Hybrid Inorganic/Organic Systems for Opto-Electronics

Collaborative Research Centre 951

Colloquium Announcement

of the Collaborative Research Centre 951
"Hybrid Inorganic/Organic Systems for Opto-Electronics"

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Nonlinear plasmonic photoemission

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Ultrafast electron and vibrational dynamics in hybrid materials from first principles

Time: Thursday, 12.11.2020, 15:15

Place: The colloquium takes place online (ZOOM)

Meeting-ID: 687 6163 8786
Password: 951951

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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 ($\hbar\omega_L$) energy is tuned through the epsilon near zero (ENZ), $\varepsilon\sim0$, condition, where ε 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, ω_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 ω_L, but rather on the internal ω_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.

[1] Hopfield, J.J. Effect of Electron-Electron Interactions on Photoemission in Simple Metals. *Phys. Rev.* **139**, A419-A424 (1965).
[2] Barman, S.R. *et al.* Electronic excitations on silver surfaces. *Phys. Rev. B* **69**, 045413 (2004).

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].

[1] J. Krumland, A. M. Valencia, S. Pittalis, C. A. Rozzi, and C. Cocchi, *Understanding real-time time-dependent density-functional theory simulations of ultrafast laser-induced dynamics in organic molecules*, J. Chem. Phys. **153**, 054106 (2020).
[2] M. Jacobs, J. Krumland, A. M. Valencia, H. Wang, M. Rossi, and C. Cocchi, *Ultrafast charge transfer and vibronic coupling in a laser-excited hybrid inorganic/organic interface*, Adv. Phys. X, **5**, 1749883 (2020).
[3] J. Krumland, G. Gil, S. Corni, and C. Cocchi, *LayerPCM: An implicit scheme for dielectric screening of layered substrates*, in preparation (2020).