Attosecond Photoelectron Spectroscopy of Metal Surfaces

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Attosecond Photoelectron Spectroscopy of Metal Surfaces
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Synopsis
We calculated the dynamic image potential of photoelectrons near an Al surface and show that this electronic self-interaction induces a considerable, XUV-frequency-dependent temporal shift in laser-streaked XUV photoemission spectra.

The sudden release and subsequent motion of a photoelectron (PE) in and in front of a solid dielectric medium provokes collective electron excitations in the solid. The back-interaction of these excitations with the PE can be modeled as a dynamic, wave-like redistribution of electronic density in the solid in terms of a complex-valued effective electron-self-interaction potential [1]. Its real part is due to virtual excitations of bulk and surface plasmons, while its imaginary part accounts for inelastic scattering and energy loss. We investigated how attosecond streaked photoemission [2] may be applied to reveal these dynamic many-body effects in solids [3].

We calculated the dynamic wake potential \( \delta U(z, \nu_z) \) assuming that the released PE moves with constant velocity \( \nu_z > 0 \) along a classical trajectory towards and perpendicular to the surface and crosses the metal-vacuum interface \( z = 0 \) at time \( t = 0 \), as expressed by the electron density \( \rho(r) = \delta(r)\delta(z - \nu_z t) \). We modeled the semi-infinite solid in jellium approximation [3] and included collective excitations in terms of the dispersion relations for bulk- and surface-plasmon excitations \( \omega_{kB} \) and \( \omega_{kB} \). For low momenta \( k = (k_z, k_x) \) these relations model single-plasmon modes, with bulk- and surface-plasmon frequencies \( \omega_B \) and \( \omega_B \), that decay into particle-hole excitations at high momenta. The plasmon field of the solid is then given by the Hamiltonian \( H_0 = \sum_{k_z, k_x > 0} \omega_{kB} b_{kB}^\dagger b_{kB} + \sum_{k_z, k_x} \sigma_{kB}^z a_{kB}^z a_{kB}^\dagger \), where \( b_{kB}^\dagger \) and \( a_{kB}^\dagger \) are annihilation (creation) operators for bulk and surface plasmons. The interaction between the PE and the solid is given by \( H_I = \int \, dr \rho(r) [\phi_B(r) + \phi_s(r)] \), where \( \phi_B(r) = \sum_{k_z, k_x > 0} B_{kB} b_{kB} \sin(k_z z) e^{i k_x x} \Theta(-z) + \text{h.c.} \) and \( \phi_s(r) = \sum_{k_z, k_x} A_{kB}^z a_{kB}^z e^{-ik_z z} e^{i k_x x} \Theta(z) + \text{h.c.} \) are the bulk and surface plasmon fields, \( |B_{kB}|^2 = 8\pi \omega_B^2/(V k_B^3 \omega_B) \) and \( |A_{kB}^z|^2 = \pi \omega_B^2/(S k_B^3 |\sigma_{kB}^z|) \) are the interaction strengths (with quantization volume \( V \) and surface \( S \)).

By comparing IR streaked XUV-photoemission spectra and their centers of energies, including the dynamic image potential \( \delta U(z, \nu_z) \) with calculations performed in the static limit \( \delta U(z, \nu_z = 0) \), we found a significant contribution to the temporal shift \( \Delta \tau \) in photoemission from the metal conduction band [1]. This shift is due to the excitation of bulk and surface plasmons in the metal during photoemission and depends sensitively on the XUV frequency as well as on solid state characteristics, such as the bulk (surface) plasmon frequency, IR skin depth, and PE transport in the solid. Our results suggest the use of state-of-the-art PE streaking as a tool for probing ultrafast electron dynamics in solids.

Figure 1. Streaked PE spectra for an Al surface calculated with the (a) dynamic image potential \( \delta U(z, \nu_z) \), (b) static image potential \( \delta U(z, \nu_z = 0) \) for \( \hbar \omega_X = 40 \text{ eV} \), and PE mean free path \( \lambda = 5 \text{ Å} \). (c) Corresponding center-of-energy shifts \( \delta E_{\text{COE}} \). The temporal shift between the traces \( \delta E_{\text{COE}} \) for dynamic and static image potentials is \( \Delta \tau \approx 100 \text{ as} \).

References

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