Low-Frequency Excitations in the Charge-Ordered Phase of (Nd_{0.5}Sr_{0.5})MnO₃

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Electronic and magnetic properties in AMnO₃ system, where A is a rare earth element, have attracted much attention because the system shows the colossal magnetoresistance. In this system, the double exchange interaction between Mn ions plays an important role. The effects of substitution in A element, which give the hole doping and/or the change of the bandwidth, have been extensively studied because it gives a drastic change of the transport, magnetic, and structural properties in the system. In the case of $(Nd_{0.5}Sr_{0.5})MnO_3$, the system is a paramagnetic insulator at $T > T_{\rm C}$ (= 250 K), a ferromagnetic metal at $T_{\rm CO}$ (= 158 K) < $T < T_{\rm C}$, and an antiferromagnetic insulator below T_{CO} . Because the lattice constants show discontinuous changes at T_{CO} , the chargeorder phase transition is a first-order one.¹ Below T_{CO} , the CE-type order of the spins and orbits was observed by the neutron diffraction measurements.² Since the phonon excitations in this system strongly interact with the magnetic ones, the study of the low-energy phonon excitations is necessary to understand the charge-order phase transition in this system. We measured Raman scattering in $(Nd_0 Sr_0 S)MnO_3$ to study the phonon excitations in this material which interacts with the spin and orbital systems.

A single crystal of $(Nd_{0.5}Sr_{0.5})MnO_3$ was prepared by the floating zone method. The symmetry of the lattice is orthorhombic at room temperature.² Below T_{CO} , the orthorhombic distortion becomes stronger.¹ The surface of the sample was polished with care to eliminate the stray light. To remove undesirable local lattice distortions due to the strain, the sample was annealed at 1050 °C for several days after the polish. All the Raman spectra were measured with a quasibackscattering geometry and we obtained the diagonal component of Raman tensor.

We observed Raman peaks at -16.9, 16.9, 79.0 and 212.0 cm⁻¹ at 15 K. The -16.9-cm⁻¹ peak is the anti-Stokes line of the 16.9-cm⁻¹ one. Above 40 K, a small peak corresponding to the anti-Stokes line of the 79.0-cm⁻¹ peak was observed. The anti-Stokes line of the 212.0-cm⁻¹ peak was not observed because of its weak intensity.

The peak frequency of the 16.9-cm⁻¹ peak decreases with increasing temperature and this mode was not observed above $T_{\rm CO}$. Therefore, this mode is thought to be the soft-phonon mode coming from the lattice distortion. But we cannot deny a possibility of the antiferromagnetic magnon, which was observed by the inelastic neutron scattering measurement in $La_{1-x}Ca_xMnO_3$.³

The frequencies and the intensities of the 79.0- and 212.0-cm⁻¹ peaks did not depend on temperature below 100 K. But at room temperature (above $T_{\rm CO}$), the 79.0-cm⁻¹ peak was not observed and all other phonon peaks in the high-frequency region, except for 212.0-cm⁻¹ peak, disappeared. At room temperature, the intensity of the 212.0-cm⁻¹ peak was very weak and its half width was very large. These results indicate that the orthorhombic distortion at room temperature is small and the symmetry of the lattice is close to that in the ideal cubic perovskite, which has no Raman-active phonon.

^{H. Kuwahara} *et al.*; Science **270** (1995) 961.
R. Kajimoto *et al.*; Phys. Rev. B **60** (1999) 9506.

³ F. Moussa et al.; Phys. Rev. B **60** (1999) 12299.