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Laser-assisted dynamics on metallic surfaces

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Using femtosecond time-resolved photoelectron spectroscopy, we observe the laser-assisted photoelectric effect on a surface. We also present experimental measurements that permit to distinguish the laser-assisted photoelectric effect from other inherent surface processes, such as, above threshold photoemission, space-charge acceleration and hot electrons excitation.

Keywords: Time-resolved photoelectron spectroscopy; laser-assisted photoelectronic effect.

Se observa el efecto fotoeléctrico asistido por láser en una superficie usando espectroscopía fotoelectrónica resuelta en tiempo en femtosegundos. Se presentan mediciones experimentales que permiten distinguir el efecto fotoeléctrico asistido por láser de otros procesos inerentes a las superficies como son la fotoemisión sobre el umbral aceleración espacio-carga y excitación de electrones calientes.

Descriptores: Espectroscoía fotoelectrónica resuelta en tiempo; efecto fotoelectrónico asistido por laser.

PACS: 68.47.De; 78.47.tp

The laser-assisted photoelectric effect (LAPE) is a powerful tool for characterizing femtosecond-to-attosecond extreme-ultraviolet (EUV) pulses, and for time-resolved spectroscopy of electron dynamics in atoms. In LAPE, EUV and infrared (IR) pulses are focused into a gas with a varying relative time. The EUV radiation photoionizes the gas and, in the presence of the IR field, the energy of the photoemitted electrons can be modulated, leading to shifting or sidebands in the photoelectron spectrum.

This change has been used in past experiments to measure the duration of attosecond EUV pulses [1], and to measure an 8 fs Auger decay lifetime [2]. Recently, we observed the equivalent process in the original manifestation of the photoelectric effect *i.e.* photoemission from surfaces [3]. We were able to extract sideband amplitudes from the continuous photoemission spectra and recorded a cross-correlation between IR and EUV beam. This result is promising as surface LAPE has the potential to study ultrafast, femtosecond-to-attosecond time-scale electron dynamics in solids and in surface-adsorbate systems—where complex, correlated, electron relaxation processes are expected. It will also make it possible to characterize lower-flux and higher energy EUV pulses, because of the orders-of-magnitude higher density of target atoms as compared to gas jets.

In our experiment, we illuminate a Pt(111) surface simultaneously with EUV and IR pulses to observe LAPE in the photoelectron spectrum. The Pt crystal is mounted inside an ultrahigh vacuum chamber with a base pressure of 2×10^{-10} torr. IR pulses of 30 fs duration and 1.5 mJ at 780 nm are produced by a Ti:sapphire laser system. A beam splitter is used to divide the beam. Approximately 80% of the 780 nm beam energy is upconverted to the EUV using phase-matched high harmonic generation in a hollow fiber. A pair of Si:Mo multilayer mirrors is used to spectrally select the 27^{th} harmonic (29 nm), and an aluminum filter blocks the infrared beam while transmitting the EUV beam. The rest of the IR beam, with variable EUV-IR delay, is also incident on the sample at a small angle. The kinetic energy of the photoemitted electrons is measured using a time-of-flight (TOF) detector.

Figure 1a shows a series of photoelectron spectra around the Fermi edge for different EUV-IR time delays, taken with the IR beam polarization chosen parallel to the direction of detection. Outside the exact time overlap of the beams, the spectra are unaffected by the presence of the IR and show the typical d-band structure of a clean Pt(111) surface. Around time zero, however, sidebands arise on both sides of the unperturbed spectrum (taken without the IR pulse present). We model the absorption and emission of IR photons by -

$$f(E-E_o) = \frac{1-2A_1-2A_2}{\sqrt{2\pi\sigma^2}} e^{\frac{(E-E_o)^2}{2\sigma^2}} + \sum_{\pm}\left(\frac{A_1}{\sqrt{2\pi\sigma^2}} e^{\frac{(E-E_o\pm\hbar\omega)^2}{2\sigma^2}} + \frac{A_2}{\sqrt{2\pi\sigma^2}} e^{\frac{(E-E_o\pm2\hbar\omega)^2}{2\sigma^2}}\right), \quad (1)$$

We fit the convolution of this LAPE response function with the EUV-only photoelectron spectrum to the photoelectron spectrum in the presence of the IR field. The amplitudes A_1 and A_2 of the first two sidebands, as well as the width σ and the peak separation $\hbar\omega$ are left as fit parameters. Figure 1b shows the result for zero time delay. The response function corresponding to fit parameters $A_1 = 0.241 \pm 0.004$;

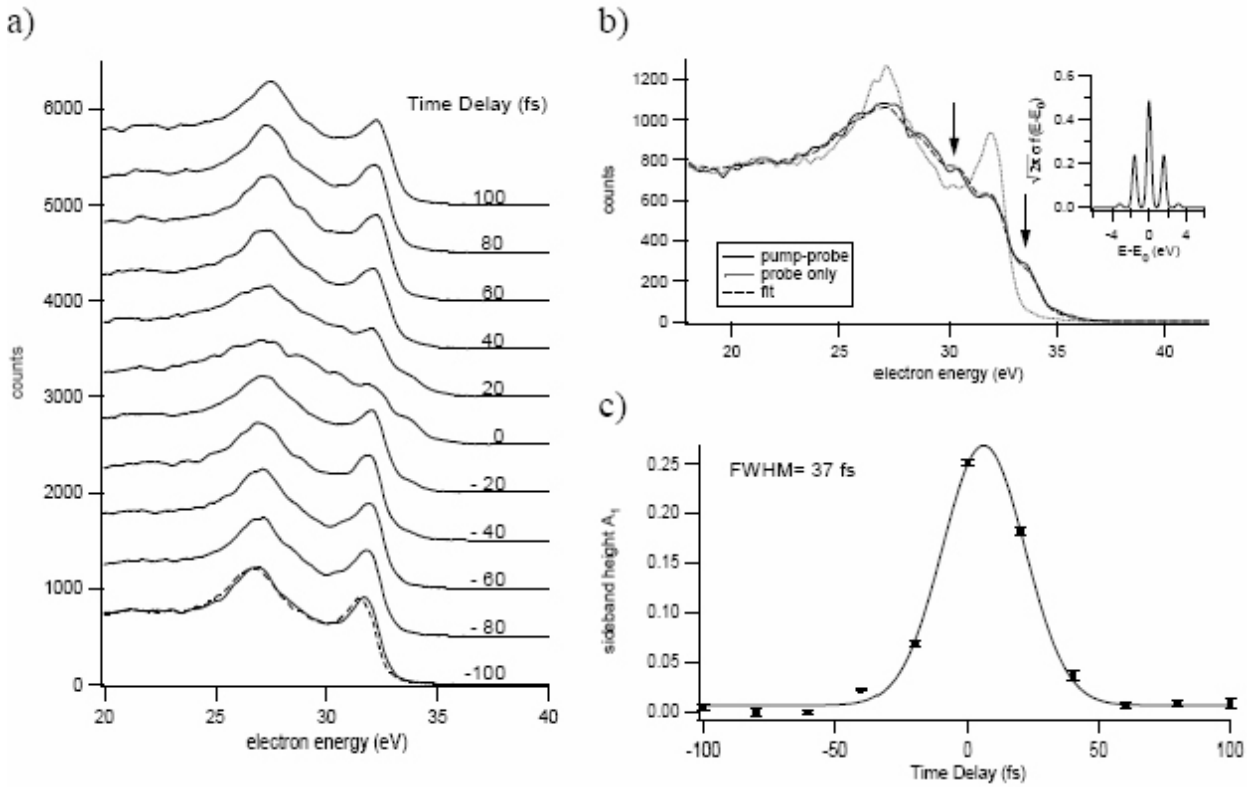

FIGURE 1. a) Photoelectron spectra taken at different time delays between EUV and IR. The dashed curve shows a spectrum taken without the IR beam present. b) The EUV-IR spectrum (solid line) taken at 0 fs time delay shows sidebands at ± 1.59 eV relative to the undressed Fermi edge position, as is shown by a fit (dashed) of the convolution of the LAPE response function [Eq. (1)] and the EUV-only data (dotted) to the EUV-IR data. c) The amplitudes of the sidebands versus time delay yields a cross-correlation between EUV and IR beams with a FWHM of 37 fs.

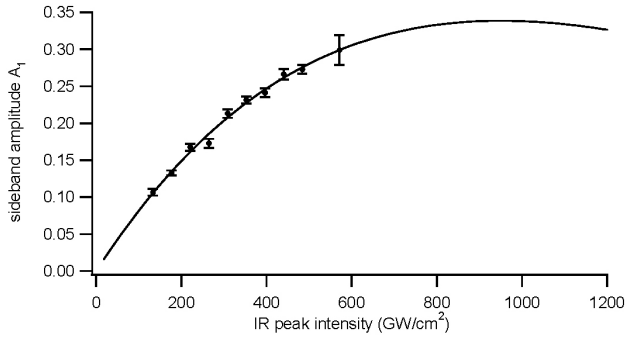

FIGURE 2. Extracted sideband intensity A_1 as a function of the effective IR beam peak intensity. The sideway intensity follows the theoretically expected square of the 1^{st} order Bessel function.

A_2=0.012$\pm$ 0.005; σ=0.23$\pm$0.03 eV; and $\hbar\omega$=1.59$\pm$0.02 eV, shown in the inset, very well reproduces the experimentally measured EUV-IR photoelectron spectrum. The sideband separation $\hbar\omega$ = 1.59 eV resulting from the fit corresponds very well to the wavelength of the IR beam of 780 nm – thus suggesting LAPE to be the underlying process. This is further supported by the fact that no sidebands are observed when the IR polarization is perpendicular to the direction of detection, and that the magnitude of the sidebands increases linearly with laser intensity.

We extract the sideband amplitudes from all spectra by similar fits, but with the peak separation as well as their width fixed to the values found at time zero. Figure 1c shows the resulting sideband amplitudes A_1 versus time delay yielding a cross-correlation between the IR and the EUV beam. From a Gaussian fit we derive an EUV pulse FWHM of 37 fs. Since the IR laser pulse duration is $\approx$ 35 fs, this cross-correlation signal indicates a much shorter EUV pulse. With more accurate characterization of the IR pulse duration, surface LAPE should yield time resolution in the fs regime or better. Moreover due to the considerably higher sample density, LAPE from solid surfaces provides much higher cross-correlation signals compared to the gaseous samples commonly used. Therefore, these observations should enable attosecond measurements of complex electron dynamics in solids.

However to extend these applications of LAPE to surfaces, it has to be unambiguously distinguished from hot electron excitation, above-threshold photoemission, and space charge acceleration as these effects can potentially lead to similar modifications of the photoemission spectrum and thus masking of LAPE [4]. Figure 2 shows the amplitudes of the 1^{st} order sidebands A_1measured at different pump laser peak intensities I. This intensity-dependence is fitted to a simple model [5] that has been found valid for the atomic case and

that predicts the sideband amplitudes to follow the square of a Bessel function:$A_n = J_n^2(a\sqrt{I})$. Here, a is a fit parameter and n denotes the number of absorbed or emitted IR photons. We find excellent agreement of our data with this model. Moreover, LAPE is the dominant process over almost the entire intensity range. Only for intensities higher than 1500 GW/cm^2, electrons from above-threshold photoemission as well as distortions of the spectrum due to space-charge acceleration start to become significant around the Fermi edge. The excitation of hot electrons has been found to cause significantly smaller modifications of the photoemission spectrum even at high intensities. Such hot electrons can be distinguished from LAPE by the increase of the average electron kinetic energy that they cause around the Fermi edge.

In conclusion, we investigated sidebands at the Fermi edge of EUV photoelectron spectra on a clean Pt(111) sample in the presence of a strong low-frequency laser pulse. The energy separation of the sidebands together with their polarization and intensity dependence supports our interpretation as the first observation of surface laser-assisted photoelectric effect. Our data also shows that LAPE is found below pump intensities that cause other typical effects encountered in ultrafast spectroscopy on metal surfaces. Furthermore, at these peak intensities (500 GW/cm2), dynamical studies of adsorbate dynamics are typically performed without substantial laser-induced desorption [6].

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