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LETTER TO THE EDITOR

Prediction of step-like occupation and inversion of states in thin films exposed to laser pulses

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Abstract
We report on many-particle simulations of the quantum electron gas in thin metal films, irradiated with intense femtosecond laser pulses at grazing incidence. A gradual population of electron states is predicted, where subsequent steps of occupation probabilities are separated by the incident photon energy $\hbar\omega_0$. During the laser interaction with pulses of the order of 100 fs, oscillations of the occupation of states in the energy domain are shown to appear, leading to an inversion of states in the conduction band. On the basis of the Fermi–Dirac distribution and resulting final occupations of states, an estimate of the average temperature of the conduction electrons in metal films subject to laser irradiation is given.

Properties of metal surfaces exposed to short laser pulses have been the topic of several recent investigations. An example is the generation of high harmonics during the interaction of laser fields with solid targets at intensities of the order of 10^{12} W cm^{-2}, which has been analysed in recent theoretical investigations by Faisal and Kamiński (1997) within a microscopic Floquet–Bloch theory of interaction of lasers with periodic structures. Calculations on the basis of this non-perturbative theory have revealed photoelectron spectra from solids with a sequence of emission bands separated by bands of zero currents at an interval of the incident photon energy (Faisal and Kamiński 1998). In recent experiments, unexpected high photoelectron emission rates have been measured in the case of the irradiation of gold surfaces by infrared laser pulses at intensities of the order of 10^7 W cm^{-2} (Farkas *et al* 1998). Earlier measurements of higher harmonics from gold surfaces subject to moderate laser fields had already shown the appearance of the fifth harmonic in the photon emission spectrum (Farkas *et al* 1992). A generation of x-ray pulses by intense laser irradiation of metal surfaces at grazing incidence

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in the simultaneous presence of a strong static electric field has been predicted recently on the basis of a dipole-layer model (Varró and Ehlotzky 1997, 1998). Another example of investigations are simulations of the dynamics of the quantum electron gas in thin metal films subject to intense fields (Schwengelbeck *et al* 1998, 2000), which have shown plasmon-induced coherent photon emissions in the ultraviolet region.

The aim of this letter is to report on non-perturbative simulations of the irradiation of metal surfaces by femtosecond laser pulses at grazing incidence, i.e. with the electric field component of the laser pointing in the x-direction, perpendicular to the metal surface. The investigation is based on a time-dependent independent-particle description of a fermion gas in connection with the Fermi–Dirac distribution function. In the following we compute the dynamics of the electron gas of a thin gold film subject to laser fields with pulse durations of the order of 100 fs at a wavelength of $\lambda_0 = 800$ nm ($\hbar\omega_0 = 1.55$ eV).

To simulate the many-particle dynamics of the system, the upper band of the metal film is represented by $N = 246$ independent particles. Initially, we assume a cold electron gas, occupying different orthogonal eigenstates associated with discrete energy levels of the model potential

$$V_0 = \begin{cases} 0 & 0 < x < L \\ U_0 & \text{elsewhere} \end{cases} \tag{1}$$

up to the Fermi energy $E_F = 5.51$ eV, where $U_0 = 9.81$ eV represents the potential value at the surface (cf Michaelson 1977) and $L = 32$ nm is the thickness of the film (workfunction $W = U_0 - E_F = 4.3$ eV, $\hbar\omega_0 < W$). On the one hand the given value of L in the nanometre region corresponds to a domain of current physical interest—on the other hand, it represents a practicable choice from the point of view of numerical computation. In the present case, the total number of discrete eigenstates of the potential (1), $\phi_i = \phi_i(x, m_s)\exp(-(\mathrm{i}/\hbar)E_i t)$, $i = 1, \ldots, N'$, is given by $N' = 2 \times 164$, where the factor 2 results from the possible electron spin orientations with $m_s = \pm\frac{1}{2}$, and E_i is the corresponding energy. It should be noted that at intensities of the order of magnitude of 10^{12} W cm^{-2}, a change of electron spin m_s is not to be expected to occur in practice. Hence, before the laser interaction, energy levels are filled twice up to the Fermi level E_F and the associated initial occupation probability of states at temperature $T = 0$ is given by $f^0(E_i) = 1$ for $E_i - E_F \leqslant 0$ and $f^0(E_i) = 0$ for $E_i - E_F > 0$, according to the Fermi–Dirac distribution (e.g. Ziman (1968))

$$f(E_i) = \frac{1}{\exp[(E_i - E_F)\beta] + 1}, \tag{2}$$

where $\beta = 1/kT$, k is the Boltzmann constant and T is the temperature. Lower energy bands of the metal, filled with electrons at virtually all temperatures, are not considered in the present investigation.

The interaction of the electrons of charge e with the laser field, polarized along the x-axis, is given in length gauge by $V = ex\xi(t)F_0\sin(\omega_0 t)$, where F_0 is the laser amplitude, ω_0 is the laser frequency, and the pulse envelope of duration τ is modelled by $\xi(t) = \sin^2(\pi t/\tau)$. The decrease in the laser field and the laser–electron interaction inside the metal film is described by an exponential function $\mathrm{e}^{-x/\delta}$, where the skin depth $\delta = c/\sqrt{\omega_p^2 - \omega_0^2}$ is related to the plasma frequency ω_p of gold and amounts to 22 nm (cf Jones and March 1985).

On starting with the above-defined electron configuration with given initial states $\psi_j(x, t_0)$, the dynamics of the single-particle wavefunctions in the conduction band of the metal film subject to the laser field is governed by the time-dependent set of Schrödinger equations

$$\mathrm{i}\hbar\frac{\partial}{\partial t}\psi_j = -\frac{\hbar^2}{2m}\frac{\partial^2\psi_j}{\partial x^2} + V_0\psi_j + V_{\mathrm{int}}\psi_j, \qquad j = 1, 2, \ldots, N \tag{3}$$

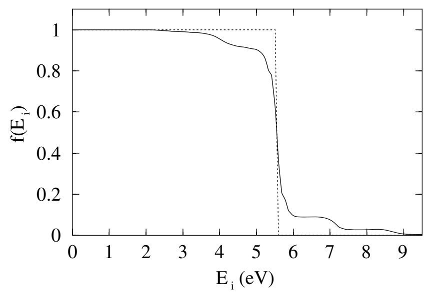

Figure 1. Occupation probability of states in a metal film of thickness $L = 32$ nm after a short laser pulse of duration 17.34 fs (solid curve), starting from the initial distribution at zero temperature (dotted curve). The interaction with laser photons of energy $\hbar\omega_0 = 1.55$ eV at peak intensity $I_0 = 10^{12}$ W cm^{-2} leads to a step-like occupation of states above the Fermi energy $E_F = 5.51$ eV (workfunction $W = 4.3$ eV) by one- and two-photon absorptions.

on the basis of negligible electron–electron interaction, which can be solved by numerical integration (cf Press *et al* (1995)). It should be noted that in contrast to two-particle quantum systems in laser fields, where the Coulomb interaction between the individual particles has substantial influence on the system dynamics (cf, e.g., Becker and Faisal (1996), Schwengelbeck (1999), Mompart *et al* (2002)), two-particle interactions between 'free' electrons in the metal electron gas can be regarded as negligible. The total wavefunction of the system is then given by the Slater determinant $(N!)^{-1/2}\det[\psi_j(x_i, m_{s_i}, t)]$ (e.g. Messiah (1970)), where the anti-symmetric linear combination of products of the N single-electron wavefunctions reflects the fact that the electrons obey the exclusion principle, so that no two electrons can be in the same dynamical state. It should be noted that in the numerical simulation continuum states are also taken into account. In the present case with short femtosecond pulses at moderate laser intensities of about 10^{12} W cm^{-2}, only a minor contribution of continuum states to the total wavefunction and the related photoemission has been found, which requires an absorption of at least three 1.55 eV photons to overcome the system workfunction $W = 4.3$ eV.

The final occupation probability of the states is given by the sum over the absolute square of projections of the $N = 246$ final single-particle wavefunctions on the eigenstates ϕ_i of the metal film potential (1),

$$f(E_i, t_f) = \sum_{j=1}^{N} \left| \int dx_j \sum_{m_{s_j}} \phi_i^*(x_j, m_j)\psi_j(x_j, m_{s_j}, t_f) \right|^2. \tag{4}$$

From the Fermi–Dirac distribution function (2) and on averaging over the total number of states N', we obtain the mean temperature $\bar{T}$ of the electron gas after the laser interaction,

$$\bar{T}(t_f) = 1/k\bar{\beta}(t_f), \qquad \text{where } \bar{\beta}(t_f) = \frac{1}{N'} \sum_{i=1}^{N'} \frac{\ln[f(E_i, t_f)^{-1} - 1]}{E_i - E_F}. \tag{5}$$

Figure 1 depicts the initial occupation probability of states at zero temperature and shows the resulting final occupation probability of states after a short laser pulse of duration $\tau = 17.34$ fs (6.5 optical cycles). The laser interaction with photon energy $\hbar\omega_0 = 1.55$ eV at a peak intensity $I_0 = 10^{12}$ W cm^{-2} leads to a step-like occupation of states above the Fermi energy $E_F = 5.51$ eV by one-photon and two-photon absorptions. A question arising is, if a similar electron structure in metal films emerges in the case of longer pulse durations and higher laser peak intensities.

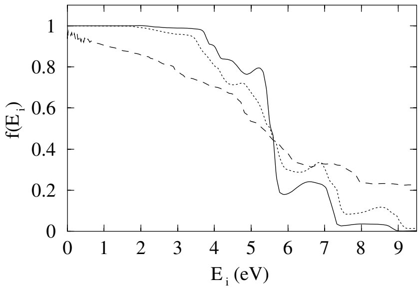

Figure 2. Occupation probability of states after a laser pulse of duration $\tau = 65.35$ fs (24.5 optical cycles) at different peak intensities, $I_0 = 5\times10^{11}$ W cm^{-2} (solid curve), $I_0 = 10^{12}$ W cm^{-2} (dotted curve) and $I_0 = 5 \times 10^{12}$ W cm^{-2} (dashed curve). In the first two cases, the final distributions appear with a step-like occupation of states, where subsequent steps are separated by the photon energy $\hbar\omega_0 = 1.55$ eV. In the case of $I_0 = 5 \times 10^{12}$ W cm^{-2}, the occupation of states is almost constantly decreasing.

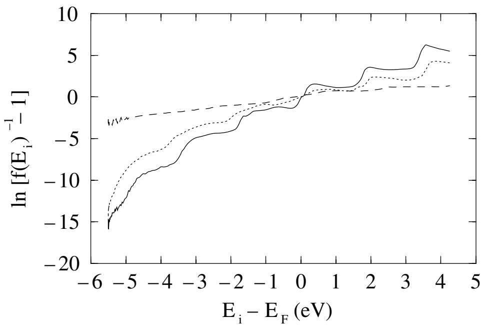

Figure 3. Plot of the logarithm of $f(E_i)^{-1} - 1$ versus $E_i - E_F$ (cf equation (5)) after a 65.35 fs pulse (24.5 optical cycles) at different peak intensities, $I_0 = 5 \times 10^{11}$ W cm^{-2} (solid curve), $I_0 = 10^{12}$ W cm^{-2} (dotted curve) and $I_0 = 5 \times 10^{12}$ W cm^{-2} (dashed curve) (same parameters as in figure 2). The plot reveals a step-like occupation structure across virtually all states. The resulting temperature (5) of the electron gas amounts to $\bar{T} = 6154$ and 9051 K in the case of $I_0 = 5\times10^{11}$ and 10^{12} W cm^{-2}, respectively. In the case of a peak intensity $I_0 = 5 \times 10^{12}$ W cm^{-2}, $\bar{T} = 20\,287$ K.

Figure 2 shows the final probability distributions after short laser pulses of duration $\tau = 65.35$ fs (24.5 optical cycles) for three peak intensities, $I_0 = 5 \times 10^{11}$, 10^{12} and 5×10^{12} W cm^{-2}. In the lower intensity cases, the figure shows step-like occupation probability distributions, where subsequent steps are separated by the photon energy $\hbar\omega_0 = 1.55$ eV. The corresponding average temperature (5) amounts to $\bar{T} = 6154$ and 9051 K, respectively. In the case of $I_0 = 5 \times 10^{12}$ W cm^{-2}, an almost constantly decreasing occupation of states with increasing energy is associated with an average temperature of $\bar{T} = 20\,287$ K ($k\bar{T} = 1.75$ eV). It is noted here that similar results with a decreasing occupation of states over the entire energy range (without an inversion of states) have also been found in case of intensities $I_0 \geqslant 5 \times 10^{12}$ W cm^{-2} with pulse durations longer than 65.35 fs. In figure 3, the logarithm of the inverse occupation probability $f(E_i)^{-1} - 1$ (cf equation (5)) is plotted for a pulse duration of 65.35 fs and the peak intensities $I_0 = 5 \times 10^{11}$, 10^{12} and 5×10^{12} W cm^{-2}. The plot reveals a step-like occupation of virtually all states after the laser interaction.

In figure 4 the occupation probability of states after laser interaction with different pulse durations up to $\tau = 129.36$ fs at peak intensity $I_0 = 10^{12}$ W cm^{-2} is shown. In case of the

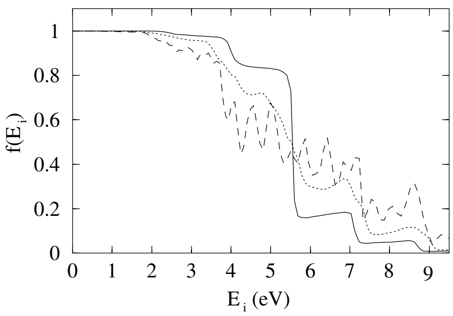

Figure 4. Occupation probability of states after different laser pulse durations, 33.34 fs (solid curve), 65.34 fs (dotted curve) and 129.36 fs (dashed curve) at peak intensity $I_0 = 10^{12}$ W cm^{-2}. In the case of the shorter pulse, the final occupation of states appears with four subsequent steps of the width of the incident photon energy $\hbar\omega_0 = 1.55$ eV. In the case of the longer pulse, corresponding to 48.5 optical cycles, the step-like occupation goes over to an inversion of states.

shorter pulse with $\tau = 33.34$ fs (12.5 optical cycles) the occupation of states appears with four steps of the width of the incident photon energy $\hbar\omega_0 = 1.55$ eV. In the case of pulse durations longer than 65 fs, the step-like occupation of states begins to break up. After pulse durations of more than 100 fs, one observes several inversions of states in terms of an oscillating occupation probability distribution. In the simulations performed so far, we have found no clear indication that the timescale for this inversion is related directly to the intensity of the pulse or to laser fluence, i.e. the integral of the square of the laser field over the pulse duration. The mean temperature in the case of a pulse duration of 33.34 fs amounts to $\bar{T} = 6269$ K. In the case of pulse durations of 65.34 and 129.36 fs, the final average temperatures of the electron gas amount to $\bar{T} = 9051.25$ and 8801 K, respectively. The lower value of temperature $\bar{T}$ in the latter case results essentially from the aforementioned inversion of states, which contributes with negative values to the average temperature of the many-particle system. Figure 5 shows the evolution of the occupation probabilities in the case of a 129.36 fs laser pulse with peak intensity $I_0 = 10^{12}$ W cm^{-2}. It can be seen that during the laser interaction, an increasing oscillation of the occupation of states in the energy domain occurs, leading to an inversion of states in the conduction band during laser interaction, as mentioned before. The reader may also compare figure 5 with the dashed curve in figure 4, which shows more clearly the occupation of states at the end of the laser pulse and for the same parameters as used in figure 5.

Similar results to the reported here have also been obtained in simulations with other laser pulse envelopes, different values of film thickness L and different particle numbers modelling the electron gas of the system. It should also be noted that similar results have been obtained in computations with different values of potential height U_0 and Fermi energies E_{F}, corresponding to other metals such as aluminium and copper.

To conclude, we have investigated the irradiation of a metal film by femtosecond laser pulses at grazing incidence by numerical simulation. To this end, an independent many-particle description of fermion gas has been employed, involving the propagation of different, each orthogonal, single-electron wavefunctions during the interaction with laser pulses. Based on the Fermi–Dirac distribution and the resulting occupation probability of states, we have given estimates of the average temperature of the conduction electrons in metal films, exposed to the laser fields. Final occupation probabilities in the case of short pulses between 12 and 65 fs at an intensity of the order of 10^{12} W cm^{-2} appear with a step-like distribution function, where subsequent steps are separated by the photon energy $\hbar\omega_0 = 1.55$ eV. In the case of longer pulses, the model calculations indicate that the step-like structure in the conduction band goes

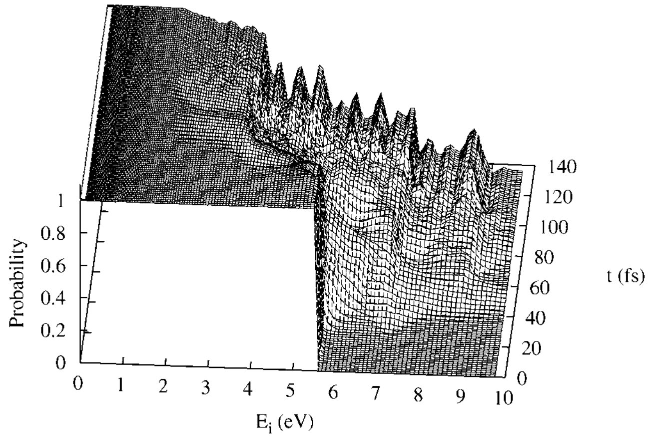

Figure 5. Evolution of the occupation probability $f(E_i)$ in the metal film during the interaction with a femtosecond laser pulse with photon energy $\hbar\omega = 1.55$ eV and peak intensity $I_0 = 10^{12}$ W cm^{-2}. During the laser interaction, increasing oscillations of the occupation of states in the energy domain appear, leading to an inversion of states (cf figure 4 and text).

over to an inversion of states. Similar results have also been obtained in simulations using different system parameters such as film thickness L and different numbers of particles N to model the electron gas in the metal films.

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