

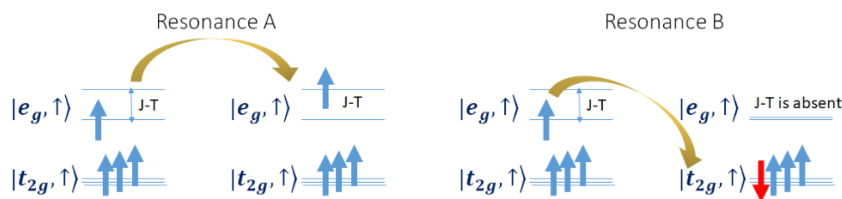
Spin, lattice and orbital couplings in manganites under electromagnetic fields

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Correlated systems display a complex interplay between different degrees of freedom (DOF). Manganites are a particularly interesting case, since charge, spin, lattice and orbital DOFs are highly intertwined. Early experiments in manganites identified d - d intersite transitions in the energy range of ~ 2 - 3 eV. Our recent investigations shed new light on the nature of these transitions, where Jahn-Teller interactions play a major role. First, by using magneto-optical spectroscopy, we show that d - d photoexcitations in $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$ split into two separate resonances, depending on whether the spin is preserved (resonance ~ 2 - 2.2 eV) or it is inverted (resonance ~ 2.6 - 2.8 eV) [1], see Figure. We further demonstrate that these resonances are a consequence of spin-orbital mixing [2], in which the spin-preserving photoinduced transfer corresponds to d - d transitions involving two e_g orbitals, while the spin-flipping resonance involves a transfer between an e_g and a t_{2g} orbital. Critically, using a group-theoretic modelling, we suggest that local Jahn-Teller distortions are essential in this process. We also performed ultrafast optical spectroscopy measurements that confirm the two resonances in the spectra, displaying periodic oscillations that match known phonon frequencies in the system. Interestingly, the two resonances exhibit coupling to different phonon modes, indicating the importance of different structural distortions, including Jahn-Teller interactions and electron-phonon coupling in the emergence of the two resonances.



Two types of intersite d - d excitations. In resonance A, the end state is Jahn-Teller (J-T). In resonance B, the end state is non J-T (or weakly).

[1] Casals et al., *Phys. Rev. Lett.* **117**, 026401 (2016)

[2] A. S. Miñarro and G. Herranz *Phys. Rev. B* **106**, 165108 (2022)