Observation of Long-Lived Coherence in the Metalorganic System Cobalt/Alq3

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The hybrid metalorganic interface Co/Alq3 is investigated with kinetic-energy-resolved coherent 2D photoemission spectroscopy. Long-lived coherent excited states of the adsorbate are probed via coupling to the substrate in a single-photon photoemission process.

In order to exploit the great potential of hybrid metalorganic interfaces for future applications, an understanding of where and how charge transport takes place is crucial. Here we investigate the coherent electron dynamics in optically pumped (400 nm) molecular states of the metalorganic complex tris(8-hydroxyquinolinato)aluminium (Alq3) [1] deposited on a cobalt surface (Fig. 1a) by means of time-resolved photoemission spectroscopy (Fig. 1b).

Combining established two-dimensional spectroscopy techniques with photoemission electron microscopy [2], ultrafast 2D spectra of the metalorganic interface Co/Alq3 are determined (Fig. 1c). Here, two excited states are identified with an energy spacing of about 77 meV. Their linewidths are 11 meV and 48 meV, corresponding to coherence lifetimes of about 370 fs and 87 fs, respectively. Measuring the kinetic energy of the photoelectrons, these features exist over the entire accessible energy range of about 1 eV. We explain the observed features by long-lived coherent excited states of the adsorbate that decay among others via coupling to excited electrons in the substrate giving rise to a photoemission signal that depends linearly on the 800 nm laser intensity. The appearance of such narrow spectral features indicates that electronic excitations in an individual adsorbate state can be surprisingly long-lived and thus can play an important role in determining charge-transfer efficiencies at the interface.

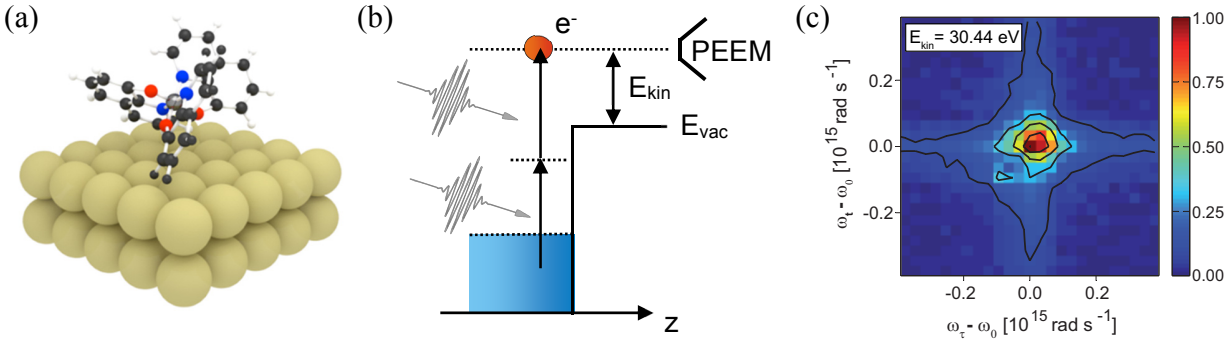

Fig.1 (a) The hybrid interface between cobalt and the metalorganic complex Alq3 is investigated with coherent 2D photoemission spectroscopy. (b) In the photoemission process electrons are excited above the vacuum potential E_{vac} and the kinetic energy E_{kin} of released electrons is resolved with PEEM. (c) Experimental 2D spectrum (absolute value) of the energy-resolved photoemission signal after phase cycling.

[1] S. Steil *et al.*, Nature Physics **9**, 242 (2013)
[2] M. Aeschlimann *et al.*, Science **333**, 1723 (2011)