Polarized Raman scattering in single crystals of \( \text{Nd}_{0.7}\text{Sr}_{0.3}\text{MnO}_3 \)

M PATTABIRAMAN\(^1\), G RANGARAJAN\(^1\), KWANG-YONG CHOI\(^2\),
P LEMMENS\(^2\), G GUENTHERODT\(^2\), G BALAKRISHNAN\(^3\), D M\( ' \)K PAUL\(^3\)
and M R LEES\(^3\)
\(^1\)Department of Physics, Indian Institute of Technology, Chennai, India
\(^2\)II. Physikalisches Institut, RWTH, Aachen 52056, Aachen, Germany
\(^3\)Department of Physics, University of Warwick, Coventry CV4 7AL, UK
\(^*\)Email: pattu@physics.iitm.ernet.in

Abstract. We report polarized Raman scattering in single crystals of \( \text{Nd}_{0.7}\text{Sr}_{0.3}\text{MnO}_3 \). The temperature dependence of the MnO\(_6\) octahedral bending and stretching modes observed in the XX spectra points to the existence of local lattice distortions, possibly polarons. The XY spectra have been analyzed using a collision-dominated model, which allows the extraction of the carrier scattering rate.

Keywords. Raman scattering; colossal magnetoresistance; polaron; manganite.

PACS Nos 75.30.Vn; 71.38.+i; 71.30.+h

1. Introduction

It is now well-established that the physics of manganites exhibiting colossal magnetoresistance [1] is characterized by a strong interplay between spin and lattice degrees of freedom [2]. \( \text{Nd}_{0.7}\text{Sr}_{0.3}\text{MnO}_3 \) (NSMO 0.3) is one such compound, which has been the center of attention of many research groups due to its interesting magnetic and transport properties. The largest reported CMR has been observed in thin films of NSMO 0.3 (as large as \( 10^6 \)\% at 60 K and 8 T) [3]. Magnetization and dynamical studies of the time-dependent magnetization of NSMO 0.3 have shown the presence of magnetic disorder and spin frustration below \( T_C \) [4]. Neutron scattering measurements show that the spin correlation length remains approximately 20 \( \AA \) at \( T_C \) and grows to 100 \( \AA \) only at about 0.95\( T_C \) [5]. Recent NMR measurements have shown evidence for strong carrier localization below \( T_C \) resulting in a ‘phase separation’ into carrier-rich and carrier-poor regions. An understanding of the interplay between spin and lattice degrees of freedom as a function of temperature is essential to correlate all the above interesting experimental findings. Raman scattering is well suited for such a study since it has proved to be a powerful and simple tool for the detection of Jahn–Teller distortions and for the study of phase transitions in magnetic semiconductors such as the La-based manganites and EuB\(_6\) [7]. In this report we present Raman scattering measurements on single crystal samples of NSMO 0.3 from 295 K to 5 K.
2. Experimental

The single crystal was grown in an infrared image furnace by the floating zone technique. Raman measurements were performed in the spectral range of 5–1100 cm\(^{-1}\) using a CCD detector with a 5145 Å Ar laser. The temperature of the sample was varied from 5–300 K using a continuous flow cryostat.

3. Results and discussion

The XX spectra at different temperatures are shown in figure 1a. The spectra consist of peaks at 65 cm\(^{-1}\), 250 cm\(^{-1}\), 480 cm\(^{-1}\) and 660 cm\(^{-1}\). Recent lattice dynamical calculations for orthorhombic LaMnO\(_3\) [8] indicate that the 660 cm\(^{-1}\) peak corresponds to an in-phase stretching mode of the MnO\(_6\) oxygen cage where the O\(^{2-}\) ions of the \(xz\) plane vibrate along the Mn–O direction. The 480 cm\(^{-1}\) peak is associated with an out-of-phase bending mode of the oxygen cage where the O\(^{2-}\) ions vibrate perpendicular to the Mn–O line. The 480 cm\(^{-1}\) peak has been identified to couple with the \(e_g\) electron in the well studied Jahn–Teller compound LaMnO\(_3\) [9]. The low frequency modes are associated with rotations of the oxygen cage and do not involve internal distortions of MnO\(_6\).

The temperature dependence of the 65 cm\(^{-1}\), 250 cm\(^{-1}\) and 480 cm\(^{-1}\) peak positions, obtained from Gaussian fits after correcting the spectra for the background and the Bose–Einstein thermal factor \([1 + n(\omega)] = [1 - \exp(-\hbar\omega/k_B T)]^{-1}\) are shown in figures 1b, 1c and 1d. The 250 cm\(^{-1}\) peak exhibits a sudden hardening across \(T_C\) (200 K) and the 480 cm\(^{-1}\) peak exhibits a softening below \(T_C\). The hardening of the 65 cm\(^{-1}\) and 250 cm\(^{-1}\) modes represent an ordering of the MnO\(_6\) octahedra due to a reduction in the Jahn–Teller

Figure 1. (a) XX spectra. (b)–(d) Temperature dependence of the observed modes.
Polarized Raman scattering in NSMO 0.3

distortion in agreement with earlier reports [7]. It was not possible to obtain accurate Gaussian fits for the 660 cm$^{-1}$ mode as it appears as weak shoulder (figure 1a).

Neutron diffraction studies of NSMO 0.3 [10] at 300 K and 1.7 K show that the lattice parameters and the Mn–O bond lengths are weakly temperature dependent. This suggests that the anomalous temperature dependence of the 480 cm$^{-1}$ peak reflects the presence of local lattice distortions (due to the Jahn–Teller effect [11]) and does not represent a structural transition. Similar temperature dependence has been observed in the corresponding phonon mode in La$_{1.2}$Sr$_{1.8}$Mn$_2$O$_7$ [12] across $T_C$ indicating a strong spin-lattice coupling. However, the major difference between the present study and previous ones is that in NSMO 0.3 the 480 cm$^{-1}$ mode softening is observed well below $T_C$ ($T_C = 0.75T_C$) whereas in La$_{1.2}$Sr$_{1.8}$Mn$_2$O$_7$, it coincides with $T_C$. The softening of the 480 cm$^{-1}$ mode represents a reduction in the spin-lattice coupling pointing to a decrease in the lattice distortions. Since the 480 cm$^{-1}$ mode is expected to couple with the $e_g$ electron which is mobile in NSMO 0.3, the distortions too are mobile resulting in the formation of polarons.

The anomalous softening of the 480 cm$^{-1}$ mode corresponds to a reduction in the Jahn–Teller distortion and an increase in electron mobility. In NSMO 0.3 this occurs well below $T_C$ unlike the La-based manganites where such effects coincide with $T_C$ [12, 13]. Thus between 150 K and $T_C$ the Jahn–Teller distortions are the source of the strong carrier localization observed in NMR [6] and the small spin–spin correlation length observed below $T_C$ in neutron diffraction [5]. The existence of local lattice distortions and charge localization below $T_C$ suggests that the dynamic Jahn–Teller effect is the likely source of the phase separation observed in NMR below $T_C$ in NSMO 0.3.

In order to study the temperature dependence of electronic excitations we have studied the Raman response in the XY geometry (figure 2a). Previous Raman studies have shown that the low frequency response of manganites in the paramagnetic phase is ‘collision-

---

**Figure 2.** (a) XY spectra. (b) Typical fit to the collision-dominated model. (c) Temperature dependence of the carrier scattering rate.
dominated’ associated with diffusive hopping of carriers. Below $T_C$ there is a crossover to a response typical of strongly correlated metals [14]. Figure 2a shows the change in the low frequency response upon cooling across $T_C$, which is illustrative of this crossover. The XY spectra were analyzed using a collision-dominated model [14] in which the electronic Raman scattering intensity is given by

$$I(\omega) = \left[1 + n(\omega)\right]\frac{B_L\omega\Gamma_L}{\omega^2 + \Gamma^2},$$

where $B_L$ is the symmetry-dependent scattering amplitude and $\Gamma_L$ the carrier scattering rate. A typical fit is shown for the spectrum at 180 K in figure 2b. In the La-based manganites a frequency-dependent carrier scattering rate of the form $\Gamma_L = \Gamma_0 + \alpha \omega^2$, where $\alpha$ reflects electron correlation effects, has been used to improve the fitting quality. In the present study we found that it was not necessary to consider a frequency dependent form for $\Gamma_L$. This suggests that correlation effects are small in NSMO 0.3 probably due to the strong carrier localization discussed above. The temperature dependence of the scattering amplitude obtained from fitting eq. (1) to the XY spectra is shown in figure 2c. The values of the scattering rate obtained for NSMO 0.3 are much higher than that reported for Pr$_{0.63}$Sr$_{0.37}$MnO$_3$ (PSMO: $T_C = 300$ K) [14] pointing to the larger disorder in NSMO 0.3 due to its lower $T_C$. At 5 K the scattering rate for NSMO 0.3 is an order of magnitude larger than that for PSMO. The reason for such a large difference in the scattering rates at a temperature well below the $T_C$’s of both samples is at present not clear.

Acknowledgement

The work in UK was supported by a grant from the EPSRC, UK.

References

Polarized Raman scattering in NSMO 0.3


