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Time-resolved photoelectron emission from surfaces

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Abstract. The attosecond streaking technique provides a unique tool to study the motion of particles with unprecedented accuracy. While originally developed for investigations of atomic systems streaking is now also applied to condensed-matter systems. We have developed a model to simulate the motion of electrons under the combined influences of crystal atoms, other target electrons, and the external pump and streaking fields. Good agreement between this semiclassical model and experimental results is reached.

1. Introduction

Direct observation of electronic motion in extended systems has always been a dream for physicists. With the advent of the attosecond streaking technique [1] this goal has recently come into reach. As in the case of the streak camera, an ultrashort flash (here an XUV pulse of about 100 eV and a duration as short as about 100 as) first excites the system. Then, a picture of the evolving system is taken, i.e. an emission spectrum is recorded. The “camera” in the case of attosecond streaking is the field of a phase stabilized linearly polarized IR pulse (wavelength $\sim$ 750 nm, duration $\sim$ 5 fs) collinear with the XUV pulse. The momentum of emitted electrons after excitation is shifted due to the streaking IR field depending on the time of emission $t_0$ [1]:

$$\Delta \vec{p} = \int_{t_0}^{\infty} \vec{F}(t) \, dt = -\vec{A}(t).$$

(1)

$F(t)$ and $A(t)$ are the electric field and the vector potential of the IR laser pulse, respectively. By shifting the XUV and IR pulses in time relative to each other it therefore becomes possible to take a snapshot of the IR vector potential [2]. If more than one peaks are present in the energy spectrum (emission from different target states) relative timing information of different groups of electrons can be gained [3] which has raised hope to get access to timing information of intra-atomic processes. However, theoretical work shows that $\Delta p$ is not exactly $A(t_0)$ but rather $A(t_0 + \Delta \tau)$ [4]. In some cases, $\Delta \tau$ can even become negative indicating emission prior to excitation. The reason for this apparently advanced emission is the combined influence of the Coulomb and IR fields on the electron trajectory neglected in Eq. 1. This so-called Coulomb-laser coupling in the exit channel can be described classically [4]. Even for simple atomic systems, extracting timing information has proven to be non-trivial and exciting. This is all the more true for extended and condensed-matter systems.
In the ground-breaking streaking experiment using a tungsten target [5] lines from the excitation of conduction-band as well as core-level lines were measured. Evaluating the streaked spectra it turned out that a theoretical analysis of the data is challenging due to various effects which cannot be deconvoluted. Most importantly, an absolute reference time $t_0$ is difficult to establish relative to which delays are to be derived. Many contributions to the relative delay between two lines cancel out if groups of electrons of similar origin are compared. In such cases, the duration of electron transport within the target can be measured. One candidate are targets consisting of a metal bulk and single or multiple adlayers of other material. Others are free-electron metals where the second line is provided by electrons which have excited a plasmon on their way to the detector.

In this communication we present the simulation of streaking spectra for a tungsten bulk covered with one or two magnesium adlayers and bulk Mg. In the following section we will briefly review the methods, in section 3 results of our simulation are presented.

2. Simulation

Our simulation method has been described in detail elsewhere [6]. We therefore give here only a short overview of its features.

Three distinct steps can be distinguished (see Fig. 1): first, the incoming XUV laser pulse (blue line in Fig. 1) excites electrons within the target. Their energy is shifted by the XUV-photon energy (in this work $\hbar \omega \approx 130 \text{ eV}$). The angular distribution of the excited electrons is, in general, non-isotropic (the energy-dependent parameter $\beta$ is for many isolated atomic systems well known). For the solid-state environment we use, for simplicity, isotropic source distributions. Tests with $\beta$ ranging from 0 (isotropic emission) to 2 ($\cos^2$-distribution) have shown only small differences in the result.

After its release, the electron in the target is subject to the electric field of the streaking laser pulse (red line in Fig. 1) and may be deflected from a straight-line trajectory by scattering events at target atoms (elastic) or electrons (inelastic). Elastic scattering cross sections of electronic wavepackets driven by the light pulse scattered at muffin-tin potentials representing the target constituents have been calculated using the ELSEPA code [7]. The mean free paths and the angular distributions in inelastic scattering events are derived from the energy and momentum dependent dielectric function $\varepsilon(q, \omega)$ which, in turn, can be extracted from independent experimental optical data [8, 9]. In between scattering events electrons are transported using the free-electron dispersion relation $E = k^2/2$. Deviations of measured delays from simulated results may then indicate a contribution to the delay due to the crystal structure of the target.

If an electron overcomes the surface barrier and leaves the target it is propagated in the streaking field alone. As the IR pulse is polarized linearly in the plane normal to the surface,
the resulting above-surface part of the momentum shift is the dominant part of the observed energy shift in the streaked spectrum. The influence of the distribution of escape times on the measured delay has been discussed in detail in [6]. One challenge for the interpretation of the original tungsten surface experiment [5] is that the energy difference between neighboring lines of W coincides with a pronounced peak in the loss function (probability for inelastic scattering $\sim \Im {-1/\varepsilon(q, \omega)}$). In such a case, the run-time difference contains contributions from conduction-band electrons having undergone one inelastic scattering event generating a plasmon on the way [6]. The run-time difference is, thus, no longer uniquely attributable to the difference between conduction and core-level emission.

Other unknowns contributing to the delay are e.g. the electric field at the surface or the life-time of holes in the photoexcited target atoms [4]. To prevent such complications and to disentangle transport from other contributions we study differences between spectra instead of single spectra. In particular we investigate the following targets: a (almost) free electron-gas metal (magnesium) and a tungsten bulk covered by one and two magnesium layers (Fig. 2). In the latter case the relative delay of tungsten lines with respect to the Mg lines, i.e. the time it takes an electron to cross the Mg layer(s), can be measured. For bulk Mg the relative delay between the conduction-band line and the line resulting from excited electrons having excited one plasmon in the remaining electron gas is studied (Fig. 2a).

3. Results
3.1. Bulk Mg

Usually, the evaluation of streaking spectra is restricted to target lines which are shifted by the IR streaking field only. One therefore excludes lines originating from electrons having undergone inelastic collisions within the target. One the one hand, loss processes might lead to the overlap of different groups of electrons (as in the case of W [5, 6]), on the other hand, the lines are usually weak and overshadowed by the background signal. For nearly free electron gas metals (e.g. Al and Mg), however, the loss function features one prominent peak at the plasmon frequency $\omega_{pl} = \sqrt{4\pi n}$ (in atomic units; $n$ . . . density of the electron gas; Fig. 3) leading to a high excitation probability. Furthermore, the conduction-band line is well separated from any core-level lines (Mg: excitation of 2p line at 50 eV; Fig. 3) allowing for an unambiguous identification of the plasmon line in the spectra.

The photon energy of the XUV pulse was chosen to be about 130 eV to match recent
experimental conditions. Taking the loss function from Fig. 3 the inelastic mean free path of electrons in the material, \( \lambda_i \), is for the energy range of interest (100 to 140 eV) between 6.5 and 7.5 Å corresponding to the average escape depth of electrons from this energy range. Therefore, while conduction band electrons contributing to the high energy peak (no loss) have traveled on average only about 7 Å, electrons having excited one plasmon have an extended range. In our simulation we find a delay of about 55 as (Fig. 4) in good agreement with the average escape depth for the convolution of two escape-probability curves with equal escape depth which is given by \( \lambda_{\text{tot}} = \lambda_i + \lambda_i / 2 \). Possible uncertainties in the deconvolution of streaking spectra cancel out as they are present for both components of the spectra and any significant spectral overlap can be rules out. The observed delay is only due to transport of electrons after plasmon excitation. Changing the XUV photon energy would open the possibility to measure the dispersion relation for such high-energy electrons of the bulk material.

\[ \text{Figure 3. Loss function of Mg; the plasmon peak at the plasmon frequency } \omega_{pl} \text{ dominates the spectrum. The closest core excitations is well separated (2p-excitation at 50 eV).} \]

\[ \text{Figure 4. Streaking spectrum of bulk Mg. Two well separated lines from excited conduction band electrons can be observed. Top line: no energy loss; bottom line: electrons have excited a plasmon in the electron gas. The relative delay is about 55 as.} \]

3.2. Mg layers on W
Alternatively, one can also determine the dispersion relation perpendicular to thin layers using the surface-streaking technique. This can be accomplished by adding monolayers on top of a bulk material and observing the delay of emitted electron groups. In this work, we present
results for a W bulk covered by one and two Mg layers (Fig. 2). In the simulation all materials are considered amorphous, i.e. the Mg layers have bulk properties and a width equal to the layer spacing ($\sim 3.2 \text{ Å}$). The resulting spectra feature increasingly important additional lines at smaller energies which can be connected to the excitation of bulk plasmons in Mg. In the context of thin layers this has to be considered an artifact of our simulation method. Excitation of surface plasmons which is also included in our simulation is strongly suppressed as electron emission in normal direction (for detection perpendicular to the surface) only very weakly couples to surface plasmons. The simulated delay increases linearly with the number of layers at the rate of $\sim 59$ as per layer. This is a few attoseconds more than the value for perpendicular transmission of the layer width by 100 eV electrons (54 as) and can be attributed to slightly increased trajectory lengths due to elastic scattering events within the surface layer(s). Again, scanning the XUV energy allows for the measurement of the dispersion relation (including possible interface effects) for thin metal layers in perpendicular direction.

While we have access to absolute delays in the simulation, experimental data currently provides only relative delays between bulk and adlayer lines. Preliminary results from the Garching group for the Mg-2$p$ and the W-4$f$ lines find a difference in delays for one and two monolayers of about 55 as in good agreement with our result [10].

4. Discussion and conclusion
Recent theoretical and experimental investigations have shown that the emission time contained in streaked energy spectra contains a wealth of information which cannot be determined by other spectroscopic means [3, 4]. Even for simple atomic systems extraction of all relevant contributions is challenging. In the case of extended systems such as solid surfaces excited by energetic photons, the situation is further complicated by multiple scattering and dissipative
processes.

In this work we have concentrated on emission time delay due to transport effects for core and conduction band levels of metallic systems with electron energies of about 100 eV. We model the target as amorphous with bulk values for the atomic and electron densities together with quantum mechanically calculated cross sections providing information about scattering events (i.e. the mean free path). Possible long-range effects of the positive hole left at the point of excitation as well as interface effects are not included in our simulation. The latter can be neglected in the special cases selected for our study as they would cancel out in the determination of the relative delay between lines.

For systems for which preliminary experimental results exist, excellent agreement with simulated data is found. The approximation in terms of an amorphous target appears to be justified for electron energies $\sim 100$ eV. Pronounced crystal-structure related effects should appear for smaller excitation energies which, however, are difficult to resolve in experimental investigations due to the ATI background in the energy spectra. Pronounced crystal-momentum effects were originally assumed to cause larger delay times between conduction band and bound state lines in the experiment by Cavalieri et al. [5]. Although the revised delay of $65 \pm 20$ as lies now closer to our simulated values of 45 as than the previously reported delay of 110 as, differences still remain. Effects from different timing contributions of electrons interacting with core and conduction band holes such as dynamic screening remain to be accounted for. Crystal dependent features in the dispersion relation can, in principle, be observed using adlayer systems if the photon energy of the exciting XUV pulse is scanned.

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