Ultrafast Low-Energy Photoelectron Diffraction for the Study of Surface-Adsorbate Interactions with 100-fs Temporal Resolution

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Various types of surface-sensitive time-domain techniques have, in the past, provided detailed and comprehensive insights into ultrafast adsorbate-surface interactions involving the charge and vibrational degrees of freedom that are relevant for surface chemical reactions. Time-domain surface electron diffraction techniques such as ultrafast low-energy electron diffraction (ULEED) hold the potential to greatly enrich this research, as they can provide quantitative and direct information on how structural orders in adsorbate layers are transiently affected by such interactions [1–3]. However, electron dispersion and Coulomb interaction broadens the probing electron pulse and considerably limits the time resolution of ultrafast electron diffraction techniques in general, and it is particularly critical for the low electron energies typically used in LEED.

In this talk, I will present a surface-sensitive and ultrafast electron diffraction experiment capable of probing structural dynamics in adsorbate layers with a temporal resolution of 100 fs. In our experiment we analyze the energy-momentum distribution of low-energy photoelectrons excited by a near ultraviolet (NUV) ultrafast laser pulse in graphite that are diffracted as they pass through an ordered tin-phthalocyanine (SnPc) adsorbate layer. We experimentally demonstrate a time resolution of this ultrafast low energy photoelectron diffraction (ULEPD) technique of 100 fs, yet limited by the pulse width of the NUV laser pulse [4]. The analysis of the transient changes in the photoelectron diffraction intensity from the SnPc overlayer indicates the excitation of the adsorbate layer on a characteristic time scale of several ps. We associate the observed changes to vibrational disorder in adsorbate layer as a result of coupling to the phonon bath in graphite, which is transiently excited during the cooling-down of the photo-excited hot carrier distribution.

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