Inelastic X-ray Scattering Study of Orbitons in $LaMnO_3$

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Introduction

In strongly correlated electron systems, such as the colossal magnetoresistive manganites, the charge, orbital, spin, and lattice degrees of freedom all play important roles in determining the physical properties of the system. In particular, the orbital degree of freedom, which arises from the fact that there exists an orbital degeneracy of e_g electrons in Mn^{3+} ions, has drawn much attention recently. In the perovskite $LaMnO_3$, all of the Mn ions are trivalent, and the e_g orbitals in this material order spontaneously below 750K, as recently verified by resonant x-ray scattering [1]. In this orbital-ordered broken-symmetry phase, it was predicted that there should exist collective excitations, which are analogous to spin-waves in magnetic systems [2-4]. Recent Raman scattering experiments on $LaMnO_3$ have been interpreted as the first observation of such modes [5]. In this work, Saitoh and coworkers argued that the observed peaks in the 120- to ~170-meV range correspond to the "orbiton" or "orbital wave" modes, although other explanations are also possible [5]. In the current study, we have attempted to observe such excitations with inelastic x-ray scattering techniques. This would be an important confirmation of the Raman data. In addition, this technique has the crucial advantage that, if such excitations could be observed, their dispersion relations could be measured. Such measurements are essential in distinguishing the excitations from conventional phonon modes.

Methods and Materials

The experiment was carried out at SRI-CAT (3-ID beamline) with a high-resolution in-line monochromator consisting of two nested silicon channel-cut crystals. A detailed description of the beamline optics can be found in Ref. 6. The overall energy resolution of our experimental setup was 2.2 meV with an incident photon energy of 21.657 keV. A single crystal sample of $LaMnO_3$, grown with the floating-zone method, was used in our experiments.

Results

In Fig. 1, the energy spectrum obtained at the fixed Q-position of (2.2 0 0) and at T = 15K is shown. Note that the (2 0 0) position is a structural Bragg peak position.

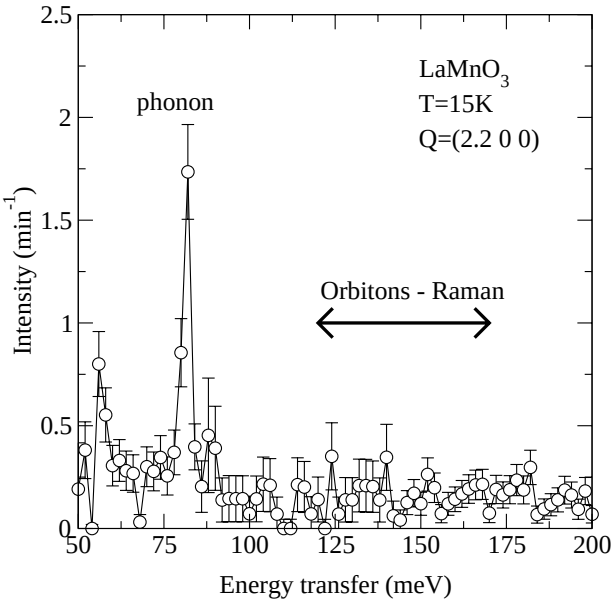

FIG. 1. Inelastic x-ray scattering scan of $LaMnO_3$ at T = 15K. The momentum transfer was fixed at (2.2 0 0). Also noted is the energy range corresponding to the orbiton modes observed in the Raman scattering study [5].

Two peaks apparent at 55 meV and 80 meV are oxygen phonon peaks corresponding to Jahn-Teller distortion mode and breathing mode, respectively. However, we could not observe any feature in the energy range where the Raman peaks were located by Saitoh et al. [6].

Discussion

There are two possible reasons that we were unable to observe the so-called orbiton scattering: (1) it is too weak to observe with the current experimental setup or (2) the orbiton is not present at these energies and there was a misinterpretation of the Raman scattering data. In regard to the first of these possibilities, it is certainly possible that the peak is much smaller than the oxygen phonon modes. Orbiton modes correspond to a single electron motion; hence, in the absence of any electron-phonon interaction, the structure factor would be much smaller than the oxygen phonon mode and unobservable. Of course, the electron-phonon interaction in this system is not small, as evidenced by the strong Jahn-Teller

distortion, and, in fact, this interpretation therefore puts an upper limit on the strength of the electron-phonon coupling in this system. In regard to the second possibility, some authors have suggested that the orbiton energy is significantly higher in this system (2 eV). In this scenario, the Raman data would perhaps be reinterpreted in terms of some multi-phonon process. Which of these two explanations is the correct one will require further experimental and theoretical effort to investigate other energy ranges and to estimate the strength of the electron-phonon coupling consistent with our experimental data. Both of these activities are presently underway.

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