



Orbital–phonon coupling in the localized limit

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Abstract

In systems with orbital order there is an elementary excitation, orbiton, due to the breaking of orbital symmetry. It is shown that there is both a correlation *and* dynamical lattice contribution to the orbital excitation, which makes the orbiton an intrinsically mixed mode. A localized model calculation shows that in the phonon excitation spectrum weak satellites appear at the orbiton energy, in qualitative agreement with Raman experiments on LaMnO₃. © 2002 Elsevier Science B.V. All rights reserved.

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LaMnO₃ is an orbital-ordered, strongly correlated electron system. There is a long-standing prediction that in an orbital-ordered state modulations of the electronic wave functions should give rise to collective excitations. Only this year the existence of these ‘orbitons’ was experimentally verified in a Raman scattering experiment [1]. There are two distinct physical mechanisms that can cause orbital order: the electron–electron repulsion and electron–lattice interaction, where the latter gives rise to a cooperative Jahn–Teller distortion in LaMnO₃. Along the lines of this dichotomy, the observed orbitons are interpreted to be either due to electron correlations [1], or due to lattice dynamics [2]. In this contribution, I show that the orbiton- and phonon-excitation spectra bare features of both correlations and lattice vibrations.

Let us consider the two-fold degenerate manganese e_g states, with one electron per site. The electron can either be in the x^2-y^2 or $3z^2-r^2$ orbital, or in any linear combination of these two states. The interaction between neighboring orbitals is mediated by the superexchange and the electron–phonon (e–p) interaction couples the electron to the two-fold degenerate Jahn–Teller phonons that have e_g symmetry. Here we consider the system in the localized limit, which is a reliable first approximation as both orbiton and phonon dispersions are usually small compared to their on-site energies [3].

In this case, the Hamiltonian [4] reduces to

$$H_{\text{orb}}^0 = J\tau^z + 2g[\tau^z Q_3 + \tau^x Q_2] + \omega_0[q_3^\dagger q_3 + q_2^\dagger q_2], \quad (1)$$

where τ_x and τ_z are (pseudo-)spin $\frac{1}{2}$ operators for the orbital degree of freedom. The phonon operators of the so-called Q_2 and Q_3 Jahn–Teller modes are $Q_i = q_i^\dagger + q_i$ and the local orbital excitation energy is given by the orbital exchange constant J , the phonon energy is ω_0 and the electron–phonon coupling constant g . In analogy with linear spin wave theory, the orbital modes can, in general, be found by performing a Holstein–Primakov transformation [3]. We introduce the bosonic orbital operators q_1^\dagger and q_1 : $\tau^z = \frac{1}{2} - q_1^\dagger q_1$ and $\tau^x = \frac{1}{2}(q_1^\dagger + q_1)$. We see from Eq. (1) that this transformation introduces a term that is linear in the phonon mode Q_3 . This is a consequence of the long-range orbital order: the lattice deforms according to which orbital is occupied. The linear term can be gauged away by introducing $q_3 \rightarrow q_3 + \eta$, where the shift η is given by $\eta = g/\omega_0$. After this shift we find

$$H_{\text{loc}} = \bar{J}q_1^\dagger q_1 + 2gq_1^\dagger q_1 Q_3 + \omega_0[q_3^\dagger q_3 + q_2^\dagger q_2] + gQ_1 Q_2, \quad (2)$$

where $\bar{J} = J + 4g^2/\omega_0$, is the sum of the local orbital exchange energy and static phonon contribution to the crystal-field splitting. The Hamiltonian without the last term, H_{loc}^0 , is exactly solvable by the canonical transformation $\bar{H} = \exp(s)H_{\text{loc}}^0 \exp(-s)$, with $s = g/\omega_0 q_1^\dagger (q_3^\dagger + q_3)$. We define the bosonic orbital and phonon Greens functions (GFs) as 2 by 2 matrices

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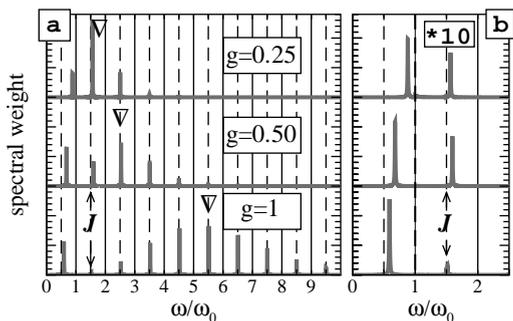


Fig. 1. (a) Orbital (Q_1) and (b) phonon (Q_2) spectral function in the localized limit. The first pole due to the orbital exchange is indicated at J . In the orbital spectrum the static crystal-field energy \bar{J} is indicated by ∇ . The spectral weight in (b) is multiplied by 10 for $\omega > \omega_0$.

$\mathbf{D}^0(v, t - t')$, where $v = 1, 2, 3$ is the index for the orbital and Q_2/Q_3 phonon mode, respectively. The matrix elements are given by $D_{11}^0(v, t - t') = -i \langle |Tq_v^\dagger(t)q_v(t')\rangle$, $D_{22}^0(v, t - t') = -i \langle |Tq_v(t)q_v^\dagger(t')\rangle$ and $D_{12}^0(v, t - t') = D_{21}^0(v, t - t') = 0$. From the canonical transformation we find that the non-interacting orbital spectral function is

$$-\frac{1}{2\pi} \text{Im} D_{22}^0(1, \omega) = e^{-4g^2/\omega_0^2} \sum_{n=0}^{\infty} \frac{2^n g^{2n}}{n! \omega_0^{2n}} \delta(\omega - J - n\omega_0). \quad (3)$$

Note that the lowest orbital excitation energy is J and the first moment of the spectral function is \bar{J} . The last term in Eq. (2) couples the orbitor and Q_2 phonon mode. We take this into account by expanding the transformed orbital operator $q_{1t} = q_1 \exp[-2g/\omega_0(q_3^\dagger - q_3)]$ for small g/ω_0 . The first term in the expansion introduces a linear mixing, which gives rise to a frequency independent self-energy. Note that the first diagrammatic correction to the self-energy is of the order $g(g/\omega_0)^2$. The self-energy Σ has the matrix elements $\Sigma_{\alpha\beta} = g$, with $\alpha = 1, 2$ and $\beta = 1, 2$. Finally, using the Dyson equation, we obtain for the interacting orbital and phonon GFs the 6 by 6 matrix

$$\mathbf{D}(\omega) = \begin{bmatrix} \mathbf{D}^0(1, \omega)^{-1} & -\Sigma & \mathbf{0} \\ -\Sigma & \mathbf{D}^0(2, \omega)^{-1} & \mathbf{0} \\ \mathbf{0} & \mathbf{0} & \mathbf{D}^0(3, \omega)^{-1} \end{bmatrix}^{-1}. \quad (4)$$

The on-site Jahn–Teller vibrational energy $\omega_0 = 80$ meV is set as unit of energy, and we take $J = 3\omega_0/2$, in accordance with Refs. [1,5]. In Fig. 1a the interacting orbital spectral function is plotted. For small e–p

coupling g , most of the spectral weight is in the pole at $\omega \approx J$, and phonon satellites with decreasing intensity are present at higher frequencies, at energy intervals ω_0 . The satellites are also known as Frank–Condon sidebands [6], and their weight increases with increasing e–p coupling strength. For larger coupling constants the average orbitor excitation energy increases, caused by the increase of the static orbital splitting \bar{J} , but low and high energy satellites are always present due to the interaction of the orbital excitation with dynamic lattice vibrations. The mixing of orbital and phonon mode gives rise to one extra phonon satellite in the orbitor spectral function, at frequencies below ω_0 . In Fig. 1b we see that, vice versa, due to the mixing a low intensity orbital satellite at $\approx J$ is present in the Q_2 phonon spectral function. The Q_2 vibrational mode softens with increasing g , in contrast to the Q_3 mode, which is not affected by the e–p interaction.

We considered localized orbital excitations coupled to Jahn–Teller phonons and find three important consequences of the orbitor–phonon coupling for systems like LaMnO₃. First, the coupling to the lattice moves the orbital excitation, on average, to higher energy. This shift has a straightforward physical meaning: it is phonon contribution to the crystal-field splitting caused by the static Jahn–Teller lattice deformation. The orbital excitation, however, strongly interacts with the Q_3 phonon, so that the orbitor is dynamically screened by the Jahn–Teller phonons, causing low and high energy satellites in the orbitor spectrum. In fact, the energy of the first pole in the orbitor spectrum—caused by electron correlations—is not affected, but rather reduced in weight. Finally, the orbital and Q_2 phonon modes mix. This implies that the true eigenmodes of the coupled orbital–phonon system have both orbital and phonon character. We therefore might interpret the weak satellites observed in recent Raman-scattering experiments on LaMnO₃, as orbitor derived satellites in the phonon spectral function.

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References

- [1] E. Saitoh, et al., Nature 410 (2001) 180.
- [2] P.B. Allen, V. Perebeinos, Nature 410 (2001) 155.
- [3] J. van den Brink, et al., Phys. Rev. B 59 (1999) 6795.
- [4] G. Khaliullin, R. Kilian, Phys. Rev. B 61 (2000) 3494.
- [5] J. van den Brink, P. Horsch, A.M. Oles, Phys. Rev. Lett. 85 (2000) 5174.
- [6] P.B. Allen, V. Perebeinos, Phys. Rev. Lett. 83 (1999) 4828.