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Ultrafast quantum modulation spectroscopy of a solid state Mott insulator

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The interplay of interactions that leads to electronic order in Mott insulators is of fundamental importance to explain several remarkable phenomena, including high-Tc superconductivity. In particular, the microscopic physics of the Hamiltonian is encapsulated in effective parameters, which include the role of all degrees of freedom to determine hopping and interaction energies. Yet, these microscopic parameters are not easily extracted from most experimental techniques, making the range of validity of a single Hubbard model often difficult to determine.
We introduce here a new type of spectroscopy that investigates microscopic couplings by detecting changes in the excitation spectrum when different low-energy modes are selectively driven. In the organic salt ET-F2TCNQ, a prototypical one-dimensional Mott insulator [1], we excite two specific local vibrational modes of the ET molecule with ultrashort mid-infrared pulses. Such technique is reminiscent of what routinely explored in optical lattices, thus bringing non-equilibrium many body dynamics of model systems to solid state compounds.
The conductivity, measured as a function of pump-probe delay over the whole infrared range, displays a pronounced red-shift of the Mott gap after excitation of a 10-μm IR active mode, due to an effective screening of the on-site correlation. Furthermore, a direct frequency modulation of the hopping leads to the formation of an intragap sideband manifold, and no Drude peak, unlike what typically observed for above gap (~eV) excitations [2,3].
A "locally-vibrating"dynamic Hubbard model is able to reproduce and explain these lineshapes, revealing a pronounced asymmetry in the behavior of holons and doublons. This is of general interest, as hole and electron doping are known to affect correlated electron systems in different ways, notably in high-Tc cuprates.
The quantum modulation spectroscopy introduced here is applicable to systematic deconstruction of the Hubbard Hamiltonian in a broad range of materials, addressing not only vibrational but also magnetic and electronic degrees of freedom.

References:
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