Orbiton–phonon coupling in the localized limit

Jeroen van den Brink

Computational Materials Science and Mesa+ Institute, Faculty of Applied Physics, University of Twente, Box 217, 7500 AE Enschede, Netherlands

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 spectrabare features of both correlations and lattice vibrations.

Let us consider the two-fold degenerate manganese eg states, with one electron per site. The electron can either be in the x²–y² or 3z²–r² 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 eg 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

\[ H^0_{\text{orb}} = J \tau^z + 2q_1 \tau^z Q_1 + \tau^z Q_2 + \omega_0 [q_1^2 + q_2^2], \tag{1} \]

where \( \tau^z \) and \( \tau^z \) are (pseudo-)spin ½ operators for the orbital degree of freedom. The phonon operators of the so-called \( Q_1 \) and \( Q_2 \) Jahn–Teller modes are \( Q_i = q_i^1 + q_i \) and the local orbital excitation energy is given by the orbital exchange constant \( J \), the phonon energy is \( \omega_0 \) 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 \( q_1^i \) and \( q_1 \): \( \tau^z = q_1^z - q_1^z q_1^z - q_1^z q_1^z \). We see from Eq. (1) that this transformation introduces a term that is linear in the phonon mode \( Q_2 \).

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 \( \eta \) is given by \( \eta = g/\omega_0 \). After this shift we find

\[ H_{\text{loc}} = J q_1^1 q_1 + 2g q_1^1 q_1 Q_3 + \omega_0 [q_1^3 q_3 + q_2^2] + g Q_1 Q_2. \tag{2} \]

where \( J = J \pm 4g^2/\omega_0 \), is the sum of the local orbital exchange energy and static phonon contribution to the crystal-field splitting. The Hamiltonian without the last term, \( H^0_{\text{loc}} \), is exactly solvable by the canonical transformation \( \hat{H} = \exp (s) H^0_{\text{loc}} \exp (-s) \), with \( s = g/\omega_0 q_1^1 q_1 [q_3^2 + q_3] \). We define the bosonic orbital and phonon Greens functions (GFs) as 2 by 2 matrices.
orbital and phonon GFs the 6 by 6 matrix $Q$ term in Eq. (2) couples the orbiton and we find that the non-interacting orbital spectral function is 

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Note that the lowest orbital excitation energy is $D$ is set as unit of energy, and we take $J = 3e_0/2$, in accordance with Refs. [1,5]. In Fig. 1a the interacting orbiton spectral function is plotted. For small electron–phonon coupling $g$, most of the spectral weight is in the pole at $\omega_0 > \omega_0$, and phonon satellites with decreasing intensity are present at higher frequencies, at energy intervals $\omega_0$.

The satellites are also known as Frank–Condon sidebands [6], and their weight increases with increasing electron–phonon coupling strength. For larger coupling constants the average orbiton excitation energy increases, caused by the increase of the static orbital splitting $J$, 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 orbiton spectral function, at frequencies below $\omega_0$. In Fig. 1b we see that, vice versa, due to the mixing a low intensity orbital satellite at $\omega = J$ 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 orbiton–phonon coupling for systems like LaMnO$_3$. 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 orbiton is dynamically screened by the Jahn–Teller phonons, causing low and high energy satellites in the orbiton spectrum. In fact, the energy of the first pole in the orbiton 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 LaMnO$_3$, as orbiton derived satellites in the phonon spectral function.

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References