4.5 TDDFT and Classical Dynamics for Investigation of Intense and Ultrashort-Pulse Lasers Interaction with Surfaces

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Progress in laser technology during the past few years has made it possible to generate ultrashort laser pulses at high intensities and optical wavelengths ($\lambda \approx 800$ nm). In some cases the laser period has become comparable with the full width at half maximum τ of the temporal intensity profile [1]. In this regime many processes become sensitive on the carrier-envelope or "absolute" phase φ. The reproducible preparation of such laser pulses is essential e.g. in attosecond inner-shell spectroscopy or generation of coherent X-ray radiation for biological microscopy. Although the phase of the laser can be fairly precisely controlled, the determination of its value has failed so far. Photoionization of atoms in strong fields is a promising process for the determination of φ [2,3], but detectable phase effects could only be observed for circularly-polarized light. Another candidate to determine φ could be photoemission from metals. Up to now there exist two studies on photoemission from metal (jellium) surfaces. One of them is based on quasi-static ionization rates [4]. Although a considerable dependence on the absolute phase was found using this model, its validity is restricted to the high intensity regime ($I \geq 10^{14}$ W/cm^2), where other (destructive) processes become increasingly important. This reduces its attractivity considerably from an experimental point of view. The other study uses time-dependent density functional theory (TDDFT) to describe photoemission [5]. This calculation supports the previous description at high intensities. Surprisingly, a strong phase dependence of the photocurrent is found already at moderate intensities (I$\approx 10^{12}$ W/cm^2).

In order to better understand the results of the quantum mechanical calculations we combined it with a semiclassical simulation of electron motion in a strong external laser field. The model used for the determination of the photocurrent is a classical-trajectory Monte Carlo method in 1D with including tunneling of electrons through the surface barrier. This method has been used for the determination of the photoionization rates of the H atom giving fairly accurate results [6]. The time dependent potential determining the motion of the electrons escaping from the surface was calculated using TDDFT. All calculations presented here were performed for a metal in jellium approximation with a Wigner-Seitz radius (radius of the sphere a metal electron occupies on the average) of $r_s = 3$ a.u. The applied external potential described a laser pulse with a peak intensity of $I = 1.57 \times 10^{13}$ W/cm^2 (E_0=0.02 a.u.) and 4 fs duration (Gaussian envelope). Its wavelenghts was $\lambda = 790$ nm.

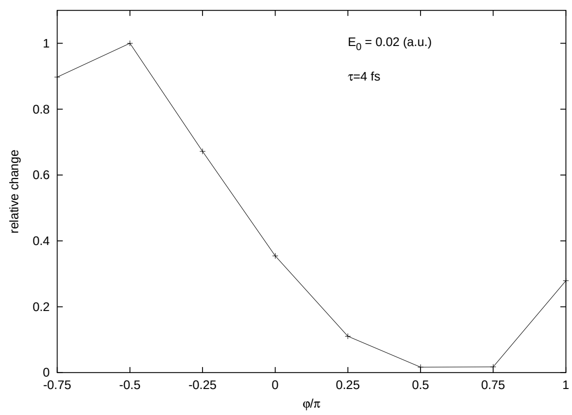

Figure 1. Relative change of the photocurrent as a function of the absolute phase φ.

Our semiclassical model also shows a phase dependence of the photocurrent. The relative change, however, is much stronger compared to the predictions of the quantum mechanical calculations. This overestimation of the phase effect can be most likely explained by the way the emission process is included in our model. The surface density of states (SDOS) is replaced by a δ-function at the Fermi energy of the jellium. Even small excitations of the electrons to the empty states of the conduction band thereby increasing the tunneling probability are so far not included. In such a model emission crucially depends on the maximum field strength (i.e. the absolute phase) reached.

To study the dependence of the photocurrent on the laser intensity we increased the pulse duration to 10 fs (phase dependence disappears for pulse $\tau \geq 10$ fs). The maximum field strength was varied between $I \approx 0.5 \times 10^{12} - 10^{14}$ W/cm^2 ($E_0 = 0.004 - 0.05$ a.u.).

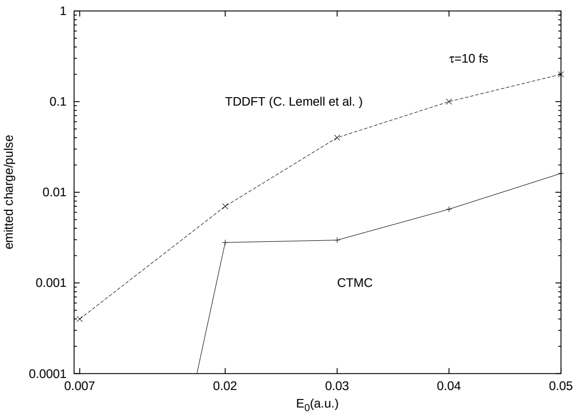

Figure 2. Total amount of charge emitted per pulse as a function of the maximum field strength.

The other parameters of the laser were the same as in the case of the phase dependence calculations. We find that the total emission in the strong field regime ($E_0 \geq 0.02$ a.u.) is by an order of magnitude smaller than predicted by TDDFT. In the multi-photon regime the photoemission is, as expected, largely suppressed by the increased width of the potential barrier.

The next step in the development of our simulation will be aimed at a more accurate description of the SDOS by also Monte-Carlo averaging the initial conditions of the electron trajectories over the complete energy range of the occupied states of the jellium in ground state. Furthermore we plan to include excitation processes on a phenomenological level by simulating the motion of the electron due to the (screened) external potential also inside the surface.

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