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Orbital excitations in $LaMnO_3$[1]

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We have investigated the excitation spectrum of the orbital ordered $LaMnO_3$ by resonance Raman scattering and by spectral ellipsometry. By resonance Raman scattering for incident photon energies between 1.8 eV and 5.0 eV we probe the orbital exciton and its coupling to the lattice degrees of freedom. From the variation of the intensity of the first and second order scattering by the Jahn-Teller phonons as a function of incident photon energy, we derive the so called resonance profile and the corresponding exitonic states. In comparison with the dielectric function we show that the resonant behavior of the Jahn-Teller phonon cross section corresponds to the absorption due to the gap in $LaMnO_3$ at 2 eV and to charge transfer processes at 4.4 eV. We assign the 2 eV resonance behavior to the orbital exciton in contrast to lower energy-scales which have been estimated for the orbital excitation previously (E. Saitoh et al. Nature 410, 180 (2001)). The strong second order scattering of the phonons can be understood by theoretical calculations based on the Franck-Condon mechanism activating multi-phonon Raman scattering in first order of the electron-phonon coupling, strongly outlining the importance of the electron-phonon interaction. However, the estimated width of the resonance is too broad compared to the experimental results, possibly indicating other contributions such as electronic correlations within a Mott-Hubbard type picture as has been recently suggested (N.N. Kovaleva et al. Phys. Rev. Lett. 93, 147204 (2004)).

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