ARPES spectra of doped oxides from first principles: crossover from polarons to Fermi liquids

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Abstract: The coupling of infrared-active phonons to electrons (Fröhlich interaction) can strongly affect the electronic properties of polar semiconductors and insulators. For example it can lead to the formation of polarons, i.e. electrons dressed by a phonon cloud. Conducting oxides represent an ideal playground to investigate this phenomenon. In angle-resolved photoemission spectroscopy (ARPES) the signature of polarons is the appearance of spectral satellites below the conduction band. Despite their ubiquitous role in a broad range of technologies, first-principles investigations of the electron-phonon interaction in polar materials are still relatively scarce. Here we present a general formalism for calculating the electron-phonon coupling in polar semiconductors and insulators from first principles, which can be used either as a straightforward post-processing operation or in conjunction with ab initio interpolation based on maximally localized Wannier functions [1]. We demonstrate the validity of this approach by calculating the temperature-dependent broadening of the emission linewidth in the hybrid lead halide perovskite CH3NH3PbI3 [2], and the scattering rates in anatase TiO2. We calculate ARPES spectra from first principles by combining accurate ab initio calculations of the electron-phonon coupling with the cumulant expansion method [3]. For the paradigmatic example anatase TiO2, we show that the transition from polarons to a weakly-coupled Fermi liquid with increasing doping observed in experiments [4] originates from nonadiabatic polar electron-phonon coupling. We show that a similar mechanism also applies to the ferromagnetic semiconductor EuO. Finally, we discuss how the coupling

of electrons to carrier plasmons may have similar effects to the Frohlich interaction, and we show that EuO can host tunable plasmonic polarons.

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[3] C. Verdi, F. Caruso, and F. Giustino, Nat. Commun. 8, 15769 (2017)
[4] S. Moser et al., Phys. Rev. Lett. 110, 196403 (2013).