factor 10 (see FIG 3.7). Therefore, one can expect the magnitude of the electric field at the surface to be on the order of $10^{11} \ \text{Vm}^{-1}$. FIG. 5.5 shows the magnitude of the surface electric field at the metal surface when the laser beam of a given intensity is present at that surface (see equation (5.1)).

For now, suppose that the electric field remains on for an infinitely long time and its frequency is zero. In this case the upper limit to the electron’s energy resulting from acceleration in an evanescent field can be calculated as: [7]

$$\Delta U_{kin} = e \int_{0}^{\infty} E(z)dz = eE_{0} \int_{0}^{\infty} dz \exp\left[-\frac{z(t)}{\lambda_{ev}}\right] = eE_{0}\lambda_{ev}.$$ (5.3)

Given this condition, if the electric field strength at the surface were $10^{11} \ \text{Vm}^{-1}$ then the electron would gain 80 keV. This is a rather large energy. As noted previously, the field oscillates with the frequency of $f = 2.36 \times 10^{15} \ \text{Hz}$. Therefore, the electron’s behaviour is complex and in order to analyse it in detail, numerical simulations of its motion in the evanescent field were performed.

5.4. NUMERICAL SIMULATION OF MOTION OF THE ELECTRON

The motion of an electron in an optical evanescent field was first solved numerically using the Verlet algorithm, which is widely used for molecular dynamics simulations. [15] In the Verlet algorithm the particle is treated in a classical way and its position is calculated in a series of steps of width $\Delta t$. The position of the electron...
at time \((t+\Delta t)\) is determined from its position at the previous two time intervals \((t\) and \(t-\Delta t)\) and the force \(f(r,t)\),

\[
 r(t + \Delta t) \approx 2r(t) - r(t - \Delta t) + \frac{f(r,t)}{m} \Delta t^2. \tag{5.4}
\]

The velocity and the kinetic energy can be calculated from the positions and are described by:

\[
v(r,t) \approx \frac{r(t + \Delta t) - r(t - \Delta t)}{2\Delta t}, \tag{5.5}
\]

\[
 U_{\text{kin}}(r,t) = \frac{mv^2(r,t)}{2}. \tag{5.6}
\]

Since the laser frequency is extremely high \((\omega = 2.36 \times 10^{15} \text{ Hz corresponding to a period of } 2.7 \text{ fs})\) the time step must be sufficiently small to sample the position of an electron frequently enough. During all numerical simulations, the time step of \(\Delta t = 1 \text{ as} \) was used. When the time step was decreased further, no significant changes were observed in the simulations. The starting position of the electron was always assumed to be at the origin i.e. \(r_i(t_0 = 0) = 0\). The second step was calculated from the given electron’s initial energy \(U_{\text{kin}}(r_i,t_0)\) as

\[
 r_2(t_0 + \Delta t) = r_1(t_0) + \Delta t \sqrt{\frac{2U_{\text{kin}}(r_i,t_0)}{m_e}}. \tag{5.7}
\]

The initial energy of an electron arises from its momentum inside metal film at the moment of liberation. The length of the simulation was typically 200 to 3000 fs. The computer code for this simulation is presented in Appendix 1.

The force \(f(r, t)\) acting on the electron as it moves through the evanescent optical field was modelled by three main elements:

- the exponential part with the maximum value \(E_0\), the maximum electric field strength at the surface and the decay constant equal to the optical field decay length \(\lambda_{\text{ev}}\),

- the cosine part with \(\omega\) set to be equal that of the driving laser field and a given initial phase \(\varphi_0\).
• the Gaussian pulse shape simulating the laser intensity envelope with set FWHM pulse duration $\tau_p$ and the offset time $\tau_s$ - a time after which the maximum of the pulse arrives at the surface.

FIG. 5.6. The numerical simulation of the position of one electron as a function of time as it is accelerated in the evanescent optical field. The electric field strength at the surface was set up to $E_0 = 10^{11} \, \text{V/m}$, pulse duration of FWHM $\tau_p = 100 \, \text{fs}$; the value of the surface electric field when the acceleration starts was set to $E_{\text{start}} = 10^6 \, \text{V/m}$; the initial energy of an electron was set to zero and the phase $\phi_0 = 270^\circ$; the inset shows an enlargement of the oscillatory motion.

The maximum surface electric field $E_0$, the angular frequency $\omega$ and the duration of the pulse $\tau_p$ were chosen to resemble the best experimental values. The evanescent wave decay length $\lambda_{ev}$ was calculated numerically using the method presented in Chapter 2. The initial phase of the laser pulse was a variable during simulations and freely chosen in a range of a full angle. The offset time $\tau_s$ was the parameter whose value was awkward to estimate. The $\tau_s$ parameter determines the strength of the surface electric field at which the electron emission process starts $E_{\text{start}}$. This was typically set to $10^6 \, \text{V/m}$ and consequently the time at which the maximum of the Gaussian envelope arrives at the surface $\tau_s$ was calculated. The three parameters $\phi_0, E_{\text{start}}, \tau_s$ set the initial conditions of the acceleration process. They can be assessed
from the analysis of the emission process of the electron from a metal film and will be discussed in more detail in Chapter 7.

The calculated electron position as a function of time is shown in FIG. 5.6. As the electron interacts with the optical evanescent field, its motion consists of two parts, namely the net acceleration away from the origin and the fast oscillatory part superimposed onto it. It is noticed that within a time of one optical cycle the net displacement in the oscillatory motion is nearly zero whereas the changes in the progressive displacement are negligible. When the electron leaves the evanescent field region its oscillatory motion vanishes and the electron progresses with a steady velocity. As a result of this interaction, the electrons gain kinetic energy $U_{\text{kin}}$, which can be calculated using equation (5.6).

**FIG. 5.7.** The simulation of the kinetic energy of an electron as it interacts with the evanescent optical field; the initial parameters were the same as for the simulation presented in FIG. 5.6; as the result of an oscillatory motion in an evanescent field, the electron gains 25 eV; the area in the lower left corner of the graph where the there is apparent energy oscillation at twice the frequency of the oscillatory motion arises from the negative value of the electron velocity, e.g. the electrons travels toward the metal film.

The energy of the electron oscillates in time and in the energy varies in the range from 0 eV to more than 200 eV, as shown in FIG. 5.7. When the electron leaves the evanescent field it gains a certain final energy. This energy critically depends on all
parameters of the ponderomotive force such as the maximum amplitude of evanescent field \( E_0 \) and its decay constant \( \lambda_{ev} \), the time which an electron spends in the optical field \( (\tau_p, \tau_s) \), the frequency of oscillations \( \omega \) and the initial phase at the time of generation \( \phi_0 \). To understand more fully the electron’s interaction with the evanescent field, an analytical solution of electron motion within the field must be found.

5.5. ANALYTICAL SOLUTION OF THE EQUATION OF MOTION

5.5.1. INTERACTION WITH A INFINITELY LONG PULSE

Consider an electron moving in an electromagnetic (EM) field, whose amplitude decays exponentially as a function of distance and oscillates quickly as a function of time. Assuming that the electron’s velocity is much less than the speed of light, a classical solution is possible. Let \( z \) be the propagation direction of an electron as indicated in Fig 3.1. The \( z \) component of the electric field vector of the evanescent wave can be expressed as,

\[
E(z,t) = E_0 \cos(\omega t + \varphi_0) \exp\left[-\frac{z(t)}{\lambda_{ev}}\right],
\]

where the \( E_0 \) is the electric field at the interface, \( \lambda_{ev} \) the decay length of the evanescent field and \( \omega \) its angular frequency. \( \varphi_0 \) is the initial phase of the optical field.

The force on the electron exerted by the electric field is defined as \( F(z,t) = eE(z,t) \), where \( e \) is the absolute value of electron charge. The Lorentz equation of motion of an electron with the mass \( m \) in the field \( E(z, t) \) (equation (5.7)) is:

\[
\ddot{z}(t) = \frac{F(z,t)}{m} = \frac{eE_0}{m} \cos(\omega t + \varphi_0) \exp\left[-\frac{z(t)}{\lambda_{ev}}\right].
\]

For simplicity of the derivation, equation (5.8) can be re-written in the from,
\[ \ddot{z}(t) = \alpha \cos(\omega t + \varphi_0) \exp \left[ \frac{-z(t)}{\lambda_{ev}} \right], \quad \alpha = \frac{eE_{\omega}}{m}. \quad (5.9) \]

When the electron travels in the rapidly oscillating field it traverses the evanescent wave region on a smooth path, whilst executing small oscillations of angular frequency \( \omega \) around this path (see FIG. 5.6). Using the Lifshitz-Landau [3] approximation, it is possible to separate these two motions and rewrite them in the form of a sum:

\[ z(t) = Z(t) + \xi(t), \quad (5.10) \]

where \( Z(t) \) is the slow coordinate, which changes only slightly with time and \( \xi(t) \) is the fast coordinate. The mean value of \( \xi(t) \) over period \( 2\pi/\omega \) may be approximated to zero. Substituting equation (5.10) into equation (5.9) gives:

\[ \ddot{Z}(t) + \ddot{\xi}(t) = \alpha \cos(\omega t + \varphi_0) \exp \left[ \frac{-Z(t)}{\lambda_{ev}} \right] \exp \left[ \frac{-\xi(t)}{\lambda_{ev}} \right]. \quad (5.11) \]

Now equation (5.11) can be Taylor expanded upon the exponential containing the fast oscillating coordinate \( \xi(t) \). Only the first two terms will be considered. This simplifies equation (5.11) to:

\[ \ddot{Z}(t) + \ddot{\xi}(t) \approx \alpha \cos(\omega t + \varphi_0) \exp \left[ \frac{-Z(t)}{\lambda_{ev}} \right] \left[ 1 - \frac{\xi(t)}{\lambda_{ev}} \right]. \quad (5.12) \]

Since \( Z(t) \) varies slowly within one oscillation period, its derivative will be close to zero. As noted previously, within that time the displacement due to the oscillatory motion is also approximately zero. In this case, equation (5.12) takes the form:

\[ \ddot{\xi}(t) = \alpha \cos(\omega t + \varphi_0) \exp \left[ \frac{-Z(t)}{\lambda_{ev}} \right]. \quad (5.13) \]

It is now easy to integrate equation (5.13) twice with respect to time whilst keeping \( \dot{Z}(t) \) constant,

\[ \xi(t) = -\frac{\alpha}{\omega^2} \cos(\omega t + \varphi_0) \exp \left[ \frac{-Z(t)}{\lambda_{ev}} \right]. \quad (5.14) \]
Inserting equations (5.13) and (5.14) back into equation (5.12) gives:

\[
\ddot{Z}(t) = \frac{\alpha^2 \cos^2(\omega t + \phi_0)}{\lambda_{ev} \omega^2} \exp\left[\frac{-2Z(t)}{\lambda_{ev}}\right].
\] (5.15)

The laser pulse arrives at the surface with a random phase and the electron spectra observed in experiments were averaged over exposure to several laser pulses. Therefore, it is possible to average equation (5.15) with respect to time taking the mean value of squared cosine as \(\cos^2(\omega t + \phi_0) = 1/2\). The average ponderomotive force is then,

\[
m\ddot{Z}(t) = \frac{e^2 E_0^2}{2\lambda_{ev} \omega^2} \exp\left[\frac{-2Z(t)}{\lambda_{ev}}\right] = \frac{dU_{ev}}{dZ},
\] (5.16)

where:

\[
U_{ev}(Z) = \frac{e^2 E_0^2}{4m \omega^2} \exp\left[\frac{-2Z(t)}{\lambda_{ev}}\right],
\] (5.17)

where the \(\alpha\) symbol was changed back to its original physical values, see equation (5.9).

At this point the first conclusions about the electron’s final kinetic energy can be drawn. If an unrealistic assumption were made that the laser pulse is of a square shape and infinitely long, e.g. \(Z(t) \to \infty\) then the electron, subject to such a field, would gain the final energy:

\[
U_{ev,final} = \frac{e^2 E_0^2}{4m \omega^2}.
\] (5.18)

The equation (5.18) has the same form as the ponderomotive energy that the electrons gains when it interacts with a tightly focused laser beam. [14].
5. 5. 2. INTERACTION WITH A FINITE PULSE

Although the assumption of an infinitely long pulse helps to calculate the maximum energy that an electron can gain when accelerated by the ponderomotive force (see equation (5.18)), it is insufficient to explain the results of acceleration obtained with the pulsed laser. Consider again the differentiation equation (5.16). It can be rearranged in the form:

\[ \ddot{y}(t) = g\ e^{-y(t)}, \quad (5.19) \]

where \( y(t) \) is the generalised position of the electron:

\[ y(t) = \frac{2Z(t)}{\lambda_{ce}}, \quad (5.20) \]

and \( g \) is the generalised energy:

\[ g = \left( \frac{eE_0}{\lambda_{ce} m \omega} \right)^2. \quad (5.21) \]

Conservation of energy requires that the total energy of the system, kinetic and potential must be preserved if the system is isolated from the environment*. Keeping that in mind, equation (5.19) can be rearranged and integrated in the form:

\[ \frac{1}{2} \dot{y}^2 + ge^{-y} = 0. \quad (5.22) \]

The first term of the equation (5.22) corresponds to the kinetic energy of the system since it only depends on the velocity of an electron. Therefore, the second term corresponds to potential energy of the system since it depends directly on the position of the electron. The total energy of the system at the beginning of acceleration process is equal to the total energy at the end:

---

* The conservation of energy of the considered system can be explained within the classical limit in the terms of the multiphoton absorption by the electrons. The electrons that interacts with the evanescent field, absorb photons therefore their energy increases. In quantum mechanical terms, one can say that the electron that undergoes the oscillatory motion in the evanescent field, in response emits electromagnetic radiation that is in the opposite phase to the evanescent field. As a result the absorption of light can be observed.
\[ U_0 = \sqrt{\frac{1}{2} \dot{y}^2(0) + g e^{-y(0)}} = \sqrt{\frac{1}{2} \dot{y}^2(t) + g e^{-y(t)}}. \] (5.23)

Rewriting equation (5.23) in the form:

\[ \frac{dy}{dt} = \sqrt{2U_0 - 2ge^{-y}}, \] (5.24)

gives the integral equation:

\[ \int_{y(0)}^{y(t)} \frac{dy}{\sqrt{2U_0 - 2ge^{-y}}} = \int_0^t dt. \] (5.25)

This can be evaluated as:

\[ t(y) = \frac{2}{\sqrt{2U_0}} \ln \left[ 2\sqrt{U_0 e^y} + 2\sqrt{U_0 e^{-y} - g} \right]_{y=y(0)}^{y=y(t)}. \] (5.26)

Equation (5.26) expresses the dependence of time of electron interaction with the evanescent field as a function of the electron’s position. This is a rather unusual representation of a function in physics. However, it is the easiest way possible to solve equation (5.25). To obtain physically meaningful result the inverse function needs to be found. This can be done numerically and will be presented later.

Since the purpose of this evaluation is to find how electron kinetic energy depends on time, equation (5.26) will be re-written in the terms of the generalised kinetic energy \( \dot{y}(t) \) only. From equation (5.23) follows:

\[ e^y = \frac{g}{U_0 - \frac{1}{2} \dot{y}(t)}. \] (5.27)

The electron starts it motion at the surface, i.e. \( y(0)=0 \) then \( U_0 \) in terms of initial generalised kinetic energy \( \dot{y}_0^2 \) is defined by equation (5.23) as:

\[ U_0 = \sqrt{\frac{1}{2} \dot{y}_0^2 + g}. \] (5.28)

Inserting equations (5.27) and (5.28) back into equation (5.26) gives:
t(y) = 
= \frac{2}{\sqrt{y_0^2 + 2g}} \ln \left[ 2 \sqrt{\frac{g(y_0^2 + g)}{(\frac{1}{2}y_0^2 + g - \frac{1}{2}y^2)} - 2 \sqrt{\frac{g}{(\frac{1}{2}y_0^2 + g - \frac{1}{2}y^2)}}} \right]_{y_0}^{y} \tag{5.29}

Now coming back to the original variables (equations (5.20) and (5.21)) the kinetic energy of an electron is:

\[ U_{\text{kin}} = \frac{m\dot{Z}^2}{2}. \tag{5.30} \]

Therefore the generalised kinetic energy \( y^2 \) in the terms of energy is:

\[ y^2 = \frac{8}{m\lambda_{ev}^2} U_{\text{kin}}. \tag{5.31} \]

The equation (5.31) and its limit at the start of the acceleration \( y^2(U_{\text{kin,0}}) \) can be substituted back into equation (5.29) together with equation (5.21) to form the final solution. However, it is difficult to draw any conclusions from the above equation about the energy of the electron, which it gains as a result of its interaction with the evanescent optical field because of its complex form. In the following section a numerical solution of equation (5.29) is found and the inverse function will be presented graphically.

### 5.5.3. Interaction with a Finite Pulse – Graphical Interpretation

Assume that the laser pulse intensity envelope is square shaped such that the probability \( P(t) \) of emitting an electron at any time during the laser pulse duration \( \tau_p \) is constant and equal to:

\[ \int P(t)dt = \frac{1}{\tau_p} \int_0^{\tau_p} dt \tag{5.32} \]
The probability that an electron gains any energy anywhere during the pulse $\tau_p$ is equal to unity. Time $\tau_p$ is related to the electron’s kinetic energy, as given by equation (5.29). Now, substituting equation (5.29) into equation (5.32) results:

$$
\int P(t)dt = \frac{1}{\tau_p} \int dt(U_{kin}) = \int \left[ \frac{\partial t(U_{kin})}{\tau_p \partial U_{kin}} \right] dU_{kin}.
$$

(5.33)

Therefore, the probability of finding the electron at a given energy $dU_{kin}$ is:

$$
P(U_{kin})dU_{kin} = \frac{\partial t(U_{kin})}{\tau_p \partial U_{kin}} dU_{kin}.
$$

(5.34)

Using equation (5.34) it is possible to calculate and plot the probability distribution functions. The final energy of an electron can be evaluated using the Mathematica software package. The Mathematica code is shown in Appendix 2. The solution involves numerical evaluation of the equation (5.29). It diverges very slowly around zero, therefore only an approximate solution in this region was found. The calculated curves were integrated once more to check that they normalized properly. All values of these integrals were always close to one.

5.5.3.1. PROBABILITY OF ACCELERATION AS A FUNCTION OF SURFACE ELECTRIC FIELD AND ANGULAR FREQUENCY

Several different calculations were performed and their results are presented below. During the calculations the accelerating optical field was defined with:

- $E_0$ the maximum value of the surface electric field;
- $\lambda$ the wavelength of the exciting optical field;
- $\lambda_{ev}$ the decay length of the evanescent field;
- $\tau_p$ the duration of the optical pulse;

To simplify the calculations it has been assumed that the pulse has a square envelope. For each set of calculations, all but one variable were kept constant. In all cases it was assumed that the electron starts its acceleration process at the surface with zero initial kinetic energy.
FIG. 5.8. The probability of an electron gaining a given energy as a result of acceleration in the evanescent optical field; different curves \(a-f\) represent the distribution function for range of the surface electric field strength \(E_0\) 1x10^{11}, 3x10^{11}, 4x10^{11}, 5x10^{11}, 6x10^{11}, 7x10^{11} \text{ Vm}^{-1} \text{ respectively}; other parameters were set to \(\lambda = 800\text{nm}\) and \(\tau_p = 100\text{ fs}\).

FIG. 5.8 presents the probability of an energy distribution that an electron can gain in the evanescent optical field for a range of maximum values of surface electric field. Despite the unrealistic assumption of a square optical pulse the calculated probability distribution functions resemble the experimental functions (see FIG 4.3 b). The probability is nonzero up to a certain cut-off \(U_{max}\) that depends on the maximum value of the surface electric field. This wide and continuous spread of energies is caused by the start time of the acceleration process. The time at which the electron starts its acceleration process determines the duration of electron interaction with the ponderomotive force. The electron can start its acceleration process at any time during the duration of an optical pulse. The generation at the beginning of the optical pulse allows the longest interaction with the evanescent field hence the probability that the electron gains the maximum energy is high. However, when the acceleration process starts later during the pulse, the interaction time of an electron with the evanescent field is shorter. Therefore, the final electron kinetic energy is
lower than the maximum value. When the intensity of the accelerating field increases the probability of gaining the maximum energy in the evanescent field also increases. This is presented in FIG. 5.9.

**FIG. 5.9.** Plot a: the maximum electron energy after acceleration in an optical evanescent field as a function of the surface electric field $E_0$; the line is quadratic fit; plot b: the efficiency of the evanescent acceleration as a function of the surface electric field $E_0$; the fitted line is that of an exponential growth function; the conditions of the calculations are the same as those used for the data presented in FIG. 5.8.

To generate plot a in FIG. 5.9 the maximum value of energy for which the probability is different from zero (the cut-off energy) was taken from graph FIG. 5.8 and presented as a function of the surface electric field $E_0$. This curve follows a quadratic dependence. It is consistent with equation (5.18) that predicts a quadratic dependence of $U_{\text{final}}$ on the amplitude of the electric field vector $E_0$ of the driving optical field. The b plot shows the effectiveness of the acceleration process as a function of the surface electric field $E_0$. Values for this plot were again recalculated from the data presented in FIG. 5.8. Each data point represents the probability value of achieving the highest energy in the evanescent field $P(U_{\text{max}})$ with respect to the number of emitted electrons. In this case the effectiveness was defined as:
The effectiveness of acceleration defined with the above equation is an exponentially rising function of the maximum value of the surface electric field \( E_0 \). This suggests that the higher intensity of the incident light, the more effectively the electrons are accelerated in the evanescent field.

\[
\eta = P(U_{\text{max}})U_{\text{max}}. \tag{5.35}
\]

**FIG. 5.10.** The probability distribution of an electron acceleration in an optical evanescent field for various wavelengths of the exciting light \( \lambda \), curves a–f correspond 700, 800, 900, 1200, 1300, 1500 nm respectively; the surface electric field was constant for all calculations and set to \( E_0 = 3 \times 10^{11} \) V/m and the pulse duration was \( \tau_p = 100 \) fs.

FIG. 5.10 shows the probability distribution function for a range of different wavelengths of the optical field \( \lambda \) but constant surface electric field \( E_0 \) and pulse duration \( \tau_p \). With increasing wavelength \( \lambda \) the probability of gaining high kinetic energy by an electron in the evanescent field increases. Again, the energy spectra are continuous and always extend from 0 eV to a certain cut-off energy \( U_{\text{max}} \) beyond
which the probability is equal to zero. $U_{\text{max}}$ is inversely proportional to $\omega^2$, which is in agreement with equation (5.18), see plot b, in FIG. 5.11. As before, the effectiveness of the acceleration (equation (5.35)) was calculated as a function of the angular frequency of the driving optical field and is shown in FIG. 5.11 (curve b). Again the effectiveness of the acceleration process increases when the electron interacts with the optical field of a longer wavelength. The probability of gaining the highest energy rises exponentially as an inverse function of $\omega^2$.

FIG. 5.11. a curve: the efficiency of the acceleration on the evanescent field as a function of the angular frequency $\omega$; b curve: the maximum energy of an electron accelerated in the evanescent optical field as a function of a frequency of the driving EM wave $\omega$; the fitted line is the reciprocity of a $\omega^2$ function; other parameters of the calculations were the same as those for data presented in FIG. 5.10.

The above results can be explained by studying the oscillatory motion of an electron. When the strength of the surface electric field increases the value of the force that acts on the electron during every half cycle of the pulse also increases. This suggests that the electron experiences higher accelerating force within half of one optical cycle of the evanescent field. This means that the amplitude of the
oscillatory part of its motion $\xi(t)$ (see equation (5.10)) is magnified. Increasing the wavelength (i.e. decreasing the frequency) of the driving EM field changes the period $T = 2\pi / \omega$ of the oscillatory motion. This means that the path length of an electron during half of an optical cycle of the evanescent field increases whereas the accelerating force is the same. In both cases the overall result is indeed the enhancement of the maximum energy of the electron. The effectiveness of the acceleration shows similar frequency dependence. Once electrons are subjected to stronger optical fields or radiation of a lower frequency, more and more electrons within one bunch can be accelerated to the maximum energy that is described by equation (5.18). It is important to remember that a uniform electron distribution at the start of the acceleration process was assumed and the initial phase of the laser pulse was not considered in the calculations. Furthermore, the unrealistic assumption of a square pulse was taken. It is probable that this assumption causes the sharp energy cut-off and it may affect the value of the effectiveness of the acceleration process. This will be a subject of Chapter 7.

5. 5. 3. 2. DURATION TIME OF OPTICAL PULSE

The next parameter that can be changed experimentally is the duration time of the optical pulse $\tau_p$. This was not included in equation (5.18) as the solution was given for an infinitely long pulse. However, using equation (5.29) it is possible to set any value of the pulse duration time $\tau_p$.

FIG. 5.12 presents the set of electron energy probability distribution functions for the range of different duration times $\tau_p$ of the optical pulse, from 20 fs to 200 fs. The surface electric field was constant throughout all calculations and was set to $E_0 = 3 \times 10^{11}$ V/m. The wavelength was also constant and was set to $\lambda = 800$ nm.
FIG. 5.12. The probability of the ponderomotive acceleration of an electron for a variable duration time $\tau_p$ of the optical pulse; the curves a – f correspond to 20, 40, 60, 100, 150, 200 fs optical pulse duration $\tau_p$ respectively; the surface electric field was constant for all calculations and was set to $E_0 = 3 \times 10^{11}$ V/m and the wavelength was set to $\lambda = 800$ nm.

Again the individual spectra span continuously from 0 eV up to a certain cut-off energy. When the duration of the optical pulse is increased, the electron can be accelerated to the higher energy. However, above a certain threshold value (about 160 fs for $E_0 = 3 \times 10^{11}$ V/m and $\lambda = 800$ nm), the maximum energy reaches the value defined by the equation (5.18) and does not increase any further. This is presented in FIG. 5.13 a. At the same time, the effectiveness of the acceleration process rises exponentially (curve b in FIG. 5.13).
FIG. 5.13. a curve: the maximum energy of an electron accelerated in the evanescent optical field as a function of the optical pulse duration $\tau_p$ (squares), the value of the final energy of an electron accelerated by the CW laser (straight line); b curve: the effectiveness of the acceleration in the evanescent field as a function of optical pulse duration $\tau_p$; other parameters of calculations were the same as those for FIG. 5.12.

When the electron interacts with the optical evanescent field, a certain minimum interaction time is required in order to attain the maximum possible energy $U_{final}$ for a given surface electric field. When $\tau_p$ is shorter than the threshold value $\tau_{threshold}$, there is insufficient time for the acceleration to reach $U_{final}$. Once the pulse is sufficiently long, the energy of the evanescent field is fully converted into the electronic kinetic energy. Therefore, all electrons, which are generated at times longer than or equal to the threshold value of $\tau_p$ will be accelerated to the $U_{final}$ defined by equation (5.18). Hence for long laser pulses, $\tau_{pulse} > \tau_{threshold}$, only the effectiveness of acceleration can increase.

The calculations presented above show that there are several possible ways to improve the yield and the efficiency of the evanescent wave acceleration once the electrons are libarated form the metal. Maximising the duration of the optical pulse and its intensity together should yield the best results. Furthermore, the laser pulse need to be sufficiently long to accelerate the whole electron bunch to the maximum...
possible energy that is given by equation (5.18). However this discussion does not take into account the excitation processes in the presence of long (longer the 200 fs) and high intensity (more the 10 GWcm\(^2\)) laser pulse. The excitation process of the electrons inside the metal film in the presence of a femtosecond laser pulse will be discussed in Chapter 7. Furthermore, such beams can cause permanent damage to the surface that is irradiated. The damage threshold depends on the intensity and the time of interaction of the laser beam with the surface. If strong laser light is applied to the sample for a sufficiently long time then the ion lattice starts to heat up and ion emission from the sample should be expected. The intensity of the ion emission and the extent of the damage caused by the laser heating will depend on the intensity and the duration time of the laser pulse. The usual value of the laser intensity above which damage occurs is a few tens of TW/cm\(^2\) for ultrashort laser pulses [16-18] In this work the typical laser pulse duration was in the ultrashort regime and the laser intensity did not exceed TW/cm\(^2\) at the metal layer. Therefore, ion emission should not be observed and the material should not be damaged.

In this chapter the equation of averaged motion in an optical evanescent field was solved within the classical limit. It was shown that the energy the electron can gain when it interacts with the evanescent field depends on the value of the surface electric field and its frequency. The electron can gain the maximum possible energy defined by equation (5.18) if it interacts with the evanescent field longer than a certain threshold interaction time (see FIG. 5.13) that depends on the parameters of the field itself. Of course to fully understand the acceleration process a detailed analysis of the initial condition of acceleration needs to be performed. Therefore, the results of the electron emission spectra will be presented in the next chapter and this will be followed by the analysis of the emission process in the presence of intense, ultrashort laser irradiation.
5. 6. REFERENCES


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CHAPTER 6

ELECTRON ACCELERATION IN AN EVANESCENT FIELD

In the previous chapter, the basic theory of evanescent wave acceleration was introduced. The derivation was based on a simplified model of the accelerating force. The equation of average motion in the evanescent field was solved using a Landau-Lifshitz approximation of two independent motions of an electron in the evanescent field assuming a square shape for the laser pulse envelope. On the basis of this theory the probability distribution function for the kinetic energy gained by an electron as a result of an interaction with the evanescent field was calculated. In this chapter, experimental results will be presented and compared with the theory. The energy spectra of electrons emitted from thin gold and silver metal films in the Kretschmann geometry under various experimental conditions will be discussed. Finally, a numerical model of accelerated motion in an evanescent field will be presented. This will lead to new aspects of electron – evanescent field interactions.

6.1. SUMMARY OF THE EXPERIMENTAL TECHNIQUE

The results presented in this chapter show the electron emission spectra measured for silver and gold films in the standard Kretschmann configuration. Two TOPS laser systems described in Chapter 2, section 2.3.4 were used in the experiments presented here. A note will be added to the description of each experiment to indicate, which laser system was used. A basic time-of-flight set up was used throughout the experiments and is described in detail in Chapter 2. Data sets for each metal film were recorded for a range of incident light intensities, ranging from 0.4 GW/cm$^2$ to 40 GW/cm$^2$ and variable duration of the laser pulse. All experiments were performed using a wavelength of 800 nm.
FIG. 6.1. A typical averaged electron pulse trace as observed with a digital oscilloscope (curve a) and a trigger signal from the Ge photodiode (curve b); only a small fraction of the total laser beam power was directed onto the triggering photodiode to avoid saturation.

FIG. 6.1 shows a typical electron pulse signal as detected by the multichannel array plate and recorded with a digital fast sampling oscilloscope and TOPS BNF laser system. Afterwards each data set was recalculated in order to present spectra in the energy domain using the formulae given in equations (2.5) and (2.6). This procedure resulted in a high density of data points near 0 eV and a lower density of data points for higher energies. The results were binned into 0.02 eV size cells to selectively average them.

6.1.1. DEPENDENCE ON INCIDENCE POWER DENSITY

6.1.1.1. EMISSION FROM SILVER FILMS

FIG. 6.2 presents a semi-logarithmic plot of the energy distribution of the electrons emitted from silver films for a series of incident beam intensities, ranging from 0.4 GW/cm$^2$ to 10.3 GW/cm$^2$ provided by the TOPS BNF system. The FWHM pulse length was fixed at 100 fs. The incident beam was focused with a 1 m focal length lens and placed 0.7 m away from the metal film. This resulted in a 4 mm laser beam cross-section at the silver film and allowed the beam to be focused behind the
prism material, preventing possible self-phase-modulation effects in the prism material. However, only a small fraction of the laser beam reached the metal at the exact SP resonance angle $\theta_{SP}$. This arrangement led to narrowing of the interaction area of the laser beam with the metal film. This was evident by examination of the image of the beam as it left the prism. A sharp dark line appeared across the laser beam projection at a flat surface, see FIG. 6.3. The position of the line depended on the angle of the incidence of beam.

![Semi-logarithmic plot](image)

**FIG. 6.2.** Semi-logarithmic plot of the energy spectra of photoelectrons emitted from a silver film in the Kretschmann configuration for a range of peak power densities, from a to e: 0.4, 3.0, 4.6, 6.4, 10.3 GW/cm$^2$; the pulse duration was 100 fs, the beam was focused with a 1 m focal length lens and its diameter at the metal layer was 4 mm.

The experimental data presented in FIG. 6.2 resemble the probability distribution functions calculated using the theory described in Chapter 5 FIG 5.7. The energy distribution spreads out continuously from nearly 0 eV up to a few tens of eV. As the laser power density increased, the electrons reached higher kinetic energy. No individual, uniformly distributed peaks were observed, supporting the theory that the emission does not occur via a multiphoton process. For the lowest power density it is found that the distribution has a width (HWHM) of about 0.2 eV.
With increasing laser power density, the peak broadens with a maximum at about 0.5 eV and there appears to be a cut-off energy. This cut-off is proportional to the incident laser power and is about 50 eV at 10.3 GW/cm$^2$. This corresponds to about 33 photons that would need to be absorbed by the electron to be excited to 50 eV.

![FIG. 6.3. Schematic diagram of ray arrangement in the beam as it reaches the metal film; left: focused beam; right: collimated beam; the focusing results in only a small fraction of the whole laser beam reaching the sample at the SP resonance angle $\theta_{SP}$, and part of the laser beam cross section is absorbed in the metal film; collimation results in absorption of the laser beam across its whole cross section as the SP resonance conditions are fulfilled for each ray in the beam.](image)

The evanescent field acceleration theory states that the maximum number of electrons should be detected with 0 eV energy, which does not quite agree with the experimental results (see FIG. 6.4). It is possible that such discrepancies are caused by the assumption of the square laser pulse during the derivation of the equation of motion in the evanescent field. It results in a constant magnitude of the surface electric field throughout the whole duration of the laser pulse. However, the Gaussian or the Lorentzian shape best approximates the envelope of the optical pulse. This leads to the variations of the maximum value of the electric field vector strength from one optical cycle to another. Alternatively, it could be the result of the initial condition of the acceleration process. To verify this, numerical calculations will be presented where the laser pulse envelope will be approximated with a Gaussian shape and the initial phase coefficient will be incorporated.

The data presented in FIG. 6.4 reveal one more unexpected feature. When the laser power density is increased above 5 GW/cm$^2$, oscillations of a constant frequency appear to be superimposed onto the data sets. The frequency of the oscillations is constant for all plots and approximately equal to 14 MHz. Their
amplitude increases with increasing laser power density. It is possible that this is a result of the plasma oscillations in the electron bunch immediately after the generation process at the metal vacuum interface. It is supported by the fact that the oscillations are only observable for high laser power densities when the number of electrons in the bunch is highest.

The definition of the plasma frequency is [1]:

$$\omega_p = \sqrt{\frac{ne^2}{\epsilon_0 m_e V}}, \quad V = \pi r^2 l_p \quad (6.1)$$

where $n$ is number of electrons in a bunch; $e, m_e$ and $\epsilon_0$ have their standard meaning of absolute value of the elementary charge, the electron mass and the permittivity of free space respectively. $V$ is a volume of the electron bunch and can be calculated using its radius $r$ in the plane parallel to the metal surface and its length $l_p$ in the direction perpendicular to the surface.
In this way, it may be possible to estimate the duration of the electron pulse. However, it is difficult to estimate the exact number of electrons emitted from the metal layer and the length of the pulse $l_p$. As the electron beam travels across the time-of-flight tube, the electrostatic forces within the bunch cause the expansion of the beam, the so-called space charge effect. Therefore, the number of detected electrons depends on the cross section of the multichannel array plate and the length of the time-of-flight tube. To verify this, further measurements need to be done, which exceed the scope of this work.

6.1.1.2. EMISSION FROM GOLD FILMS

The spectra of electrons emitted from gold films are similar to those generated from silver films. The results are presented in FIG. 6.5 and FIG. 6.7. Again each dataset was recorded for different laser power densities, ranging from 1.6 GW/cm$^2$ to 38.2 GW/cm$^2$ provided by the TOPS BNF system. The laser beam was collimated and its diameter was 5 mm. The pulse duration was set to 100 fs as before.

![Image](image.png)

**FIG. 6.5.** Semi-logarithmic plot of energy spectra of photoelectrons emitted from a gold film in the Kretschmann configuration for various laser peak power densities in the range (a-e): 1.6, 9.5, 17.5, 25.5, 36.2 GW/cm$^2$; the pulse duration was 100 fs, the beam was collimated and its diameter was 5 mm.
The maximum energy that is gained by photoelectrons emitted from gold films is lower compared to those emitted from silver films. At a 10 GW/cm\(^2\) incident laser intensity the maximum energy is 55 eV for electrons emitted from silver and 30 eV for electrons emitted from gold. This gives a ratio of 1.8. This value is very close to the theoretically calculated field enhancement ratio for silver and gold films using the SP excitation theory, which is 1.7 (see Chapter 3, FIG 3.7). One can conclude that there is no significant difference between the energy spectra of the electrons generated from gold or silver films apart from the different value of the field enhancement factor.

![Graph showing the maximum electron kinetic energy as a function of the incidence laser power. The two data sets correspond to the final \(U_{\text{kin}}\) of the photoelectrons emitted from silver (squares) and gold (dots) films in the Kretschmann configuration; the fitted straight lines are of a slope of \(~5\) (silver) and \(~3\) (gold).](image)

**FIG. 6.6.** The maximum electron kinetic energy as they interact with the accelerating force as a function of the incidence laser power; the two data sets correspond to the final \(U_{\text{kin}}\) of the photoelectrons emitted from silver (squares) and gold (dots) films in the Kretschmann configuration; the fitted straight lines are of a slope of \(~5\) (silver) and \(~3\) (gold).

When the laser intensity is increased further, electrons can gain higher final energy, but the cut-off energy becomes less pronounced. This is not supported by the theory described in Chapter 5 and suggests that there might be another process that takes part in electron acceleration. The magnitude of this interaction would need to be dependent on the surface electric field strength \(E_0\) and therefore the laser power density. Analysis of the cut-off energy as a function of the laser incidence power
leads to similar conclusions (see FIG. 6.6). If one assumes that the ponderomotive force is the only accelerating force then the maximum electron energy should scale as a square function of the surface electric field, see equation (5.18) and FIG 5.8. However, FIG. 6.6 shows that the maximum energy scales almost linearly with the laser intensity within the experimental error.

**FIG. 6.7.** Linear plot of the low energy part of the electron energy spectra presented in FIG. 6.5 for various laser peak power densities in the range (a-e): 1.6, 9.5, 17.5, 25.5, 36.2 GW/cm$^2$; all other experimental details remain the same.

Close examination of the low energy part of the spectrum shows that there are no electrons detected with zero kinetic energy as in the case of emission from silver films (see FIG. 6.4). Also, there is no oscillation pattern superimposed on the spectra. The only other difference between the two experiments is the geometry. The oscillations were observed only when the laser beam was focused during the experiment. FIG. 6.7 presents the results when the laser beam was collimated resulting in a uniformly defined wave front. The absorption and therefore the electron emission occurred across its whole cross section at the metal layer (see FIG.
6.3). Such an arrangement causes the wavefront to sweep across the beam cross-section. This creates an optical path difference among the individual rays in the laser beam as they reach the surface at the SP resonance angle $\theta_{SP}$. If the collimated 4 mm diameter beam hits a metal surface at 42°, the SP resonance angle for the gold film, then the optical path difference between the outermost rays is 27 fs (see FIG. 6.8). This causes progressive SP excitation across the beam cross-section and in the same way delays the electron emission. The calculated delay time corresponds to about 37.5 optical cycles at 800 nm. This might lead to a lower photoelectron density, $n/V$ immediately after the generation process and longer spatial length $l_p$ of the electron bunch, see equation (6.1).

![FIG. 6.8](image)

**FIG. 6.8.** The optical path difference between the outermost rays of the collimated laser beam as it hits a surface at the SP resonance angle $\theta_{SP}$; the optical path difference results only from the geometry of the arrangement.

### 6.1.1.3. TOTAL NUMBER OF ELECTRONS

Analysis of the number of electrons in the bunch as a function of the incident laser power yields interesting results. Two different values of the electron number were taken into consideration: the total number of detected electrons and the number of electrons at the peak of the energy spectrum. The energy distribution peaks at about 0.5 eV, (see FIG. 6.4 and FIG. 6.7). This means that probing the electron number at the peak value probes the number of low energy electrons effectively. The results are presented in FIG. 6.9.
When the laser intensity is low (up to a few GW/cm$^2$), the electron number in the emitted bunch increases but its population mostly consists of low energy electrons. When the power density of the laser beam is increased the total number of electrons does not rise any further but at the same time the number of electrons at the spectrum’s peak value decreases. This means that more electrons gain higher final energy. It is possible that in the first instance when the intensity of an exciting laser beam is less then a few GW/cm$^2$ the generation rate of electrons rises but the surface electric field $E_0$ is not strong enough to accelerate electrons efficiently. When the laser power is increased above a certain threshold value, the absorption in the metal layer saturates and no more electrons can be generated. However, higher laser power density results in a stronger accelerating force, therefore more electrons are accelerated to higher kinetic energies.
6. 1. 2. TIME OF INTERACTION IN THE EVANESCENT FIELD

FIG. 6.10 shows electron energy spectra for various duration times of the optical pulse $\tau_p$ and constant strength of the surface electric field $E_0$. The electrons were emitted from a gold film in the Kretschmann configuration. The Alfa TOPS laser system was used and the laser beam was collimated and its diameter set to 20 mm. The duration of the laser pulse was altered by varying the separation between the grating pair in the compressor part of the laser system. Depending on the direction of the change (increasing or decreasing the distance between the grating pair) a positive or negative chirp in the laser pulse was introduced. The electron spectra were measured for both chirps and no significant changes were noticeable. FIG. 6.10 shows only two spectra, the reason for such a poor statistic is that the measurements were taken with a new laser system (Alfa TOPAS laser) that still needed final adjustment. The laser beam showed signs of astigmatism. A proper telescope to reduce the beam diameter could not be build without compromising the temporal shape of the laser pulse.

FIG. 6.10. Linear plot of the electron energy distribution for different FWHM duration times $\tau_p$: a: 50 fs (unchirped), b: 100 fs (chirped); the laser beam was collimated and its diameter was 20 mm, the intensity was constant and set to 2.2 GW/cm$^2$; the emission was observed from a gold film in the Kretschmann configuration.
In the experiment presented in FIG. 6.10, only the time duration of the laser pulse is changed during the experiment, whereas the intensity remains constant. The constant peak power density provides a constant value of the electric field at the surface of the metal. Only the interaction time of electrons with the EM wave is affected. In both cases the final kinetic energy of the majority of electrons is low, about 0.5 eV. When the laser pulse duration $\tau_p$ is increased by a factor of two the number of electrons at the peak of the spectrum also increases by the same factor. However, the total number of detected electrons rises by a factor of 2.6. Such a result can lead to two possible conclusions. Firstly, a longer laser pulse prolongs the interaction of the EM wave with the metal surface affecting the emission conditions. This aspect of an evanescent wave acceleration will be discussed in more detail in the following chapter. Secondly, the interaction time of the evanescent field with free electrons is affected by increasing the pulse duration. As described in Chapter 5, longer interaction time causes more efficient evanescent wave acceleration. For longer laser pulses, the maximum electron energy should asymptotically approach the energy value defined by the equation (5.18), see FIG (5.12). Indeed the maximum value of the energy in the spectra presented in FIG. 6.10 increases by a factor of 1.6.

6. 1. 3. COMPUTER SIMULATIONS

A one-dimensional numerical model was developed to simulate the acceleration process in an optical evanescent field. The Verlet algorithm [2] was used to calculate the position and the final kinetic energy of an electron. This has already been described in detail in Chapter 5 and the computer code is presented in Appendix 1. It was assumed that the motion of an electron can be treated classically at all times. Each step during numerical evaluation is calculated from the previous two steps and the force that acts on an electron. Each step was again set to 1 as. For clarity, the accelerating force $F_a$ will be defined again here. $F_a$ is a function of position and time and can be expressed as:

$$F_a(z,t) = eE(z,t)$$  \hspace{1cm} (6.2)

The electric field vector has only one non-zero component that is perpendicular to the metal surface, the $z$ direction, as it was explained in Chapter 3. The maximum
$E_0$ of an electric field vector $\vec{E}$ is at the metal surface e.g. at $z = 0$. Its value decays exponentially for all $z \geq 0$ and is nearly constant for all $z < 0$. As a function of time the value of the $\vec{E}$ vector follows the envelope of the optical pulse and simultaneously oscillates at the frequency of the driving optical field $\omega$. This can be expressed as:

$$F_0(z,t) = eE_0 \cdot \text{evanescent}(z) \cdot \text{gauss}(t) \cdot \cos(\omega t + \varphi_0). \quad (6.3)$$

**FIG. 6.11.** The schematic representation of the time (right) and position (left) dependence of the strength of the electric field vector $E$; left: the amplitude of $\vec{E}$ decays exponentially as a function of position away from the metal surface, inside the metal its value is small compare to its maximum value $E_0$; right: $\vec{E}$ oscillates with frequency $\omega$ and its value follows the envelope of the Gaussian shape with $\tau_p$ as FWHM duration time.

The evanescent and the Gaussian components are defined as:

$$\text{evanescent}(x) = \begin{cases} \exp \left[ -\frac{z}{\lambda_{ev}} \right], & z \geq 0 \\ \text{const.}, & z < 0 \end{cases}, \quad (6.4)$$

$$\text{gauss}(t) = \exp \left[ -2 \ln(2) \left( \frac{t - t_0}{\tau_p} \right)^2 \right], \quad (6.5)$$
where $\lambda_{ev}$ is the decay length of the evanescent optical field, $\tau_p$ and $\tau_0$ are the FWHM duration time of the optical pulse and the time of arrival of the maximum of the Gaussian pulse at the surface respectively, and $\phi_0$ is the initial phase of the laser pulse.

This detailed definition of the accelerating force gives an opportunity to analyse the acceleration process with respect to all variables. The numerical results presented below can be compared with the graphical representation of the analytical solution of the equation of motion in the evanescent field (see Chapter 5). The dependence of the electron energy distribution with respect to the maximum surface electric field $E_0$, the duration of the optical pulse $\tau_p$ and the frequency of the driving field $\omega$ were discussed. However, in the analytical solution the result was the averaged function of time and did not include the initial conditions of the acceleration process. Here, the analysis can be tested using the complimentary numerical method. All these variables are the parameters governing the electron motion throughout the whole acceleration process. Additionally, the initial phase $\phi_0$ of the optical field can be chosen arbitrarily. This parameter defines the initial conditions of an acceleration process, consequently pointing toward the generation process analysis.

6.1.3.1. THE ELECTRON PATH

Using the Verlet algorithm it is possible to follow a path of one individual electron that starts its motion at a specific time with given initial conditions. Initially, a path of a few individual electrons will be followed. All optical field parameters will be kept constant and the initial phase $\phi_0$ will be varied over $2\pi$. FIG. 6.12 presents the position of an accelerated electron in the evanescent field as a function of time for several different values of the initial phase of the optical field $\phi_0$. To follow the path of the electron in all cases, a simplified definition of the evanescent part of the accelerating force was used:

$$evanescent(z) = \exp\left[-\frac{z}{\lambda_{ev}}\right], \text{ for all } z. \tag{6.6}$$

The maximum value of the surface electric field $E_0$ was set to $10^{11} \text{ (V/m)}$ and the surface electric field at $t = 0$, $E_{\text{start}}$ was set to $10^{10} \text{ (V/m)}$, the definition and the
importance of the $E_{\text{start}}$ parameter is explained in Chapter 5. The FWHM pulse duration time $\tau_p$ was set to 100 fs and the decay length of the evanescent field $\lambda_{ev}$ set to 1 $\mu$m (as it was calculated using the method presented in Chapter 3). The simulation step was carried out using time steps of $10^{-18}$ s.

**FIG. 6.12.** Main graph: simulated plot of the position of an electron as it is accelerated in the evanescent optical field; different curves correspond to different initial phases $\phi_0$ of the optical pulse at the $t=0$ fs (a-d): 90, 150, 180, 270°; inset: the final kinetic energy as a function of the initial phase $\phi_0$ of the EM field at the surface (curve e); the maximum value of the surface electric field was set to $E_0 = 10^{11}$ V/m, the value of the surface electric field at the beginning of simulation was set to $E_{\text{strat}} = 10^{10}$ V/m and the FWHM duration time of the optical pulse $\tau_p$ was set to 100 fs; the final kinetic energy was recorded after 500 fs of acceleration; for clarity of the presentation $\cos(\phi_0)$ is plotted (curve f)

Different values of the initial phase $\phi_0$ give effectively different values of the surface electric field at the beginning of the acceleration process $E_{\text{start}}$:

$$E_{\text{start}}(t = 0) = E_0 \text{gauss}(t) \cos(\omega t + \phi_0).$$  \hspace{1cm} (6.7)
The electron path crucially depends on the strength of the surface electric field at the beginning of the acceleration process. Its final kinetic energy varies over a wide range of values, as is shown in the inset of FIG. 6.12. An electron gains the highest possible energy if the phase of the EM wave at the beginning of generation is \( \varphi_0 = \frac{3\pi}{2} + 2k\pi, \ k \in \{0, N\} \). When the initial phase is in the range \( \varphi_0 \in (2k\pi, \pi + 2k\pi), \ k \in \{0, N\} \) during the first phase of acceleration the electron is “pushed” backward into the metal (where \( z < 0 \)) and re-emerges out of it again after some time. In a real system the electric field inside the metal film is nearly zero. The non-zero value of the electric field inside the metal arises from the fact that at 800 nm the sample cannot be treated as perfect conductor. This indicates a non-zero imaginary part of the dielectric function of the metal. (see calculations presented in FIG. 3.7). Therefore, in a real system the electron needs to be liberated again via the emission process to start the acceleration process again. During further numerical simulations, once the position of an electron became negative its final energy was not calculated. This behaviour however, points out that the initial phase of the EM field plays an important role in the evanescent wave acceleration process.

6.1.4. ACCELERATING A BUNCH OF ELECTRONS

A simple numerical model was developed to simulate the acceleration process of a bunch of electrons. It is based on the assumptions:

- The initial energy of all electrons is always zero and is independent of the value of the surface electric field at the moment of generation;
- The threshold value of the surface electric field \( E_{strat} \) was chosen arbitrarily, \( E_{strat} \) is the lowest value of the surface electric field vector above which acceleration can start (see FIG. 6.14);
- The subsequent electrons start the acceleration process at uniform time intervals \( \delta t \) during the laser pulse duration (see FIG. 6.14);
- If the electron position at any time during the acceleration process is negative e.g. \( z < 0 \) then its final kinetic energy is not calculated as was explained in the previous section;
- Electrons do not interact with each other during the acceleration process;
6.1.4.1. THE TIME OF GENERATION

The kinetic energy was calculated from the velocity of an electron as explained in Chapter 5. The final energy of an electron was recorded when the acceleration time in the simulation is larger than a set value $\tau_{\text{end}}$. $\tau_{\text{end}}$ was found empirically and chosen as the smallest value of the acceleration time when no significant changes to the final kinetic energy were observed. It was typically set to 500 fs for $\tau_p = 100$ fs FWHM optical pulse duration.

![Graph showing the simulated final kinetic energy of 20,000 electrons as a function of the generation time.](image)

**FIG. 6.13.** Main graph: the simulated final kinetic energy of 20,000 electrons as a function of the generation time; the maximum surface electric field was set to $E_0 = 10^{11} V/m$, the FWHM pulse width was set to $\tau_p = 100$ fs; the initial phase $\phi_0$ was set to 270°; each value of the final kinetic energy was recorded after 500 fs; the inset shows the enlargement of the main graph as a function of phase $\phi = \omega t_{\text{generation}} + \phi_0$ at the generation time of the individual electrons.

**FIG. 6.13** shows the value of the final kinetic energy as a function of the generation time. The total number of electrons was set to 20,000, the maximum value of the surface electric field $E_0$ was chosen to be $10^{11}$ (V/m) and the surface electric field at which the acceleration started was set to $10^{10}$ (V/m). The duration of the
optical pulse at FWHM was $\tau_p = 100$ fs. The accelerating force definition introduced in equation (6.3) was used. The initial phase $\varphi_0$ was set to zero. The subsequent electrons started their generation process at the intervals of $\delta t = 12.8$ as. Effectively this procedure resulted in different values of the effective phase $\varphi_{\text{eff}}$ of the optical field at the beginning of the acceleration process for every individual electron:

$$\varphi_{n,\text{eff}} = \omega n \delta t + \varphi_0, \quad n = 1 \ldots 20000.$$  \hfill (6.8)

![Diagram showing the Gaussian envelope of the optical pulse and the definitions of the parameters used in the computer simulations.](image)

**FIG. 6.14.** The schematic diagram showing the Gaussian envelope of the optical pulse and the definitions of the parameters used in the computer simulations; $E_{\text{start}}$ is the threshold value of the surface electric field, above which the acceleration process is allowed, $E_0$ is the maximum value and $E_n$ is the value of $\vec{E}$ at the beginning of the acceleration process of the n-th electron, $t_g$ is the generation time, $\delta t$ is the time interval at which the individual electrons start their acceleration process, $n$ is n-th electrons, whose acceleration process started at $t_{g,n}$ and $n_{\text{maximum}}$ is the total number of electrons.

This assumption leads to the condition that an electron can only be accelerated when the following inequality is fulfilled (see FIG. 6.14):

$$E_n > E_{\text{start}} \rightarrow E_{n,\text{gauss}}(t_{g,n}) \cos(\varphi_{n,\text{eff}}) > E_{\text{start}},$$  \hfill (6.9)
and its position does not become negative at any simulation substep. This is true when the effective phase of the optical field falls in the range of angles (see the inset of FIG. 6.13):

\[ \phi_{\text{eff}} \in (\pi + 2k\pi, 2\pi + 2k\pi), \quad k \in \{0, N\} \]  

(6.10)

6.1.4.2. SIMULATED ENERGY DISTRIBUTION PLOTS

The statistics of the kinetic energy can be calculated from the data presented in FIG. 6.13 resulting in the energy distribution shown in FIG. 6.15, which is the averaged energy distribution for several simulations of the acceleration of an electrons bunch for various values of the initial phase \( \phi_0 \). Such a procedure had to be undertaken as the experimental data are the average result of electrons emitted from several laser pulses and the averaging was used to minimise the signal-to-noise ratio. The initial phase \( \phi_0 \) varies at random from one laser pulse to another. Additionally the laser peak intensity of the pulse fluctuates in time. Therefore all experimental data are averaged with respect to the initial phase and the fluctuations of the laser intensity.

Similar to the experimental data the distribution is uniform and spans the range zero eV to few hundred eV where it peaks. Two other main features are the peak at the low energy part of the spectrum and the cut off energy at high energy. In the simulation the energy of the majority of electrons is zero. When moving toward higher energies the number of electrons decreases. At about 25 eV there is a well-defined peak. Furthermore, the number of electrons still decreases uniformly and at about 200 eV a sharp cut-off appears. There are still some electrons accelerated to higher energies (up to 250 eV) the number over the 50 eV range remains nearly constant.
FIG. 6.15. top: Semi-logarithmic plot of the kinetic energy distribution for 20,000 simulated electrons; the data were averaged for several different values of the initial phase $\phi_0$ that was varied over $2\pi$; the total number of electrons at 0 eV is 380 (not shown in the plot to present the data at high energies more clearly); solid line: running average, all other parameters of the simulation are identical as in the data presented in FIG. 6.13. bottom: the electron energy distribution of electrons emitted from a flat gold metal film in the Kretschmann configuration at the laser intensity of 11.5 GW/cm$^2$ and the laser pulse duration of 100 fs.
The main difference between the experiment and the theory is the value of the energy at which certain features of the spectrum appear. The peak at 25 eV in the simulation is at much higher energies compare to the experimental data, where it always appeared at energies less then 1 eV. It is possible that the arbitrary defined values of the starting electric field $E_{\text{start}}$ together with the assumption of the uniform electron distribution at the beginning of the acceleration process results in the unrealistic very high and low energy values for certain number of electrons. Also, the assumption that the number of electrons that start the acceleration process do not change with intensity of the electric field strength, need to be verified. Furthermore, it is possible that the Coulombic interaction of electrons during the acceleration process also plays an important role.

6.1.4.3. DEPENDENCE ON THE FWHM, SURFACE ELECTRIC FIELD STRENGTH AND ANGULAR FREQUENCY

Using the numerical method, it is possible to analyse the final kinetic energy of an electron with the respect to:

- Surface electric field $E_0$;
- Angular frequency of the driving electromagnetic wave $\tau_p$;
- The FWHM duration time of the optical pulse $\tau_p$.

A path of only one electron at a time can be followed. Therefore the appropriate initial phase was chosen to assure that an electron will gain the highest possible energy. This condition is fulfilled when the starting surface electric field $E_{\text{start}}$ is at its maximum, e.g. $E_{\text{start}}=E_0$ (see FIG. 6.13) and the initial phase $\phi_0$ is set to $\frac{3\pi}{2}$ (see FIG. 6.12, inset). The results are in agreement with the solution of the analytical equation of the acceleration in the evanescent field. For brevity only one of the graphs will be presented and analysed.

FIG. 6.16 presents the dependence of the final kinetic energy of an electron as a function of the FWHM duration time of the laser pulse $\tau_p$. The calculations were made for several values of $\tau_p$ ranging from 20 fs to 200 fs. All other parameters were constant. The maximum and the starting surface electric field was set to $3 \times 10^{11}$ V/m,
and the wavelength of the driving optical field was $\lambda = 800$ nm. The time step in the Verlet algorithm was set to 1 and the final kinetic energy was calculated after 800 fs. When the $U_{\text{kin}}$ was calculated at longer times no significant changes were observed in data.

![Image](image.png)

**FIG. 6.16.** The normalised with respect to the maximum value in each plot, simulated final kinetic energy as a function of the laser pulse duration time $\tau_p$; the individual plots are the data sets for different values of the initial phase as indicated; the acceleration process started at the maximum value of the surface electric field; the kinetic energy was probed after 800 fs; the maximum strength of the surface electric field was set to $10^{11}$ V/m in all cases.

The dynamics of acceleration of an individual electron dramatically depends on the exact value of the surface electric field at the start of the acceleration process. Or more precisely the dynamics of the accelerating force during the acceleration process. The shape of the curves presented in FIG. 6.16 is similar to these obtained using the evanescent wave acceleration theory, see FIG 5.12. For clarity of presentation all plots were normalised. It was found that all plots tend asymptotically to a certain maximum final energy. When the initial phase $\phi_0$ was set to 90° this energy was reached for the shortest pulses. In Chapter 5 this energy was identified as
the ponderomotive energy, defined by equation (5.18). For the parameters used in the simulation, this energy should be 79.3 eV. This is in contrast to results presented in FIG. 6.16 where this value is about 217 eV. The unrealistic high value of the kinetic energy was previously observed in the data presented in FIG. 6.15.

It is possible to simulate the experimental and analytical results using the numerical method for certain fixed values of the initial parameters in the simulation. However, it is evident that the acceleration process depends not only on the magnitude of the accelerating force itself but also on its dynamics. Furthermore, the emission process of the electrons at the metal surface in the presence of the intense laser irradiation needs to be investigated in more detail.

The next chapter will present certain aspects of the electron emission in intense (above TWcm\(^{-2}\) intensity), ultrafast laser light (less than few tens of femtosecond). The theory of above threshold ionisation (ATI) and the tunnelling effect in the regime of the ultrashort and intense laser radiation will be introduced and compared with experimental and numerical results.

6. 2. REFERENCES

CHAPTER 7

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CHAPTER 7

ELECTRON EMISSION IN THE PRESENCE OF INTENSE LASER IRRADIATION

In the previous chapter, the experimental results were presented along with numerical simulations. It was concluded that the acceleration process does not depend only on the magnitude of the ponderomotive force but also on its dynamics at the beginning of the acceleration process. In this chapter, the emission of the electrons in the presence of intense laser irradiation will be presented. The liberation mechanism of an electron from the metal will be discussed in detail and the acceleration mechanism with the respect to the phase of the ponderomotive force will be analysed.

7. 1. IONISATION MODELS

The commonly used approach to studying the electron emission process in the presence of strong optical fields is known as “quasi-static” or “semiclassical” and it involves a two-step mechanism. Firstly, the ionisation process that describes the mechanism of electron transition from the bound state to the vacuum level is usually treated quantum mechanically. Secondly, the electron’s interaction with the external electric field is usually treated classically. The reason for such a treatment is that the ponderomotive potential affects the “original” electron energy distribution and obscures the emission effects. [1, 2]. Therefore, the ionisation processes in the presence of intense light will be described first, followed by the analysis of the initial conditions in the acceleration process in the rapidly oscillating field.
7. 1. 1. ABOVE THRESHOLD IONISATION

There is a number of papers where the emission mechanism from metals in the presence of surface plasmons (SPs) is discussed. [3-6] Typically the intensity of the laser beam applied to the metal surface to observe photoemission was in the range of 10-100 GW/cm$^2$ in the visible to infrared spectral range. The most commonly accepted theory of electron emission in the presence of intense laser light is the multiphoton ionisation (MPI) effect. [7-12] It can be described most simply by the classic Einstein equation:

\[ E_n = nh\nu - \phi, \]  

(7.1)

where $E_n$ is the energy of an electron that absorbed n-photons and $\phi$ is the metal work function.

The laser field is assumed to be too weak to noticeably modify the potential barrier at the metal interface. Therefore, the barrier suppression mechanism is neglected in the low laser intensity approximation (see case B in FIG. 7.1). The potential barrier at the metal/vacuum interface is constant and equal to the work function $\phi$. Photoemitted electrons need to absorb the minimum number of quanta to overcome the potential barrier. The typical value of the work function for noble metals is about 3 to 6 eV. It usually takes 3 to 4 photons (from the visible spectrum) to overcome the potential barrier if the electrons are excited with near infrared radiation. The presence of surface plasmon (SP) excitations will modify this simple model. This process was discussed in detail previously in Chapter 3.

There are several schemes that describe the ionisation models in the presence of an intense, ultrashort laser pulse. Most of the work in this field involves ionisation of a noble gases since it is convenient and controllable method for heating plasmas [1, 2, 13, 14]. The ionisation process of noble gases usually involves ultraintense laser fields at intensities of the order of $10^{18}$ W/cm. High power densities are usually obtained by tightly focusing the laser beam in a low density noble gas environment. The typical ionisation potential for noble gases is 10 eV or more, therefore high power densities are needed to make the ionisation possible. Although these laser intensities are much higher compared to those used in the work described here, the
research gives good insight into the electrons’ behaviour in strong, pulsed optical fields.

**FIG. 7.1.** Schematic drawing of the modification of the potential barrier at the metal interface by an external electric field; the energy at infinite distance away from the metal is always zero, the Fermi energy ($E_F$) is shifted with respect to this level by the applied external electric field; **A**: a negative electric field is applied; **B**: negligible small field is applied; **C**: a positive electric field is applied; the multiphoton process involves an absorption of several energy quanta to overcome the potential barrier; the field emission theory assumes the suppression of the potential barrier by the external electric field allowing the electron to tunnel through it.; above threshold ionisation involves absorption of many photons and emission over the suppressed potential barrier.

If an atom is subjected to a high intensity laser light, it is possible that an electron can absorb more than the required minimum number of photons to overcome the potential barrier. This process is known as above threshold ionisation (ATI) and is usually studied on the basis of a multiphoton ionisation process [12, 15]. The ATI process can be described by modifying the classical Einstein equation (7.1) to: [15, 16]

$$E_n = (n+m)\hbar \nu - \phi,$$

(7.2)

where $m$ is the number of excess absorbed photons by an electron.
In the ATI models the external optical field excites the electrons inside the ion/solid and at the same time effectively suppresses the potential barrier at the interface. However, if the optical field oscillates fast enough, electrons do not have enough time to undergo tunnelling emission through the thin potential barrier. Therefore, electrons overcome the barrier by absorbing $n + m$ number of photons, (see case C FIG. 7.1). The energy spectra of electrons emitted via the ATI process consist of a set of high order evenly spaced emission peaks, corresponding to absorption processes of different order. [1, 14] There is no explicit dependence on the phase of the optical field in the ATI formalism. However, it is possible that in the fast oscillating fields the whole atomic energy structure follows the driving optical field. The two extreme states are schematically presented in FIG. 7.1, case A and C. Therefore, the sensitivity of the ATI process to the driving optical field should be expected.

7.1.2. FIELD EMISSION

In the regime of a high intensity slowly varying laser field, the potential barrier at the metal surface can be suppressed long enough to allow electrons to tunnel through it. The barrier suppression mechanism or field emission has well been investigated for static dc fields [17-19]. At the metal surface a strong, electric field can suppress the potential barrier and therefore allow electrons to tunnel through it. If the electric field is applied at the metal/vacuum interface at 0 K, then the number of electrons emitted per unit time can be calculated with the classical Fowler-Northeim formula: [18, 20]

$$\frac{dN}{dt} = \frac{4}{3}\frac{16\pi me}{\hbar^3(\phi + E_f)b^2} E^2 \exp\left( -\frac{b\sqrt{\phi^3}}{E}\right),$$

(7.3)

where $\phi$ is the metal work function, $E_f$ is the Fermi energy, $E$ the applied external dc electric field, $b$ a correction constant to account for the image potential barrier lowering mechanism. The Fowler-Northeim theory assumes that the electrons in a
metal obey the Fermi-Dirac distribution and the metal surface is perfectly flat and clean.

The energy distribution of electrons emitted in the field emission process is uniform with the maximum energy corresponding to the Fermi energy of an atom at 0 K (FIG. 7.2, curve a). At higher temperatures the distributions broaden as shown in FIG. 7.2, curves b - e. At 0 K, the majority of electrons tunnel from the Fermi level while some electrons originate from lower energy levels of the ionised atom. At higher temperatures, it is possible for electrons to occupy energy levels above the Fermi level, therefore they can be emitted with higher energy with their distribution resembling the Fermi-Dirac distribution inside the solid at the appropriate temperature.

![FIG. 7.2. The simulated total energy distributions for field-emitted electrons; a to e curves represent the emission at temperatures: 0, 50, 100, 273, 295 K; the energy is measured from the vacuum level, the calculations were performed for silver $\phi = 4.5$ eV.](image)

If the applied potential is of optical origin then the dc interpretation of the tunnelling model needs to be re-examined. The peak value of the laser pulse can be strong enough to suppress the potential barrier at the metal interface and to make the
tunnelling process possible (see case C FIG. 7.1). If the change in electric field is relatively slow, as it is in the case of infrared lasers, there might be enough time for electrons to tunnel through the suppressed barrier. This problem was first solved by Keldysh [21] and afterwards re-examined by others [22], [1, 23-25].

**FIG. 7.3.** Solid line: the ionisation rate as a function of the optical field phase \( \varphi = \omega t + \varphi_0 \) at the beginning of the acceleration process e.g. \( t = 0 \), calculated with the ADK theory; dashed line: the electron’s field momentum as a function of the optical field phase; the optical field was assumed to be in a form: \( E_x = E_0 \cos(\varphi) \) and \( E_0 = 3 \times 10^{10} \) V/m, the ionisation potential was chosen to be the silver work function \( \phi_{Ag} = 4.5 \) eV.

The simplest tunnelling model in the presence of the optical fields is the barrier suspension ionisation (BSI) model [26]. In this model the optical field is approximated with a static electric field that suppresses the ionisation potential of an atom or ion. It is assumed that the emission occurs only at the peak of the electric field. Later, the BSI model was developed to incorporate the electron dynamics inside the atom or ion during the ionisation process. The most significant are Keldysh [21], Ammosov-Delone-Krainow (ADK) [24, 27], and Keldysh-Faisal-Reiss (KFR) [22, 23]. All these formulae concentrate on different atoms that are ionised, from simple hydrogen like molecules [21] to more complex multienergy...
levels ions [22]. For brevity, only the simplified ADK formula will be recalled here [24, 27]:

$$\eta_{ADK}(\phi) = A\phi \left( \frac{\phi^{1.5}}{|E(\phi)|} \right)^{B} \exp \left( -\frac{2(2\phi)^{1.5}}{3|E(\phi)|} \right),$$

(7.4)

where $A$ and $B$ are constants dependent on the electronic structure of the ionised species, $\phi$ is the ionisation potential or the work function, and $E(\phi)$ is the electric field expressed here as a function of the phase of the optical field $\phi = \omega t_0 + \phi_0$.

The above formula provides the ionisation rate for complex atoms in an ac electric field. The initial state of an electron is described by an effective quantum number and the angular and magnetic quantum numbers, which determine the constants $A$ and $B$. The ionisation rate will depend on the exact knowledge of the electronic structure of an ion or atom to be ionised.

In the experiment presented here thin gold and silver metal films were used. The electronic structure of bulk gold and silver crystals is known. However, the exact structure of thin films depends crucially on the conditions of the evaporation process and sample storing environment [28, 29]. The work function values used in the calculations were measured for clean and perfectly flat metal surfaces. The metal surfaces were exposed to air before the emission spectra were measured. Therefore, it should be expected that the experimental values of the work function were different from the tabulated ones. Moreover, the electronic structure of the thin film in the presence of SPs is different from the bulk material structure hence the value of the $A$ and $B$ constants in the equation (7.4) can only be estimated. Although each of the emission theories proposed in the recent literature describe the emission dynamics in unique ways, the significant conclusion from the Keldysh-type theories is that the emission process in the tunnelling regime is dependent on the phase of the electric field that is applied to the atom or ion (see FIG. 7.3). The liberation of an electron can occur for a finite range of the phase of the optical pulse during one cycle.
7.1.3. MULTIPHOTON OR TUNNEL IONISATION

Keldysh [21] defined the adiabacity parameter $\gamma$ to distinguish the boundary between multiphoton and tunnelling emission. This parameter is the ratio of the electron tunnelling time to the laser period. In its simplest form it can be defined as: [25, 26, 30]

$$\gamma = \sqrt{\frac{\phi}{2U_p}},$$  \hspace{1cm} (7.5)

where $U_p$ is the ponderomotive energy defined by equation (5.18). Electron spectroscopy [25, 30, 31] measurements showed that when $\gamma \ll 1$, the majority of electrons are liberated through the tunnelling process, whereas for $\gamma >> 1$, the multiphoton process dominates. However, experimental results were reported where tunnelling, rather then the MPI, was observed for $\gamma$ in the range of 1 to 8. [30, 32]

**FIG. 7.4.** The schematic drawing of the oscillation of the electric field vector during one optical pulse at $z = 0$; the peak intensity of the pulse is $E_0$; the dashed line marks the boundary between the regions where the Keldysh adiabacity parameter $\gamma$ is less or more than unity as indicated; the inset shows an enlarged section of the electric field strength oscillations and the sign of the derivative of $E$ vector with respect to time ($dE/dt$).
In the experiments presented in this work the typical peak value of the electric field was \( E_0 = 3 \times 10^{10} \text{ V/m} \) at 800 nm. The commonly accepted values of the work functions for silver and gold are 4.5 eV and 5.3 eV respectively [33]. Hence, from the equation (7.5) the Keldysh parameter is equal to \( \gamma_{Ag} = 0.56 \) and \( \gamma_{Au} = 0.61 \). A study of the electron emission from gold films in the presence of SPs [30] suggests that the tunnel ionisation process should be expected in this case. However, in the experiments presented in this work the electron emission was also observed for lower values of strength of the electric field than the quoted maximum value. This suggests that the photoemission conditions are in the transition region between the multiphoton and the tunnelling formalisms. A complex emission mechanism such as multiphoton assisted tunnel ionisation can be considered. This can be the subject of future work.

7.2. FIELD MOMENTUM

After the ionisation process, the electron’s dynamics is determined by calculating the Lorentz force that acts on the electron in the external electric field. Firstly, it is the net energy that the electron gains in the evanescent field due to the ponderomotive force that arises from changes of the field intensity across the electron path. This was the main subject of Chapter 5 Additionally two other mechanisms give rise to the electron’s final kinetic energy. The initial kinetic momentum is inherited from the SPs momentum at the metal surface and the field momentum is due to the electric field phase at the moment of ionisation. [27, 34-36].

The canonical momentum of an electron in the optical field can be expressed as: [37]

\[
\vec{P}(r,t) = \vec{p}_{\text{kin}}(r,t) + \frac{e}{c} \vec{A}(r,t),
\]

where \( \vec{p}_{\text{kin}}(r,t) \) is the residual kinetic momentum from SPs and \( \vec{A}(t) \) is the vector potential of the evanescent field. The electric field vector oscillates in the plane of
incidence in the \( z \) direction perpendicular to the metal surface (see FIG 3.1). Only the \( z \) component of \( \vec{A} \), \( \vec{p}_{\text{kin}}(r,t) \) and \( \vec{P} \) will be considered throughout the calculations.

The field momentum of an electron is gained from the electric field outside of the metal film at the moment of liberation. The kinetic momentum is inherited from the electron dynamics inside the metal film. Since the kinetic momentum originates from the SP momentum, it does not depend explicitly on the phase of the optical field outside the metal. Therefore, for the purpose of this discussion, it can be ignored. The field momentum as a function of the phase of the electric field can be re-written as:

\[
p_{f,z}(\varphi) = \frac{e}{c} A_z(\varphi), \quad \varphi = \omega t + \varphi_0.
\]  

The vector potential is related to the electric field vector as:

\[
E_z(\varphi) = -\frac{\partial \psi}{\partial z} - \frac{1}{c} \frac{\partial A_z(\varphi)}{\partial t},
\]

where \( \psi \) is the scalar potential, which depends only on the position of the electron. At the moment of ionisation, \( t = 0 \) and \( z = 0 \) hence \( \varphi = \varphi_0 \) and \( \frac{\partial \psi}{\partial z} = \text{const.} \). To simplify calculations it will be set to zero. The electric field vector is defined as \( E_z = E_0 \cos(\varphi) \) therefore the vector potential can be expressed as:

\[
A_z(\varphi) = -\frac{cE_0}{\omega} \sin(\varphi).
\]  

By inserting equation (7.9) back into equation (7.7), one gets:

\[
p_{f,Z}(\varphi) = -\frac{eE_0}{\omega} \sin(\varphi).
\]  

The field momentum is out of phase with the driving electric field by 90 degrees. It’s value is zero only at the peak of the optical cycle, as shown in FIG. 7.3. Only the BSI theory predicts that the liberation of an electron occurs at the peak of the optical cycle. In this case the field momentum would not contribute to the final kinetic
energy. However, the Keldysh type theories and the ATI allow the electron to be liberated at the off peak values of the optical cycle, where the field momentum is non-zero. Therefore, one of the initial conditions of the electron interaction in the evanescent field is determined by the phase of the optical field at the moment of the liberation.

Indeed, the dependence of the initial phase at the time of generation was observed in the numerical simulations, presented in Chapter 6. The electron’s final energy was calculated only in the case when the electron did not re-enter the metal e.g. the electron position had to be always a positive value. This happened when the initial phase was in the range of $\varphi \in [\pi, 2\pi]$, (see FIG. 6.12). The electron gained maximum energy if the initial phase was set to 270 deg. This observation is consistent with equation (7.10). Whenever the value of the field momentum vector is positive, then the electron’s position is positive. Consequently, the electron should gain its maximum energy if $\sin(\varphi) = -1$, e.g. $\varphi = 270$ degrees.

### 7.3. FINAL KINETIC ENERGY DUE TO INITIAL PHASE

The general expression for the rate of kinetic energy change of a particle that interacts with the electromagnetic field is given by: [37]

$$\frac{dU_{\text{kin}}}{dt} = \vec{v} \cdot \frac{d\vec{p}}{dt},$$

(7.11)

The above equation can be rewritten in terms of the phase of the electric field $\varphi = \omega t + \varphi_0$. The velocity of the particle will be expressed as a function of its position:

$$\frac{\partial U_{\text{kin}}}{\partial \varphi} = \frac{1}{\omega} \frac{\partial^2 \vec{r}(\varphi)}{\partial \varphi^2} \cdot \frac{\partial \vec{p}(\varphi)}{\partial \varphi},$$

(7.12)

where $\omega$ is the angular frequency of the driving optical field.

In the case discussed in this chapter the kinetic energy is the energy $U_{\text{field}}$ gained from the field momentum $p_f$, the so-called quiver energy. In the evanescent field of
the SPs both vectors, momentum and position, have only components in the z direction since the electric field vector $E$ oscillates only in this direction. The two vectors can be only in phase or out of phase by 180 deg therefore $\cos\left(\vec{p}_j, \vec{r}\right) = \pm 1$. However, the electron can start its acceleration process only when momentum and position vectors are in phase $\cos\left(\vec{p}_j, \vec{r}\right) = 1$. Combining equations (7.10), (7.12) and (5.8) at $t_0 = 0$ yields:

$$U_{\text{field}} = -\frac{e^2 E_0^2}{4 \omega^2} \cos(2\varphi) \exp\left(-\frac{z}{\lambda_{ov}}\right).$$

(7.13)

The initial momentum is acquired at $z = 0$, therefore $\exp\left(-\frac{z}{\lambda_{ov}}\right) = 1$. Therefore, the kinetic energy due to the initial phase of the optical field is:

$$U_{\text{field}} = -\frac{e^2 E_0^2}{4 \omega^2} \cos(2\varphi),$$

(7.14)

and the initial phase is in the range of $\varphi \in [\pi + 2k\pi, 2\pi + 2k\pi]$, $k \in \{0, N\}$, see equation (7.10). The energy due to the field momentum can be varied in a range from $-\frac{e^2 E_0^2}{4 \omega^2}$ to $\frac{e^2 E_0^2}{4 \omega^2}$. The electron can gain the maximum kinetic energy from the field momentum if the initial phase of the optical field is $\frac{3\pi}{2}$ at the moment of its liberation. Indeed, the same result was obtained from numerical calculations presented in Chapter 6.

7.4. TOTAL ENERGY

The total electron’s energy $U_{\text{total}}$ is sum of the energy gained due to the interaction with the ponderomotive force and the energy gained from the initial field momentum. Combining equations (5.18) and (7.14) the total energy can be expressed as:
\[ U_{\text{total}} = U_{\text{ponderomotive}} + U_{\text{field}} \]
\[ = U_{\text{ponderomotive}} \left( 1 - \cos(2\varphi) \right), \quad (7.15) \]

and the initial phase is in the range of \( \varphi \in [\pi + 2k\pi, 2\pi + 2k\pi], \quad k \in \{0, N\}. \) \( \cos(2\varphi) \) changes in the range of a full cycle of the cosine function. Therefore, the total energy of an electron can be in the range from 0 to \( 2U_{\text{ponderomotive}}. \) Indeed, this was observed in the numerical calculations when the initial phase was varied freely in the range of \( \varphi \in [\pi, 2\pi], \) and the maximum energy was always gained when the initial phase was set to \( \frac{3\pi}{2}. \)

In the experiments presented here, the laser pulse reached the metal surface with a random phase. Furthermore, the experimental energy spectra are averaged over several laser pulses. The averaging technique was used to maximise the signal-to-noise ratio, as was explained in Chapter 2.

If the electron is liberated in the narrow range of the initial phase \( \varphi \in (\alpha, \beta), \) \( \alpha < \beta \) and \( \alpha, \beta \in [\pi + 2k\pi, 2\pi + 2k\pi], \quad k \in \{0, N\} \) \( U_{\text{field}} \) will contribute to the final kinetic energy of an electron. The average value of the \( U_{\text{field}} \) is:

\[ U_{\text{field, av}} = \frac{e^2 E_0^2}{8m\omega^2} \frac{\cos(2\alpha) - \cos(2\beta)}{\beta - \alpha}, \quad (7.16) \]

and the final kinetic energy of an electron takes the form:

\[ U_{\text{total}} = U_{\text{ponderomotive}} \left( 1 - \frac{\cos(2\alpha) - \cos(2\beta)}{2(\beta - \alpha)} \right), \quad (7.17) \]

where \( \alpha \) is the phase of the optical field at which the liberation process starts and \( \beta \) is the value of the phase at which the liberation process stops.

### 7.5. Angular Distribution of Photoelectrons

The presence of the additional force in the evanescent wave acceleration process can be responsible for the non-symmetrical distribution of emitted electrons and the spread of energies of electrons especially at the cut-off energy point.
It was observed that in the plane of polarisation of the laser beam the majority of electrons were emitted perpendicularly to the metal film. In the direction out of this plane (perpendicular) there was no distinguishable distribution. Refer to FIG. 4.5. This observation was not consistent with the multiphoton emission process, which states that electrons emerge out of the metal film with randomised momentum (see section 4.6). However, it does confirm that electrons are accelerated in the evanescent wave acceleration process. When the electrons are accelerated by the ponderomotive force, their acceleration depends on the intensity change of the accelerating field but not the direction of oscillation of the electric field vector. If a Gaussian profile of the incident beam is considered then the distribution of emitted electrons should be symmetrical in all directions perpendicular to the metal surface. However, the quiver energy that results from the electron initial conditions defined by the equation (7.14) acts only in the direction of polarisation of the incidence laser beam. Therefore its effect can be only visible in the plane of polarisation of the laser beam, leaving the uniform distribution of electrons in all other directions. Indeed, the non-symmetric electron distribution accelerated in the optical field has already been reported. [27, 34-36]

The theoretically calculated probability distribution functions (see FIG. 5.7, 5.9 and 5.11) revealed that due to evanescent wave acceleration the spectrum should have sharp energy cut-off. This was never observed in the experiments even with the best possible laser beam profile. Equation (7.17) suggests that one should expect that some electrons acquire an additional momentum at the beginning of the acceleration process that might be responsible for their further acceleration.

In this chapter it was shown that the electron’s energy, originating from the interaction with the SPs electric field, consist of two main components: the ponderomotive energy and the quiver energy. The ponderomotive energy is gained as a result of interaction with the ponderomotive force, described in detail in Chapter 5. It is related to the intensity gradient of the driving optical field, and it expels electrons from the high intensity regions to the low intensity regions. Hence, its value does not depend explicitly on the direction of the electric field vector of the driving
optical field but only on its absolute value. The quiver energy is gained at the beginning of the interaction and is calculated from the field momentum at the moment of the electron’s liberation from the solid. It depends on the phase of the optical field at the beginning of the acceleration process, therefore it is directly related to the electric field vector direction and its magnitude at that moment. The effects of the quiver motion can be observed only in the direction of polarisation of the driving EM wave. Furthermore the non-symmetric electron distribution was observed in the experiments presented in this work as shown in FIG 4.5. A more sensitive experimental set-up is required to investigate the anisotropy of the electron distribution in the SPs evanescent field in greater detail. However, this result confirms that a non-symmetrical force interacts with the electrons that can be associated with the ponderomotive force.

7.6. REFERENCES


CHAPTER 8

ELECTRON EMISSION FROM ROUGH METAL SURFACES

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In previous chapters, a theory of surface-plasmon (SP) excitation at perfectly flat metal surfaces was presented. It was shown that it is possible to efficiently liberate and accelerate electrons up to a few keV in the evanescent optical field that is enhanced in the presence of the SPs. In this chapter, attention will be focused on a method that allows further enhancement of the optical field at the metal surface. First, the process of exciting localised SPs in small metal particles deposited at a plane surface will be discussed. Next, it will be shown that the electric field is enhanced near a surface of high curvature due to the lightning-rod effect. Finally, the experimental results of electron emission from rough metal surfaces manufactured in two different ways will be analysed and compared with previously presented electron emission spectra in Chapter 6.

8.1. ELECTRIC-FIELD ENHANCEMENT AT ROUGH METAL SURFACES

The enhancement of the local electric field in the presence of an EM field at the surface of small metal particles originates from two mechanisms: the local SP resonance [1-6] and the corona or the “lightning-rod” effect. [2-5, 7, 8] The first mechanism has a dynamic character and arises from the excitation of coherent plasma oscillations that are localised within one or more protrusions at the metal surface. The second enhancement mechanism has a static character and arises only from the fact that the electric field lines tend to concentrate near surfaces of a high curvature. Hence it is a purely geometrical factor. The total enhancement of the local
field at the surface of small metallic protrusion is a product of the dynamic and static enhancement mechanisms: [9, 10]

\[ \gamma_{total} = \gamma_{SP} \gamma_{LR}. \]  

(8.1)

Many models have been proposed describing the interaction of small metallic particles with EM waves. The fundamental work was published in 1908 by Gustav Mie. There he described interaction of a perfectly spherical particle with an EM wave to explain various colours of small colloidal particles of gold in water. The detailed review of the Mie theory is given by Bohern and Huffman in [11]. Later, it was further extended to the interaction of arbitrarily shaped small particles with EM radiation. [2, 7, 10, 12-14] Mie theory was applied to many diverse branches of physics such as meteorology or astrophysics to explain scattering properties of small particles such as raindrops creating rainbows and glories [15, 16] or interstellar dust. [17] The Mie formalism was also used to analyse large electric field enhancements in surface science. It was applied to explain large field enhancements in Surface Enhanced Raman Spectroscopy (SERS) [2, 18], in aperture-less near-field optical microscopy. [9, 19] and in high order nonlinear harmonic generation at metal surfaces. [10, 14]

8. 1. 1. LOCALISED SURFACE PLASMONS

The theory of surface plasmon (SP) modes on perfectly flat metal surfaces was discussed in Chapter 3. However, the real surface of a solid is characterised by a certain degree of roughness that typically ranges in 10-100 nm. [20, 21] The conditions of the SPs excitation at the real surface are different to those calculated for an ideal model. The perfectly flat metal surface is a good first approximation of a real system. However, if the roughness of the surface is of the order of a wavelength of the exciting EM field or higher the theory proposed in Chapter 3 needs to be revised. The sensitivity of the SPs excitation condition to the quality of the metal surface has been employed in the SP spectroscopy technique to characterise the smoothness of the surface of different solids. [22]
A real metal surface can be modelled as a collection of small randomly distributed protrusions settled on a flat plane that is surrounded by another dielectric medium. For each medium the protrusions, the plane and their environment can be characterised with a frequency dependent dielectric function, as shown in FIG. 8.1. It is possible to solve the boundary conditions (see equations 3.6 and 3.7) in a one particle system and afterwards extend it to the whole assembly. However, it is very tedious as a spherical set of coordinates is required. Exact solutions were derived by several authors and can be found in the work of Bohren and Huffman [11] or Wokaun [5, 11]. In this work, however, only the final result will be discussed.

FIG. 8.1. A schematic drawing of hemispheroids on a flat substrate; the localisation directions of the SPs within one particle are shown in the enlarged section; the EM field is evanescent in both x and z directions as a result of excitation of localised SPs; each medium is characterised with dielectric functions $\varepsilon_{s}(\omega)$, $\varepsilon_{m}(\omega)$, $\varepsilon(\omega) = 1$ for the substrate, the metal particle and the surroundings (chosen to be a vacuum).

Consider a plane TM EM wave incident at a metal hemispheroid deposited on a substrate and exposed to vacuum. The dipole approximation imposes that the hemispheroid needs to be small with respect to the wavelength of the exciting EM field so that the electric field induced inside it is uniform over its volume. The EM wave can couple efficiently to the SP modes only if one or more components of its wavevector match that of SPs modes inside the particle. When the SP modes in thin metal film were considered, this condition was fulfilled only in the direction perpendicular to the thin film surface, here chosen as the z direction, $k_z = k_{SPs}$ see
Chapter 3, FIG 3.4. Therefore, the SP modes were localised only in the direction perpendicular to the surface, and delocalised in the plane of the film. The coupling conditions were fulfilled at the ATR angle that was unique for a given configuration (see Fig 3.5).

In a small particle, in contrast to the plane metal film, not only the \( k_z \) wavevector can couple efficiently to the SP modes but also the \( k_x \) vector of the TM EM wave. Therefore, the SP modes are localised in both directions (see FIG. 8.1). In metal spheres, SPs can be excited at any angle of incidence of the EM wave since there is no preferred direction of SP excitation due to the symmetry of the system. If the metal particle is of a lower symmetry, there exists a preferential axis where the excitation of localised SPs leads to the highest enhancement of the local field at the surface of the particle. A good approximation of a protrusion is a hemispheroid as presented in FIG. 8.2. The maximum local field enhancement is obtained at the tip of the particle when the electric field vector oscillates along its longer axis. Furthermore, in this arrangement the local SP enhancement can coincide with the enhancement due to the corona effect.

**FIG. 8.2.** Schematic drawing of a hemispheroid deposited on a flat surface; \( \varepsilon_m(\omega) \), \( \varepsilon_s(\omega) \) and \( \varepsilon(\omega) \) are the dielectric functions of the particle, substrate and the environment respectively; \( a \) and \( b \) are the two semi-axes of the particle; if the incoming electric field vector \( \vec{E}_0 \) oscillates along the longer axis, then the highest field enhancement due to SPs is expected at the tip of the particle.
A simple 2D model can be constructed to calculate the field enhancement at the tip of the hemispheroid due to its symmetry along the longer axis. The total field enhancement at the tip of the particle shown in FIG. 8.2 can be calculated as a ratio of the amplitude of an incoming electric field $E_0(\omega)$ and the amplitude of the local field at the tip of the particle $E_{loc}(\omega)$,

$$\gamma_{total} = \left| \frac{E_{loc}(\omega)}{E_0(\omega)} \right|.$$  \hfill (8.2)

If the electric field vector oscillates along the longer semi-axis of the hemispheroid the total enhancement of the local field may be expressed as: [5, 12, 19, 23]

$$\gamma_{total} = \left| \frac{\varepsilon_m(\omega)}{1+(\varepsilon_m(\omega)-1)A} \right|,$$  \hfill (8.3)

where $A$ is a size dependent depolarisation constant and $\varepsilon_m(\omega)$ is the particle dielectric function that is frequency dependent and is in general a complex number. To derive the above equation, it was assumed that the particle is suspended in vacuum and the interaction with the substrate is neglected.

The SPs resonance occurs in the particle when the real part of the denominator in equation (8.3) is zero: [5, 12]

$$\text{Re}\left[1+(\varepsilon_m(\omega)-1)A\right] = 0.$$  \hfill (8.4)

Assume initially that the $A$ constant is purely a geometrical factor that depends on the size of the hemispheroid. Therefore, at a given frequency for the chosen metal, the SP resonance will depend on the shape of the particle. If dynamic depolarisation effects, such as surface scattering or radiation damping, are taken into consideration then the form of the $A$ constant becomes more complicated. [5] The dipole approximation requires that the particle needs to be small to provide a uniform electric field distribution inside the hemispheroid in order to excite the localised SPs. However, with decreasing size of the particle, the damping due to the wall-collisional effect becomes prominent. [5] On the other hand, if a large particle is considered, the
dipole approximation does not hold and higher order multipoles can be excited inside the hemispheroid. Then the dynamic depolarisation effects such as radiative damping will dominate. [3, 5, 10]. There were noticeable variations between the observed enhancement and the calculated one within the electrostatic approximation if the longer semi-axis of the particle is: [12]

$$a \geq 0.02\lambda. \quad (8.5)$$

For such large particles the Lorentz-Mie theory needs to be applied. A numerical solution exists and the substantial review is given by Barber et al in [12]. However many authors reported using the electrostatic approximation for the particle size of the order of a wavelength that yields good results. [5]

8.1.2. LIGHTNING-ROD EFFECT

The electric field enhancement due to the lightning rod effect arises only from the geometry of the object that interacts with an electric field. Using Gauss’s law it is possible to show that electric field lines tend to concentrate around conductors of high curvature. Therefore, the density of a local electric field is higher close to the tip of the protrusion at the metal surface compared to that close to the perfectly flat plane. The magnitude of the enhancement due to the corona effect will depend only on the exact shape of the protrusion.

Consider again a hemispheroid characterised by the complex dielectric function $\varepsilon_m(\omega)$ and interacting with an EM field (see FIG. 8.2). The lightning rod effect is a static type of enhancement and does not depend on the frequency of the driving field but only on its amplitude. According to Drude theory, in the quasistatic limit, where the frequency of the driving field tends to zero, the imaginary part of the dielectric constant of a free electron metal also tends to zero, $\varepsilon(\omega) \rightarrow \text{Re}[\varepsilon(\omega)]$. In this case, the enhancement calculated with equation (8.3) contributes only to the enhancement due to the lightning rod effect and is frequency independent. [4, 6]

$$\gamma_{LR} = \left| \frac{\varepsilon_m(0)}{1 + (\varepsilon_m(0) - 1)\lambda} \right| \quad (8.6)$$
If a spherical particle is considered, namely \( a = b \), the \( A \) constant is equal to 1/3. [5, 6] When the particle is elongated, \( A \) depends on the ratio of its two semi-axes \( r = \frac{a}{b} \). In the quasistatic regime, the depolarisation constant for a hemispheroid can be calculated using standard tabulated functions and it takes the form: [4, 6, 23]

\[
A(r) = \frac{1}{2r^2} \int_{0}^{\infty} \frac{ds}{\sqrt{(s+1)(s+r^{-2})}}.
\]  

(8.7)

The depolarisation constant defined by the above equation depends only on the ratio of the two semi-axes, therefore it is shape but not size dependent. In the same way the enhancement due to the lightning rod effect will depend only on the shape of the particle. This is correct since the change of size of the metal particle does not change the boundary conditions in the electrostatic problem.

**FIG. 8.3.** The calculated electric field enhancement coefficient at the tip of the hemispherical silver particle interacting with a \( \lambda = 800 \text{nm} \) EM wave as a function of a ratio of its two semi-axes \( r = \frac{a}{b} \); the dashed-dot line represents the total field enhancement, the solid line is the enhancement due to the lightning rod effect only and the dashed line is the SP contribution to the total enhancement; the electric field vector was chosen to oscillate along the longer semi-axis of the hemispheroid as shown in FIG. 8.2.
If the dynamic depolarisation processes are neglected the total field enhancement $\gamma_{total}$ can be estimated by inserting equation (8.7) into (8.3) and using the particle’s dielectric function for a given wavelength. [3] The enhancement due to the lightning rod effect $\gamma_{LR}$ can be calculated in the same manner in the limit when $\varepsilon_m(\omega) = \text{Re}[\varepsilon_m(\omega)]$. Additionally, the enhancement due to SP excitation $\gamma_{SP}$ can be recalculated from the formula (8.1). FIG. 8.3 presents the calculated total electric field enhancement and the contributions of the SPs excitation and the corona effect enhancement in a silver hemispherical protrusion. All the calculations are valid within the dipole approximation.

As expected, the enhancement due to the corona effect is a monotonically rising function of the axes ratio $r$ as the tip’s curvature at the tip increases with increasing $r$. The enhancement due to SPs excitation presents different dynamics. It is sensitive to the exact value of the dielectric function of the metal particle at a given wavelength. For a silver hemispheroid it reaches its maximum value of 38 when the axes ratio $r$ is close to 7 and becomes small again when $r$ increases further. Similarly, the overall enhancement follows the same dynamics. Its maximum is nearly at the same value of the hemispheroid axes ratio as $\gamma_{SP}$ and it reaches the maximum value of nearly 570. Indeed such large field enhancement factors have been reported in various SERS experiments where silver was an active surface. For a review see the work of Moskovitz [18] and references therein.

8.2. DEPENDENCE ON INCIDENCE ANGLE OF INCOMING LASER BEAM

FIG. 8.4 presents energy spectra of electrons emitted from a rough gold film. A 50 nm thick metal film was deposited onto the hypotenuse side of a right angle prism. The metal particles were prepared by heating the film to a temperature below the melting point of gold and then cooled rapidly as described in detail in Chapter 2. Such a treatment resulted in a film of almost transparent appearance with conductivity close to zero. The size of droplets was difficult to distinguish with an optical microscope. This suggests that that the particle size was less than a wavelength from the visible spectrum.
The experimental set-up was identical with that used in experiments involving flat metal surfaces in the Kratchmann configuration, as described previously in Chapter 2. A collimated 5 mm, $\lambda = 800$ nm laser beam was used throughout the experiments. The pulse duration was constant and set to 150 fs at 1 kHz repetition rate. The intensity of the laser beam at the particle layer was varied with the combination of a half-wave retardation plate and a polarising cube, as explained in Chapter 2. First, the incidence angle at the metal film was set to $45^\circ$ and later it was changed in steps by several degrees in the range form $40.4^\circ$ to $46.5^\circ$. At all times, the same area of the metal film was irradiated.

**FIG. 8.4.** The energy spectra of electron emitted from rough gold surfaces; graphs from (a) to (d) correspond to the emission of the electrons at different angles of incidence $45$ deg, $46.5$, $43.5$, $40.5$ deg respectively; each data set (1 - 3) for each graph corresponds to different intensity of an EM wave 1.7, 6.8, 11.9 GW/cm$^2$ respectively.
Electron emission from a rough gold film was observed for angles of incidence as far away from the SPs resonance angle as 4.4 deg. This result is in contrast with the spectra of electron emission from flat metal films that were observed only at the SP resonance angle of 42.1° for gold films. The emission intensity is comparable with that from the flat metal films (see FIG 6.2 and 6.5). Similar emission intensity and distribution of electron energies indicate that the acceleration process occurs in the presence of the SP evanescent field. The absence of any dependence on the angle of incidence of the laser beam indicates that localised SPs are excited in the metal particles.

The method used to produce rough metal surfaces does not allow control over the size and arrangement of metal particles at the surface. Therefore, an assembly of particles with better defined size and shape needs to be created. However, this experiment proved that the use of a right angle prism as a substrate is unnecessary since the emission occurs at different resonance conditions than that provided by the Kretchmann configuration. Moreover, the use of flat, thin substrate will make the measurements of the particle dimensions with the Atomic Force Microscopy (AFM) possible.

8.3. EMISSION FROM NANO-PYRAMIDS

An experiment leading to direct comparison of the emission from a flat metal film and a particle assembly was performed. A silver layer was evaporated onto hypotenuse side of a right angle prism. The side length of the prism was 40 mm. The sample was mounted onto the vacuum chamber in the standard way as described in Chapters 2 and 6. Nano-pyramid arrays were produced using the method developed by the Van Duyne group. [24-27] An AFM image of the nano-pyramid assembly is presented in FIG. 8.5. This technique was described in detail in Chapter 2. The nano-pyramid assembly was deposited onto an ITO coated flat glass sample to assure the transparency and a conductive substrate (for more details see Chapter 2). The sample was mounted on the standard glass vacuum window at the vacuum side of the time-of-flight tube. The nano-pyramid assembly directly faced the interior of the chamber. The $\lambda = 800$ nm laser beam entered the vacuum window from the outside and reached the particle array at an angle close to the SP resonance angle in the
Kretschmann configuration. The laser beam was collimated and its diameter was 1.5 cm. The reason for such a large beam diameter was twofold. Firstly, it provided a large interaction area for the laser beam with the nano-pyramid array metal film leading to a larger number of emitted electrons. Secondly, the laser beam was astigmatic and therefore it was difficult to decrease its size using a standard combination of two lenses in the telescope arrangement. It was possible to improve the spatial beam profile by simple tilting one of the lenses in the telescope arrangement. This, however, led to a poor temporal profile of the beam as the wavefront travelled through different glass thicknesses in the lens. Attempts at beam size reduction always resulted in very poor quality of spatial or temporal beam profile. The laser pulse duration was set to 50 fs and the incident power was attenuated with a set of reflective neutral density filters. Such an arrangement was chosen to provide the same laser beam size in both arrangements in order to compare the emission spectra.

**FIG. 8.5.** AFM 3D image of silver nano-pyramids on ITO conductive glass; the scan is over 3 µm square; the maximum height of the metal tips is 100 nm.
FIG. 8.6. Electron emission spectra from a flat silver film (a) and nano-pyramid assembly (b); a set of curves on each graph from 1 to 4 corresponds to different laser beam intensities at the emitter of 30, 65, 100 and 175 GW/cm² respectively.

FIG. 8.6 compares the electron emission spectra from a flat silver film and the nano-pyramid assembly. The spectra were recorded for a range of laser power intensities as indicated in the figure caption. The energy spectrum of the emission from the nano-pyramids is slightly narrower compared to that recorded for the flat
metal film. However, this discrepancy is very small and can be attributed to lower effective laser beam intensity at the nano-pyramid assembly. The laser beam had to cross three interfaces: air/vacuum-window/vacuum/glass substrate before it reached the target. This contributed to the power loss due to reflection at these interfaces.

![Graph showing the number of emitted electrons per bunch from flat surfaces (squares) and nano-pyramid layer (circles)](image)

**FIG. 8.7.** The number of emitted electrons per bunch emitted from flat surfaces (squares) and nano-pyramid layer (circles); the data presented in FIG. 8.6 were used for calculations; the fitted solid line has a slope of 6.

The dependence of the total number of emitted electrons on the incidence laser power presents very similar dynamics for the emission from the flat surface and the nano-pyramid assembly. However, there are some differences compared to the power dependence presented in FIG. 4.2 for the gold flat surface. The data presented above were recorded at a 50 fs duration time of the laser pulse. Therefore, the interaction of an electron with the evanescent field was not long enough to provide a full conversion of the energy stored in the evanescent optical field to the electron’s kinetic energy, due to the lack of the characteristic saturation plateau at high laser intensities.
Theory and experiment presented in this chapter show that localised SPs can be excited in a small metal particle that can enhance the electric field of the incoming laser field. The enhancement mechanism in a small metal particle is twofold, the SP enhancement and the “lightning-rod” effect. In the experiments presented in this work the magnitude of the field enhancement is comparable with the enhancement observed at flat metal surfaces. It is not surprising since the ratio of the semi-axes of the metal particles used here was close to 1 and therefore not matching the localised SP resonance for a small particle as presented in FIG. 8.3. However, it was demonstrated that there is no dependence of the electron emission on the angle of incidence as was the case for the flat metal film in the Kretschmann configuration (see FIG. 4.1). This is a very important result since it is possible now to accelerate electrons with the laser beam using 0° incidence angle. This will provide electrons across the interaction area generated in the same phase (for reference see FIG. 6.8).

8.4. REFERENCES

CHAPTER 9

CONCLUSIONS
CHAPTER 9

CONCLUSIONS

There is always ever-growing demand to build new and more sophisticated tools to investigate basic properties of matter. Recent advances in the femtosecond laser technology have created a great opportunity to create various time-resolved schemes that can probe molecular dynamics on a femtosecond time scale. Following this trend, this work presents the outline of a theory and experiments for the generation and acceleration of short electron bunches in an evanescent optical field.

The experiments and theory presented here aimed to develop a new time-resolved spectroscopy scheme that would be surface sensitive and use short electron bunches as a probe beam. As a result a novel technique of accelerating electrons in an optical field was developed and the analysis of electron excitation and emission in the presence of an intense ultrashort laser field was attempted. It is evident that the generated short electron bunches can be used as a tool to probe chemical reactions at metal surfaces. The results presented in this work were published in Nuclear Instruments & Methods In Physics Research Section A [1], Applied Physics Letters [2] and presented at two conferences: The 12th International Conference on Ultrafast Phenomena 2000, Charleston, North Carolina, USA [3] and The Gordon Conference on Vibrational Spectroscopy, 2000, Newport, Rhode Island, USA. Furthermore, the paper published in Applied Physics Letters was summarised in the Editor Choice section of Science [4].

To study the electron bunch dynamics emitted in the presence of surface plasmons (SPs) enhanced field the basic theory of surface plasmons was summarised. The wave equation of the SP waves, the conditions of excitation, and the experimental configurations where SPs can be excited were presented and discussed. Special emphasis was put on the surface field enhancement factor in the presence of a SP wave. On the basis of this investigation, a simple electron emitter was constructed. Thin (50 nm) gold or silver films were evaporated on a hypotenuse side of a right angle glass prism to form the Kretschmann configuration. Using a TM polarised 800 nm femtosecond laser beam an electron bunch was excited out of the
metal film and accelerated by the evanescent field away from the cathode. The electron beam was found to be directional in the plane of incidence of the laser beam whereas there was no preferred direction of emission out of the plane of incidence. The dependence of the averaged total photocurrent as a function of laser intensity was measured and found to be of an order of 9 when a 30-V retarding potential was applied to the sample. Furthermore, the time-of-flight analysis of the electron beam showed a wide distribution of electron energies and no evidence of individual peaks that could be attributed to the multiphoton process. It was demonstrated that the electrons could gain an energy as high as 0.4 keV.

The anisotropy of the electron distribution and the high electron energies (expanding from 0 to 0.4 keV) suggested that there is a force that could accelerate electrons away from the metal surface. The force was found to depend on changes of electromagnetic field intensity across the electron path and therefore it was identified as ponderomotive. It was found that electrons could gain high energy after interaction with an evanescent optical field at the metal/vacuum interface that is enhanced in the presence of SP excitation at the metal surface. Therefore, the equation of averaged motion in a fast oscillating evanescent field was solved within the classical limit. It was shown that the energy an electron can gain when it interacts with the evanescent field depends on the magnitude of the surface electric field and its frequency. It was shown that after a certain time of interaction of the electron bunch with the evanescent field, determined by the optical pulse duration, the whole electron bunch could be accelerated efficiently and gain the highest possible energy for a given arrangement.

The theoretical considerations were compared with the experimental results. Experiments investigating the energy distribution of the electron bunch as a function of the laser power density and the duration of the exciting laser pulse were performed. They were found to be in the good agreement with the evanescent wave acceleration theory. Furthermore, it was shown that it is possible to simulate experimental and analytical results using a numerical method for fixed values of the initial parameters. However, it become evident that the acceleration process depended not only on the value of the electric field vector of the accelerating field itself but also on its dynamics. Therefore, it was concluded that the emission process
of the electrons at the metal surface in the presence of the intense laser irradiation needed to be investigated in more detail.

An analysis of the emission process in the presence of intense ultrashort laser pulse was performed. It was found that the electron’s energy, originating from the interaction with the SP electric field, consists of two main components: the energy gained as a result of interaction with the ponderomotive force analysed before and the quiver energy. The quiver energy is gained at the beginning of the interaction process and can be calculated from the field momentum at the moment of the electron’s liberation from the solid. It was shown that the quiver force depended on the phase of the optical field at the beginning of the acceleration process. Therefore, it was directly related to the electric field vector direction and value at that moment. The effects of the electron interaction with the quiver force could be only observed in the direction of polarisation of the driving EM wave, explaining the anisotropy in the spatial distribution of the emitted electron beam and a wide spread of electron energies.

Several attempts were undertaken to demonstrate possible applications of the electron generation and acceleration technique presented in this work. The successful experiment involved fabricating an array of nano-pyramids on a flat conductive glass substrate. It was demonstrated in the theory that it is possible to obtain an electric field enhancement factor of an order of few hundreds in small metal particles. It was shown that the electric field enhancement arises from the excitation of localised SPs in metal particles and the “lightning-rod” effect. The experimental shape and dimensions of fabricated nano-pyramids did not meet the exact requirements of the theory. However, it was shown that the localised SPs can be excited in a small particle at any angle of incidence, therefore the experimental configuration can be simplified further. This can give the advantage of a uniform wavefront at the nano-pyramid array allowing an emission of coherent electron bunch.

The experiment that did not yield results that could be explained on a basis of the theory developed so far was the time-resolved experiment. It was anticipated that a time-resolved signal could be detected from a bare gold film or monolayer of molecules deposited on a metal surface. A time-resolved experiment was constructed where the electron beam was the probe beam and 400 nm (generated using second
harmonic generation in the beta barium borate (BBO) crystal) as a pump beam. After numerous attempts, no distinguishable signal was found. The possible causes of the failure of this experiment might be that the molecules deposited on the metal surface were not bond strongly enough to the metal. Hence, the laser beam incident on the metal surface caused their evaporation. In addition, the theory of electron emission in the presence of strong intense laser pulses states that the electron signal depends on the phase of the exciting laser pulse. The results of the interactions of the pump and the probe pulse could be obscured by the averaging procedure in the detection system. However, in the light of the theory presented in this work it is possible to build a more sophisticated time-resolved electron scheme where ultrashort electron pulses can be used.

Further study of this work should involve a detailed investigation of the electron emission process in the presence of intense ultrashort laser pulses. This should lead to a better understanding of the electron emission process in the scheme proposed in this work. This should also help in building more advance time-resolved experiments where an electron beam can be used as a probe. On the other hand, there is a need to develop a method of fabricating nano-pyramid arrays that could provide an even higher field enhancement factor and therefore would provide the possibility of a very simple way of generating high-energy ultrashort electron pulses. Indeed, it is already planned to use this idea in the TOPS facility at Strathclyde University in Dino Jaroszynski’s group.


APPENDIX
Below the C++ code is presented that was used to calculate position and energy of electron interacting with evanescent optical field.

DEFINITIONS OF GLOBAL PARAMETERS USED THROUGHOUT THE SIMULATIONS

```cpp
#define fs 1e-15
#define nmax 20000
#define CONV 1.386294361 //2 ln 2 conversion factor
#define SIMULATION_LENGTH 500 //Length of simulation in fs

static double mass=9.1093897e-31; //electron mass in kg
static double lambda=800*nm; //laser wavelength
static double lambdaEff=1000*nm; //1/e length of evanescent field
static double omega=2*PI*C/lambda; //frequency corresponding to 800nm
```

DEFINITIONS OF FUNCTIONS

GAUSSIAN PULSE ENVELOPE

The gaussian shape defined below is the envelope of the electric field vector of the 800 nm pulse (see equation 6.5). All variable t, shift, sigma are in seconds.

sigma is the FWHM of the pulse envelope;
shift is a time when the maximum of the gaussian pulse arrives at the surface and can be chosen arbitrary.

Usually the threshold value of the surface electric field strength was chosen at which the emission process can start and this determined the value of the shift parameter.

```cpp
doctor __fastcall Gauss(double t, double sigma, double shift)
{
    return exp(-CONV*pow((t-shift)/sigma,2));
}
```
MAXIMUM VALUE OF AN ARRAY

double __fastcall maximum(double array[nmax])
{
    int i;
    double max=array[0];
    for(i=1;i<nmax;i++)
    {
        if (array[i]>max) max=array[i];
    }
    return max;
}

EXPONENTIAL DECAY

A step function that is zero of the position of an electron is negative and decays exponentially if the electron position is positive (see equation 6.4).

double __fastcall evanescent(double x)
{
    if (x>=0) return exp(-x/lambdaEff);
    else return 0;
}

ALGORITHM 1

The algorithm below calculates the position (see equation 5.4) and kinetic energy (see equations (5.5) and (5.6)) and writes the data to the file. Note that using this algorithm the position of an electron can be either negative or positive.

xx, xm, x (m)- the position is the electrons at time t-2dt, t-dt and t;

v, velocity (m/s)- velocity of an electron;

force (N) – the value of the force;

Dt (fs) time step

t (fs) time;

F0 (V/m) - maximum value of the surface electric field (chosen arbitrarily)

FS (V/m) - the threshold value of the surface electric field when the emission is allowed (see FIG. 6.14) (chosen arbitrarily)

energy0 (J)– the initial energy of en electron (chosen arbitrarily);

Phi (degree) – initial phase id the gaussian pulse (chosen arbitrarily);

Nsteps - number of calculation steps
double xx,xm,x,v,force,t,Dt,F0,FS,energy0,velocity,sigma,shift,phi;
int i,NSteps;
FILE *fp;

velocity=sqrt(2*energy0/mass);           // initial velocity

// initial velocity

//zeros the variables

Dt=0.001*fs;                              // the time step

//calculation of the second position

xm=x-velocity*Dt;                               // calculates the second position

NSteps=SIMULATION_LENGTH*fs/Dt;

//calculates pulse shift for a given ES

shift=sigma*sqrt(0.5*fabs(log(FS/F0)/log(2)));   // calculates pulse shift for a given ES

fp=fopen("C:/users/justyna/position.dat","w");
for(i=0;i<NSteps;i++)
{
    fprintf(fp, "%lg, %lg \n",x*10^6 ,0.5*mass*v*v)
    t=i*Dt;                                   // Simulation time
    force=E*F0*cos(omega*t+phi)*exp(-x/lambdaEff)*Gauss(t,sigma,shift);    // (see equation 5.4)
    xx=2*xm-x+force*Dt*Dt/mass;             // (see equation (5.5))
    v=(xx-x)/(2*Dt);                       // (see equation (5.5))
    x=xm;                                   // Back up old position
    xm=xx;
}

fclose(fp);

ALGORITHM 2

The algorithm below uses the algorithm 1 and calculates the kinetic energy of electrons at the time \( t = N_{\text{steps}} \times D_{t} \) for a bunch of electrons that start their accelerations process at equal time intervals determined by the number of electrons used in simulation (see FIG. 6.14). Afterwards it calculates the statistics of the energy distribution (binning).

The variables used below have the same meaning as in Algorithm 1. The additional variables are:

t\(_{\text{generation}}\) – a value of time interval inside which the electron can start its acceleration process;

\( \text{max} \) – maximum energy that a electron obtained;

\( D_{j} \) – a value of an energy bin;

\( \text{energydata}[n] \) – an array where all the values of the kinetic energy corresponding to each electrons are stored;
spectra[nmax] – an array that stores information of the binned data on an energydata array.

```c
{ double xx, xm, x, v, force, t, Dt, tgeneration, F0, FS, energy0, velocity, sigma, shift, max, Dj, phi;
 int i, j, NSteps, NSteps1, m;
 FILE *fp;
 double energydata[nmax], spectra[nmaxx];

 velocity=sqrt(2*energy0/mass);
 t=v=x=xx=xm=0;
 Dt=0.001*fs;
 xm=x+velocity*Dt;
 NSteps=SIMULATION_LENGTH*fs/Dt;
 shift=sigma*sqrt(0.5*fabs(log(FS/F0)/log(2)));  //calculates Pulse shift for a given ES
 tgeneration=2*shift;  //time of electron generation
 j=0;
 fp=fopen("C:/users/justyna/spectra.dat","w");
 for(i=0;i<nmax;i++)  //initialises energy array
 {
     energydata[i]=-1;
 }
 for (j=0;j<nmax;j++)  //creates electrons
 {
     t=v=x=xx=xm=0;
     xm=x-velocity*Dt;
     t=j*tgeneration/nmax;
     NSteps1= ceil(NSteps-j*tgeneration/(nmax*Dt));  //number of steps of the jth electron
     if (F0*cos(omega*t+phi)*Gauss(t,sigma,shift)<FS) continue;  //starts simulation only when the current value of the surface electric field is greater then the threshold value FS
     for(i=0;i<NSteps1;i++)  //calculates electron final energy, step size is reduced once electron is generated at the later time
     {
         t=i*Dt+j*tgeneration/nmax;
         force=F0*cos(omega*t+phi)*E*evanescent(x)*Gauss(t,sigma,shift);
         xx=2*xm-x+force*Dt*Dt/mass;
         if(xx<0) break;  //quits this loop if position of an electron is negative
         v=(xx-x)/(2*Dt);
         x=xm;
         xm=xx;
     }
     if(xx<0) continue;  //skips to the end if the previous loop was broken
 }
```

energydata[j]=0.5*mass*v*v*J2eV;
}

max=maximum(energydata);       //finds the maximum energy value
for(j=0;j<ceil(max/Dj);j++)    //sorts energy data into bins
{
    m=0;
    for(i=0;i<nmax;i++)    //scans all energy data and picks up those that
        //fall into one bin
        {
            if(j*Dj<=energydata[i] && energydata[i]<(j+1)*Dj) m++;
        }
    if(m!=0) fprintf(fp, "%lg, %d
", Dj*(j+0.5), m);
}
fclose(fp);
}

Other variations of the above algorithm are possible. The final kinetic energy of an electron can be calculated with respect to: surface electric field, wavelength, initial energy of an electron and pulse duration. Those algorithms are straightforward.
APPENDIX 2

Below the Mathematica 3.0 code is presented that was used to calculate the energy distribution functions discussed in Chapter 5.

The function below calculates time that is needed to gain a certain energy by an electron (see equation (5.26)). Note that this function can yield results even for energy greater then the maximum possible energy that an electron can gain defined by equation (5.18), therefore the boundary conditions are needed. The variables used in the function below are:

- \( g \) is defined by equation (5.21);
- kinetic energy \( U \) is defined by equation (5.30);
- the initial kinetic energy is defined by equation (5.28).

\[
\text{CalctimeVSenergy}[U_{kin}, U_{kin0}, F_0, \lambda, \lambda_{cv}] := \\
\text{Module}[\{g, U_0, U\}, \\
g := \left(\frac{eF_0\lambda}{\pi m_\infty \lambda_{cv}}\right)^2; (*\text{see equation (5.21)}*) \\
U := \frac{4U_{kin}}{m_\infty \lambda_{cv}^2}, U_0 := \frac{4U_{kin0}}{m_\infty \lambda_{cv}^2} + g; (*\text{see equations (5.30) and (5.28)}*) \\
\frac{2}{\sqrt{2U_0}} \log \left[ 2 \sqrt{\frac{U_0 g}{U_0 - U}} + 2 \sqrt{\frac{U g}{U_0 - U}} \right] \\
\frac{2}{\sqrt{2U_0}} \log \left[ 2 \sqrt{\frac{U_0 g}{U_0 (U_0 - g)}} + 2 \sqrt{\frac{(U_0 - g)g}{U_0 (U_0 - g)}} \right];
\]

The derivative of the above function is the probability function of gaining a certain energy for a given pulse duration \( \tau \) (see equation (5.33))

\[
\text{CalcPvsE}[U_{kin}, U_{kin0}, F_0, \lambda, \lambda_{cv}, \tau] := \\
\text{D[CalctimeVSenergy[[U_{kin}, U_{kin0}, F_0, \lambda, \lambda_{cv}], U_{kin}],];}
\]
The next function calculates the probability distribution for an electron gaining certain energy while accelerated in the evanescent optical field. It defines properly the boundary of the probable energy electron can gain. The maximum energy $U_{\text{kinmax}1}$ that the electron can gain is the value defined by equation (5.18). The electron can also gain all the energies from 0 to $U_{\text{kinmax}1}$. Those energies, $U_{\text{kinmax}2}$ can be calculated form function CalctimeVSenergy. If this value of $U_{\text{kinmax}2}$ is larger then the value of $U_{\text{kinmax}1}$ then it is assumed that the electron gains only $U_{\text{kinmax}1}$. Next the probability of gaining this energy is calculated using function CalcPvsE. This function now returns non-zero value only if the calculated energy is within the bounds defined before.

$$
P_{\text{vsE}}[U_{\text{kin}}, U_{\text{kin}0}, F_0, \lambda, \lambda_e, \tau] :=$
Module[{g, U_{\text{kinmax}}, U_{\text{kinmax}1}, U_{\text{kinmax}2}, temp1, temp2, u},

g := \left(\frac{eF_0\lambda_e}{2\pi m_e\lambda_e}\right)^2; (*\text{see equation (5.21)}*)

U_{\text{kinmax}} := \frac{m_e\lambda_e^2 g}{4} + U_{\text{kin}0}; (*\text{see equation (5.18)}*)

temp1 = \text{Solve}[\text{CalctimeVSenergy}[u, U_{\text{kin}0}, F_0, \lambda, \lambda_e] == \tau, u];

U_{\text{kinmax}} := \text{Min}[U_{\text{kinmax}}, U_{\text{kinmax}1}, U_{\text{kinmax}2}]; (*\text{only the lowest energy is the proper one}*)

temp2 := \text{CalcPvsE}[U_{\text{kin}}, U_{\text{kin}0}, F_0, \lambda, \lambda_e, \tau]; (*\text{calculates probability}*)

\text{If}[U_{\text{kin}} < U_{\text{kin}0}, 0, \text{If}[U_{\text{kin}} > U_{\text{kinmax}}, 0, \text{temp2}]] (*\text{and returns 0 if the energy is out of bounds}*)];