Publ. Astron. Obs. Belgrade No. 84 (2008), 241 - 244 *Contributed Paper*

SIMULATION OF STREAKING EXPERIMENTS AT SURFACES

C. LEMELL[1], B. SOLLEDER[1], K. TŐKÉSI[2], and J. BURGDÖRFER[1]

[1] *Institute for Theoretical Physics, Vienna University of Technology, Wiedner Hauptstraße 8-10, A–1040 Vienna, Austria*
E–mail: lemell@concord.itp.tuwien.ac.at

[2] *Institute of Nuclear Research of the Hungarian Academy of Sciences, (ATOMKI), H–4001 Debrecen, P.O.Box 51, Hungary*

Abstract. Recently, surprisingly large run-time differences of electrons emitted in laser-metal interactions have been reported. In our work we aim at simulating this experiment starting with electron excitation by the XUV pump pulse, subsequent electron transport within the target material, and finally emission into and transport in vacuum under the influence of the NIR pump pulse. Our results can only partly explain the measured data.

1. INTRODUCTION

Gaining deeper understanding of electron dynamics in condensed-matter systems is an important step towards developments in the fields of, e.g., electronics or information processing. Applying the "streaking" technique originally developed for gas targets (Kienberger et al. 2004) to photoelectrons from metal surfaces, electron dynamics in solids (time scale for electronic motion ~ 1 as $= 10^{-18}$ s) could be observed in real time (Cavalieri et al. 2007). In their setup two collinear linearly polarized laser pulses (extreme ultraviolet (XUV) and near infrared (NIR)) were directed at a W(110) surface under a grazing angle of incidence. Electrons excited by the pump pulse were transported in the field of the probe pulse changing their energy as a function of emission time. Photoelectrons escaping the target surface were detected by a time-of-flight spectrometer with the detection direction normal to the surface (see Fig. 1).

On top of a strong background signal originating from above-threshold ionization two prominent features were observed in each of the streaking spectra: a low energy peak around 55 eV and a high energy peak at 85 eV. The former was attributed to electrons excited from 4f states of tungsten, the latter to electrons from the target conduction band (5d and 6s states). A run-time difference between the two groups of electrons of 110 ± 70 as was observed.

2. SIMULATION

Our simulation includes three main ingredients (Lemell et al. 2008): 1) the interaction of the XUV pulse with the target material, i.e., the excitation of target electrons to

continuum states; 2) A classical-trajectory Monte-Carlo simulation of the transport of excited electrons through the material (Solleder et al. 2007) and 3) the propagation of electrons which have escaped the surface to the detector in the streaking field.

In photoemission experiments with a Ne gas target, the photon energy of the pump pulse was measured to be 91 eV with a full width at half maximum (FWHM) of about 6 eV. Therefore, target electrons from only 4 states can be excited to energies in the measured energy range (above 30 eV): 6s, 5d, 4f, and 5p. The former two form the conduction band of tungsten and lead to a broad peak around 83 eV emission energy, the latter two to a peak with its centroid at 56 eV. The streaked energy shift of these two peaks has been analyzed in order to determine the run-time difference between these two groups of electrons (Cavalieri et al. 2007).

Electrons excited by the pump pulse are propagated through the target material subject to elastic and inelastic scattering events and to deflection of their trajectories in the electric field of the NIR probe pulse. Doubly differential and total elastic scattering cross sections have been calculated with the ELSEPA package (Salvat et al. 2005) using a muffin-tin potential for the crystal atoms. The energy dependent elastic mean free path was derived from the total cross sections. Computation of inelastic scattering was based on an extrapolation of optical data (i.e., the frequency dependent dielectric function at $q = 0$) to the $q - \omega$ plane (Powell 1974, Penn 1984). Energy loss in an inelastic scattering event can lead to the release of a secondary electrons whose trajectory is also followed in our simulation. Between subsequent scattering events the differential equation for electron transport in the time-dependent electric laser field was solved. To assess the influence of the crystal structure two different dispersion relations (DR) were employed: the free-particle DR $E = k^2/2$ and a DR derived from ab-initio calculations (Calvalieri et al. 2007) of group velocities along the surface normal.

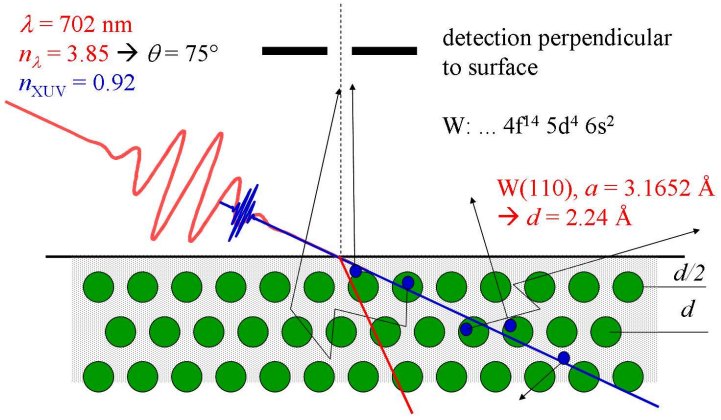

Figure 1: Schematic view of experimental setup: A 300 as XUV pulse hits the surface under a grazing angle of incidence ionizing target electrons. They are transported in the field of a 10 fs NIR pulse and detected by a TOF spectrometer mounted perpendicular to the surface.

Above the surface the electron is subject to the time-dependent streaking field $E(t)$ of the NIR laser pulse. It transfers a momentum of

$$\Delta p = \int_{t_{esc}}^{t_{end}} E(t)\, dt \tag{1}$$

to the electron where the integral is taken from the time of escape from the surface, t_{esc}, to the end of the laser pulse, t_{end}. Electrons with a final momentum perpendicular to the surface ($\theta \leq 5°$) are counted.

10^7 trajectories were started for each streaking spectrum, the time delay between pump and probe pulses was varied from -10 fs to 10 fs in 100 as steps.

3. RESULTS

Results presented in this section were calculated using the free-particle dispersion relation in our transport simulation.

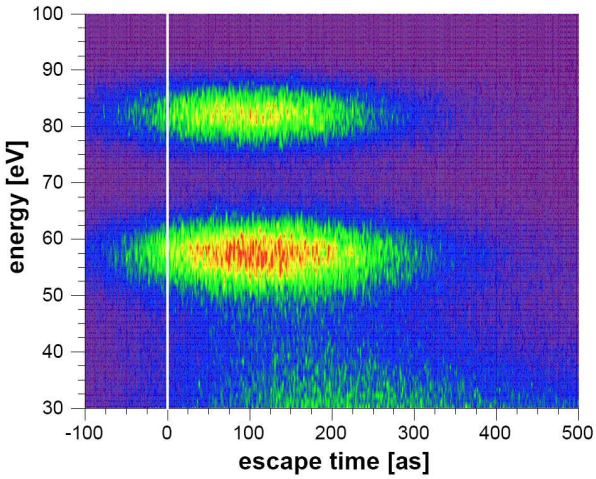

Figure 2: Energy vs. run time of electrons emitted from the surface.

In Fig. 2 the energies of electrons which have escaped the surface are plotted against their run time within the target where the zero point $t = 0$ is defined by the maximum of the envelope function of the pump laser field. Electrons with $t < 0$ have been released on the rising flank of the pulse with a duration (FWHM) of about 300 as. Two main features around 56 eV (4f and 5p electrons) and 82 eV (conduction band) can be seen. The escape time averaged over the peak regions is 159 as and 115 as for the low and high energy peaks, respectively. This difference in run time of our simulation lies at the lower limit of the experimental error bar. The present run-time difference can be accounted for by the following target properties: the inelastic mean free path (IMFP) of electrons with 56 and 83 eV emission energy is for both groups between 6 and 7 Å. Electrons which have not suffered any energy loss along their

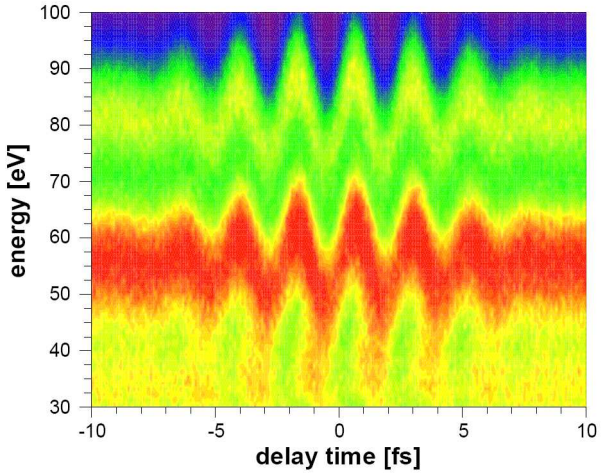

Figure 3: Streaking image for electrons detected in XUV-NIR laser-pulse surface interactions ($\theta_{esc} = 0 \pm 5°$).

trajectory in the target have traveled on the average a distance equal to the IMFP. This short traveling distance sets a lower limit for the run-time difference of only about 20 as. Scattering events increase this value to the observed 45 as. Using the crystal-structure DR the run-time difference is increased to about 80 as.

Fig. 3 shows the streaking image for the energy range from 30 – 100 eV. Momentum transfer due to the interaction with the probe laser field leads to the observed energy shift of the lines at 56 and 82 eV. We have found that influence of the streaking field on the trajectory does not change the observed run-time difference.

In conclusion, we have modeled a recent experiment by Cavalieri et al. who have succeeded in applying the streaking technique to solid-state targets. Our simulation includes excitation of target electrons, their transport through the metal, and, if they escape the target, streaking on their paths to the detector. Qualitative agreement (i.e., a run-time difference larger than derived from escape depths and electron velocities alone) with experiment is found, quantitative differences remain so far unexplained.

References

Cavalieri, A.L., Müller, N., Uphues, Th., Yakovlev, V.S., Baltuska, A., Horvath, B., Schmidt, B., Blümel, L., Holzwarth, R., Hendel, S., Drescher, M., Kleineberg, U., Echenique, P.M., Kienberger, R., Krausz, F., Heinzmann, U.: 2007, *Nature*, **449**, 1029.

Kienberger, R., Goulielmakis, E., Uiberacker, M., Baltuska, A., Yakovlev, V., Bammer, F., Scrinzi, A., Westerwalbesloh, Th., Kleineberg, U., Heinzmann, U., Drescher, M., Krausz, F.: 2004, *Nature*, **427**, 817; *ibid.* supplementary material.

Lemell, C., Solleder, B., Tökesi, K., Burgdörfer, B., Jakovlev, V.S., Cavalieri, A.L., Krausz, F.: 2008, *Phys. Rev. A*, in preparation.

Penn, D. R.: 1987, *Phys. Rev. B*, **35**, 482.

Powell, C. J.: 1974, *Surf. Sci.*, **44**, 29.

Salvat, F., Jablonski, A., Powell, C.: 2005, *Comp. Phys. Commun.*, **165**, 157.

Solleder, B., Lemell, C., Tökesi, K., Hatcher, N., Burgdörfer, J.: 2007, *Phys. Rev. B*, **76**, 075115.