Orbital correlations and Mott-Hubbard excitons: electronic excitations in transition-metal oxides studied by ellipsometry

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Orbitals play a decisive role for the low-energy physics of transition-metal oxides with strong electronic correlations. Orbitals e.g. govern both the size and the sign of the super-exchange coupling between spins. The central question in orbital physics currently is under which circumstances the orbitals themselves may be considered as a truly low-energy degree of freedom with low-energy excitations, in other words, to establish compounds with strong orbital fluctuations. In most Mott-Hubbard insulators the orbitals are frozen out by the crystal field, opening a gap in the orbital excitations of a few 100 meV or larger. However, different groups have pointed out that orbital fluctuations may be strong in the Mott-Hubbard insulators RVO$_3$, claiming e.g. the observation of a one-dimensional orbital liquid, of an orbital Peierls phase, or of (bi)-orbitons Studies based on LDA+DMFT support orbital fluctuations for larger R ions such as in LaVO$_3$.

In the optical conductivity, the spectral weight of transitions from the lower to the upper Hubbard band depends sensitively on nearest-neighbor spin-spin and orbital-orbital correlations. In the case of large crystal-field splitting such as in the d^4 manganites, the temperature dependence of the spectral weight can be described quantitatively based on the spin-spin correlations. In RVO$_3$, different groups reported contradictory data sets, none of them succeeded in explaining the T dependence of the peak structure. Our data resolve this issue. Comparison with theoretical predictions based on either "classical" orbital order or strong orbital fluctuations strongly favors the latter.

The line shape of the optical conductivity is determined by the multiplet structure and excitonic effects. Interestingly, the binding energy of such a Hubbard exciton may arise both due to Coulomb attraction and due to a lowering of the kinetic energy.