JAHN-TELLER PHONON ANOMALY AND PHASE INHOMOGENEITIES IN La_{0.7}Ca_{0.3}MnO₃¹

J. Zhang,² P. Dai, ³ J. A. Fernandez-Baca, E. W. Plummer,⁴ Y. Tomioka,⁵ and Y. Tokura^{5,6}

It is known that the richness of the novel properties of colossal magnetoresistance (CMR) manganites originates from the strong interplay between the charge, lattice, orbital, and spin degrees of freedom found in these systems. Although several mechanisms [involving double-exchange (DE), electron-phonon interactions, and electron-electron correlations] have been proposed to explain the observed CMR effect in this class of materials, a clear understanding of its microscopic origin is still lacking. In particular, if dynamic Jahn-Teller (JT) distortions of the MnO₆ octahedra are crucial to the metal-to-insulator transition (MIT) and the CMR effect, the lattice vibrations associated with such distortions (i.e., the so-called JT phonons) would be expected to exhibit unusual behavior across the MIT/magnetic transition temperature (T_c). The Tdependence of the optical phonons in La_{1-x}Ca_xMnO₃ and in La_{1 x}Sr_xMnO₃ has been studied with infrared reflectivity and optical conductivity, and the observed energy shifts across T_c were attributed to a strong electron-phonon interaction. In this work, inelastic neutron scattering is used to demonstrate that the JT phonons in La_{0.7}Ca_{0.3}MnO₃ do indeed exhibit an abnormal behavior across the MIT/ T_c . As in the other CMR manganites, the optimally doped La_{0.7}Ca_{0.3}MnO₃ exhibits a ferromagnetic (FM) to paramagnetic (PM) transition and a simultaneous MIT at $T_c \approx 240$ K. While its structure maintains the slightly distorted cubic perovskite (orthorhombicpnma) symmetry, there are small incoherent dynamic distortions across T_c . In general, the electronphonon coupling across T_c should renormalize the phonon energies without drastically changing their intensities. This should be especially true for the high-energy optical phonons because the change in their population factors is negligible across T_c . Surprisingly, it has been discovered that one of Mn-O stretching JT phonon modes shows anomalous damping with increasing temperature in the FM phase and vanishes around T_{c} in concurrence with FM ordering and metallicity of the system (Fig. 1). Such drastic *T*-dependent damping cannot be explained with simple arguments based on the dynamic JT distortions at individual J-T sites. It is argued that this behavior may be directly related to decoherence effects because of the growing *dynamic phase fluctuations* of the short-range polaron or charge/orbital ordering associated with JT-distortion in the FM metallic phase matrix as $T \rightarrow T_{c}$.



Fig. 1. (a) Relative integrated intensities of the JT breathing phonon mode as a function of temperature at the middle of the zone $(Q = \{3.7, 1, 1\})$ and the zone boundary $(Q = \{3.5, 1, 1\})$ along the [1,0,0] direction. (b) Schematic illustration of the evolution of dynamic phase segregation with increasing temperature.

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^{2.} ORNL/UT joint faculty participant.

^{3.} UT/ORNL Distinguished Scientist Program.

^{4.} Joint Research Center for Atomic Technology (JRCAT), Tsukuba, Japan.

^{5.} University of Tokyo, Tokyo, Japan.