Inelastic light scattering studies of superconducting Ru-based double perovskites

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Abstract

We report an inelastic light scattering study of double perovskite structure $Sr_2Y(Ru_{1-x}Cu_x)O_6$ systems as a function of temperature and doping (x = 0.0, 0.05, and 0.1). As the temperature is lowered, for x = 0.0, the 767 cm^{-1} A_{1g} apical oxygen phonon mode exhibits a softening and an enhancement of the linewidth in the antiferromagnetic phase. Similar anomalies are also observed for x = 0.1 when the superconducting gap opens. This suggests that a strong lattice-spin-charge coupling plays an important role in the magnetic and superconducting properties observed in these materials.

Key words: Infrared and Raman spectra; optical properties of bulk materials and thin films

1. Introduction

Recently, there has been a great deal of interest in Ru-based double perovskite structure $Sr_2Y(Ru_{1-x}Cu_x)O_6$ systems[1,2]. The parent compound is an antiferromagnetic insulator with Néel temperature $T_N \sim$ 26 K. When doped with Cu on Ru ion sites, the material becomes superconducting ($T_c \sim$ 5 K) for x = 0.04. More intriguingly is the observation of the coexistence of the superconductivity and magnetic ordering. These phenomena have been described within the framework of the double exchange mechanism[1].

In this paper, we describe Raman-scattering measurements aimed at exploring the low-frequency excitation spectra in the various phases of $Sr_2Y(Ru_{1-x}Cu_x)O_6$. Raman scattering is unique in that it allows the charge, spin, and lattice degrees of freedom to be monitored simultaneously through the magnetically ordered and superconducting phase transitions[3,4]. Indeed, we show in this study that this technique provides special insight into the nature of the novel properties of $Sr_2Y(Ru_{1-x}Cu_x)O_6$.

2. Experimental

Temperature-dependent Raman spectra were performed using a cold-finger cryostat and a Dilor XY 800 triple spectrometer equipped a liquid-nitrogen cooled charge-coupled device array detector. The mechanically polished surfaces of the ceramic samples were excited with 10 W/cm^2 of the 514.5 nm photons from an Ar^+ ion laser. The spectral resolution with these instruments was typically less than 1 cm^{-1}.

3. Results and Discussion

Figure 1 shows the room-temperature unpolarized Raman spectra of $Sr_2Y(Ru_{1-x}Cu_x)O_6$ (x = 0.0, 0.05, and 0.10). The spectrum of Sr_2YRuO_6 (Ru-2116) is

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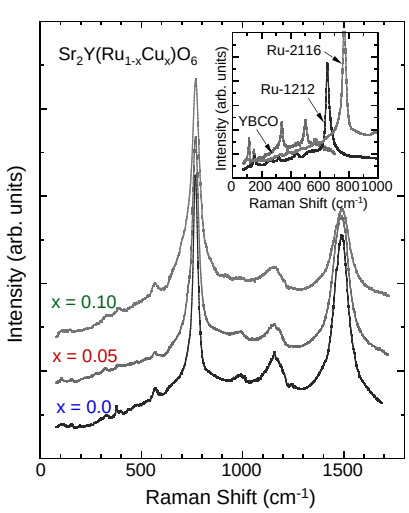

Fig. 1. Room-temperature unpolarized Raman scattering for $Sr_2Y(Ru_{1-x}Cu_x)O_6$ (x = 0.0, 0.05, and 0.10) with green (514.5 nm) excitation. For comparison, the inset shows the low-frequency Raman spectra of YBCO, Ru-1212, and Ru-2116.

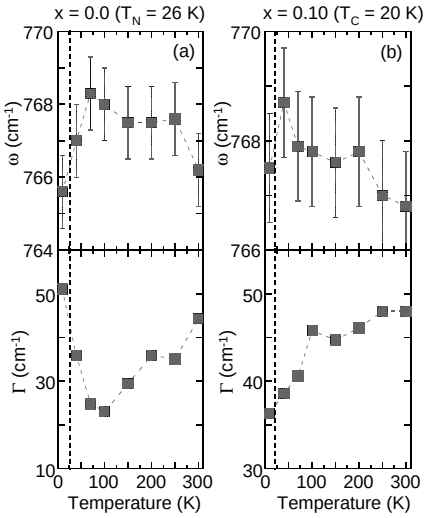

Fig. 2. Temperature dependence of the position and linewidth of the 767 cm^{-1} peak in (a) Sr_2YRuO_6 and (b) $Sr_2Y(Ru_{0.9}Cu_{0.1})O_6$. Dotted vertical lines indicate the respective critical temperatures.

composed of a strong electronic background and several optical phonon modes near 156, 328, 378, 568, 767, 998, 1160, and 1490 cm^{-1}, exhibiting slight Cu doping dependence. The exact assignment of these phonons was not possible because pure scattering geometries could not be achieved in the polycrystalline samples used. However, the intensity of the strongest peak at $\sim$ 767 cm^{-1} is much higher for parallel than for perpendicular polarization. Thus we identity this peak as the A_{1g} apical oxygen stretchinglike vibration of the RuO_6 octahedra. This mode assignment is also motivated by comparison with the similar features of apical oxygen frequencies observed at $\sim$ 504 and 654 cm^{-1} for $YBa_2Cu_3O_{7-\delta}$[3] (YBCO) and $RuSr_2GdCu_2O_8$[5] (Ru-1212), as shown in the inset of Fig. 1.

To illustrate the changes of the Raman spectra with temperature in detail, we show in Fig. 2 the temperature dependence of the fitting parameters (frequency ω and linewidth Γ) of the 767 cm^{-1} peak using a standard Lorentzian profile. As the temperature is lowered from 300 K, we first observe the expected normal hardening of the mode frequency caused by thermal contraction and a steadily decrease linewidth. However, upon crossing the magnetically ordered and superconducting phase transitions, one immediately see that the 767 cm^{-1} mode exhibits (i) a noticeable softening and (ii) a broadening for $T < T_N$ or narrowing for $T < T_c$ of the linewidth. Our results further indicate a strong modulation of the Ru-Ru exchange interaction by the apical oxygen phonon in the antiferromagnetic phase as well as the superconductivity-induced phonon self-energy effects contribute to the anomalous behavior of this mode below T_N and T_c.

Acknowledgements

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