Colloquium Announcement
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“Hybrid Inorganic/Organic Systems for Opto-Electronics”

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Nonlinear plasmonnic photoemission

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Ultrafast electron and vibrational dynamics
in hybrid materials from first principles

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Nonlinear plasmonic photoemission

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We examine the nonlinear photoemission response of silver single crystal surfaces as the photon energy is tuned through the epsilon near zero (ENZ), \( \varepsilon \sim 0 \), condition, where \( \varepsilon \) is the bulk dielectric function. At ENZ, the free electron response can no longer screen the optical field, which can penetrate a metal as a longitudinal bulk plasmon field, \( \omega_p \). By recording two-photon photoemission (2PP) spectra, we find that ENZ is the onset of a plasmonic photoemission process, which is non-Einsteinian in nature, because the photoelectron energy does not depend on \( \omega_L \), but rather on the internal \( \omega_p \). Moreover, we find that a tangential optical field can excite the plasmon response on account of an atomic scale surface corrugation. The collective longitudinal plasmonic response is detected as a peak in single particle photoelectron spectra because the plasmon decay preferentially excites electrons from the Fermi level, \( E_F \), rather than decaying evenly according to density of states into a distribution of hot electrons and holes spanning \( E_F - \omega_p \leftrightarrow E_F + \omega_p \). The preferential excitation of electrons from \( E_F \) has been predicted and attributed to time dependent screening in 1965 by Hopfield,\(^1\) and observed for silver by Horn and coworkers,\(^2\) but it challenges the diametric consensus in the plasmonic photocatalysis community that plasmon decay distributes energy democratically to single particle products. The observed plasmon decay process therefore offers a more optimistic energy transduction in plasmonic photocatalysis.


Ultrafast electron and vibrational dynamics in hybrid materials from first principles

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Understanding the dynamics of coupled electron-vibrational states in their natural sub-picosecond time scale is essential to gain insight into the fundamental processes that rule the response of materials to an ultrafast laser pulse. Real-time time-dependent density functional theory, in conjunction with Ehrenfest molecular dynamics, is becoming a popular methodology to investigate these phenomena on the nanoscale. I will demonstrate the capabilities of this approach [1] with the example of a prototypical hybrid interface formed by a hydrogenated Si nanocluster and a $p$-dopant molecule adsorbed thereon [2]. To investigate the charge-transfer dynamics in hybrid materials formed by transition metal dichalcogenide (TMDC) monolayers and C-conjugated molecules, the level alignment plays a crucial role. I will show that this quantity varies significantly with the density of the adsorbed molecules and with the composition of the TMDC, leading to different physical scenarios. Finally, I will outline our recent developments to account for screening effects of layered substrates implicitly in our calculations [3].