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Polaronic metal phases in $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ uncovered by inelastic neutron and x-ray scattering

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Among colossal magnetoresistive manganites the prototypical ferromagnetic manganite $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ has a relatively small magnetoresistance, and has been long assumed to have only weak electron-lattice coupling. Here we report that $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ has strong electron-phonon coupling: Our neutron and x-ray scattering experiments show strong softening and broadening of transverse acoustic phonons on heating through the Curie temperature $T_C = 350$ K. Simultaneously, we observe two phases where metallic resistivity and polarons coexist. The ferromagnetic polaronic metal phase between 200 K and T_C is characterized by quasielastic scattering from dynamic CE-type polarons with the relatively short lifetime of $\tau \approx 1$ ps. This scattering is greatly enhanced above T_C in the paramagnetic polaronic metal phase. Our results suggest that the strength of magnetoresistance in manganites scales with the inverse of polaron lifetime, not the strength of electron-phonon coupling.

I. Introduction

In colossal magnetoresistive (CMR) manganites a transition from a ferromagnetic (FM) metallic ground state to a paramagnetic (PM) insulating phase at elevated temperatures increases with an applied magnetic field, which favors the FM phase. In the insulating phase Jahn-Teller (JT) interactions between the carriers and the atomic lattice favor local lattice distortions that trap the charge carriers [1-3]. In many manganites these lattice distortions form superstructures on heating through T_C as a result of CE-type [4,5] short-range charge and orbital order (COO) of Mn^{3+} and Mn^{4+} ions [6-8]. According to Hund's rules, three of the four $3d$ electrons of the JT active Mn^{3+} ion in $\text{La}_{1-x}\text{Sr}_x\text{Mn}^{3+}_{1-x}\text{Mn}^{4+}_x\text{O}_3$ occupy the lower t_{2g} orbitals forming a $S = 3/2$ core spin, whereas the remaining electron can choose between the two energetically degenerate e_g levels. Such a system will lower the energy of one of the e_g levels by a structural distortion of the Mn^{3+}O_6 octahedron via the JT effect [9]. For half-doped manganites with an even mix of JT active Mn^{3+} and JT inactive Mn^{4+} ions, Goodenough proposed [5] a CE-type COO ground state with a crystal lattice distortion with a wave vector of $\mathbf{q}_{\text{CE}} = (\frac{1}{4}, \frac{1}{4}, 0)$. It is observed in $\text{La}_{0.5}\text{Ca}_{0.5}\text{MnO}_3$ [10,11], $\text{LaSr}_2\text{Mn}_2\text{O}_7$ [12], and similar compounds. For smaller doping, the imbalance between Mn^{3+} and Mn^{4+} prohibits long range COO. Here, an e_g electron is able to hop from a Mn^{3+} to a Mn^{4+} ion and the intermediate oxygen atom. Due to the strong Hund's coupling of the $3d$ electrons the hopping probability is maximized for a ferromagnetic alignment of the t_{2g} Mn core spins. Due to this double exchange (DE) mechanism, ferromagnetism and an undistorted lattice win below T_C . Above T_C JT interactions favour short range COO associated with lattice distortions localizing the charge carriers [8,13,14].

Experimentally, there is a clear correlation between the magnitude of magnetoresistance and T_C : The resistivity jump increases with decreasing T_C . Standard theory correlates this T_C reduction with increasing electron-phonon coupling (EPC) strength [1,2,15,16]. In particular,

models of CMR assume that the amplitude of the lattice distortions controls how much the resistivity increases above the Curie temperature, T_C .

Among ferromagnetic manganites, $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ (LSMO) has the highest $T_C = 350$ K, and a comparatively small CMR effect [17]. LSMO undergoes a metal-metal transition at T_C as opposed to the metal-insulator transition in, e.g. $\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$ [18] and $\text{La}_{1.2}\text{Sr}_{1.8}\text{Mn}_2\text{O}_7$ [19]. So the point of view emerged that the metallicity, high T_C , and small CMR effect in LSMO, are well explained by DE physics without JT polarons [3]. However, in a recent experiment [20] some of us demonstrated that the lattice distortions in LSMO increase at T_C similarly to other manganites having a much larger magnetoresistance effect. Small polarons in LSMO were reported by pulsed neutron diffraction as well [21]. These experiments did not have energy resolution, so no information was obtained on electron-phonon effects for specific phonons nor on the spacial or temporal character of the polarons. Chen et al. [22] performed an energy-resolved investigation using neutron scattering. We will show in the following that they observed predominantly signatures of the temperature dependent renormalization of the acoustic phonons and not, as suggested in their report, of polaronic fluctuations peaked at finite energy transfers. These results motivated us to investigate the lattice dynamics of LSMO in more detail. Using direct measurements of phonons and polarons by energy-resolved neutron and x-ray scattering, we report both strong electron-phonon coupling for phonons near \mathbf{q}_{CE} and formation of dynamic polarons of CE-type in the ferromagnetic as well as paramagnetic phases.

II. Experimental

Our sample was a high-quality single crystal of $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ weighing 4 g with the FM transition temperature measured to be $T_C = 350$ K. Because magnetic intensities of the (1,0,0) Bragg peak were measured with an imperfect energy resolution, the

measured intensity includes magnetic fluctuations. Thus we assigned T_C to zero of the second derivative of the data in Fig. 4(a). Neutron scattering experiments were carried out on the 1T triplIntroductione-axis spectrometer at the ORPHEE reactor using doubly focusing PG002 monochromator and analyser crystals. The final energy was fixed at 14.7 meV. Inelastic x-ray scattering was performed at the ID-28 beam line at the European Synchrotron Radiation Facility (ESRF) in Grenoble, France. We used the Si(999) reflection with an absolute photon energy of 17.8 keV and a resulting energy resolution in our phonon scans of 2.5 meV (FWHM). All measurements were carried out in the constant-momentum-transfer \mathbf{Q} mode, $\mathbf{Q} = \boldsymbol{\tau} + \mathbf{q}$, and $\boldsymbol{\tau}$ is a reciprocal lattice point. Data were taken along the [110] high symmetry and off-symmetry directions at $\mathbf{Q} = (2 - h, 2 + h, 0)$ and $(1.5 + h, 2 + h, 0)$ [see scheme of the scattering geometry in Fig. 1(f)], respectively. The samples were mounted in a closed-cycle refrigerator at LLB and a Nitrogen gas blower system at ESRF allowing measurements at $5 \text{ K} \leq T \leq 435 \text{ K}$ and $100 \text{ K} \leq T \leq 400 \text{ K}$, respectively. The sample for x-ray scattering was prepared from a piece cut from the large sample after the neutron scattering experiments were finished. The experimental resolution in the neutron scattering experiments was obtained from standard calculations using the “*rescal*” program package [23]. In x-ray scattering, the resolution was determined by scanning the elastic line of a piece of plastic at the beginning of the experiment.

$\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ deviates from the ideal cubic perovskite structure below 750 K for $x = 0$ [24]. At $x = 0.3$, the structure in the whole temperature range investigated by us, i.e. $T \leq 450 \text{ K}$, is rhombohedral which makes the analysis of high energy phonons complicated [25]. However, it was shown that low energy modes and, in particular, acoustic ones can still be well described by a cubic shell model [26]. Hence, we adopt the cubic notation for all wave vectors for simplicity. The wave vectors are given in reciprocal lattice units (r.l.u.) of $(2\pi/a, 2\pi/b, 2\pi/c)$, where $a = b = c = 3.88 \text{ \AA}$.

III. Results

Our experiments mapped reciprocal space in the vicinity of the Σ_3 , i.e. $[1\bar{1}0]$ polarized, transverse acoustic (TA) phonons dispersing in the [110] direction at different temperatures, T . These phonons have the same symmetry as the JT distortion of the MnO_6 octahedra [14,26,27] and that of the charge order peaks observed in the insulating paramagnetic phases of CMR manganites such as $\text{La}_{1.2}\text{Sr}_{1.8}\text{Mn}_2\text{O}_7$ [13] or $\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$ [8]. The structure factor of these phonons is large next to the $(2, 2, 0)$ reciprocal lattice vector, where it can be measured in a purely transverse geometry, i.e. $\boldsymbol{\tau} \cdot \mathbf{q} = 0$ at $\mathbf{Q} = \boldsymbol{\tau} + \mathbf{q} = (2, 2, 0) + (-h, +h, 0)$ [see Fig. 1(f)]. To compare our results more closely with published results [22] we also studied scattering intensities at $\mathbf{Q} = (1.5 + h, 2 + h, 0)$, i.e. perpendicular to the direction discussed above and intersecting at $h = 0.25$.

At low temperatures our measurements reveal well defined phonon dispersions and sharp peaks [Figs. 1(a)(d)]. The TA branch at $\mathbf{q} = (-h, h, 0)$ can be easily followed up to $h = 0.3$ where the intensity is dominated by a single peak [Fig. 1(a)]. Near the zone boundary, i.e. $h \geq 0.45$, we observe two well separated peaks at 8.5 meV and 16.25 meV. Close to $h = 0.35$ raw data show an anti-crossing with a transverse optic (TO) branch in agreement with previous low temperature measurements [26]. This happens when upward and downward dispersing phonon branches of the same symmetry come close to each other. They are not allowed to cross but can exchange eigenvectors. Hence, close to the zone boundary the TA character is transferred to the nominal first TO branch, i.e. the TA character is mostly in the peak at 16.25 meV.

Our data along the $\mathbf{Q} = (1.5 + h, 2 + h, 0)$ direction at $T = 155 \text{ K}$ [Fig. 1(d)] also clearly show the TA mode. As we leave the transverse geometry with increasing $|h - 0.25|$, the TA phonon structure factor quickly decreases and we lose the peak for $h < 0.1$ and $h > 0.4$. Heating up to room temperature, we observe a weak low energy tail of the TA phonon near $h = 0.25$ [Fig. 1(b)]. Crossing T_C , the TA phonon at $\mathbf{q}_{CE} = (\frac{1}{4}, \frac{1}{4}, 0)$ acquires a pronounced low energy tail, whereas the phonons at other wave vectors, e.g. at the zone boundary ($h = 0.5$), do not strongly depend on temperature [Fig. 1(c)]. Along the off-symmetry direction, the effect is also present only close to \mathbf{q}_{CE} [Fig. 1(e)].

For a more detailed analysis, constant-momentum-transfer scans were fitted with damped-harmonic-oscillator (DHO) functions [28] convoluted with the calculated Gaussian experimental resolution and results for three different wave vectors with $h = 0.15, 0.25$ and 0.5 are shown in Fig. 2. The experimental background for each temperature was taken from the scans at the zone boundary [Figs. 2(g)-(i)] for $E \leq 6 \text{ meV}$, and near the zone center for $E \geq 12 \text{ meV}$ [Figs. 2(a)-(c)] and approximated by a straight line. At $T = 200 \text{ K}$ (Fig. 2, left column) the phonon line widths are close to the calculated resolution and the experimental background is reached on either side of the observed modes^a.

At $h = 0.5$ (the zone boundary) the phonon softens by 3% on heating to 375 K as can be expected from thermal expansion and atomic motions leading to softer bonds. The temperature dependence of the phonon intensity follows the Bose factor at this wave vector. At $h = 0.15$ and 0.25 [Figs. 2(a)-(c), (d)-(f)] the temperature dependence is clearly unconventional with the strongest anomalies at $h = 0.25$. Here, the phonon softens in the investigated temperature range by more than 10% and the

^a In our scans we see also a small signal around 8 meV ($h = 0.15$) and 10 meV ($h = 0.25$), which we ascribe to spurious scattering. It is most pronounced at low temperatures and harder to observe when the phonon broadens and thereby masks the spurious scattering. As the signal intensity is small compared to that of the TA mode it did not affect our analysis.

peak visibly broadens already at 300 K, where neutron counts do not reach the background at low energies at 300 K [Fig. 2(e)]. At 375 K [Fig. 2(f)], this effect is strongly enhanced and our DHO fit now gives finite phonon intensity even down to zero energy transfer.

The temperature dependence of the integrated phonon intensity is set by the Bose factor built into the DHO fits and gives a very good description of the measured phonon intensities at the zone boundary. For 0.25 our analysis indicates that only half of the scattering intensity at $E = 2 - 3$ meV and $T = 375$ K can be attributed to the TA phonon [Fig. 2(f)]. Put differently, phonon line broadening cannot explain all scattering intensity at low energies at $h = 0.25$. The discrepancy between data and DHO fit is smaller at $h = 0.15$ but still detectable [Fig. 2(c)]. The fits to the data can be much improved by introducing a Lorentzian centered at zero energy indicating the presence of quasielastic (QE) scattering. Unfortunately, the neutron scattering data are not sufficient to independently extract the QE scattering amplitude and line width because we could not measure below an energy transfer of 2 meV for technical reasons.

To further investigate this QE scattering, we performed high-energy resolution inelastic x-ray scattering (IXS), which has an effectively unlimited energy range. Scans at $\mathbf{Q} = (2 - h, 2 + h, 0)$, $100 \text{ K} \leq T \leq 400 \text{ K}$ and $-22 \text{ meV} \leq E \leq +22 \text{ meV}$ were analyzed assuming that (1) phonon intensities vary according to the Bose factor and (2) incoherent elastic scattering due to deviations from the perfect crystal is T -independent. Zone boundary data at $\mathbf{Q} = (1.5, 2.5, 0)$ where no quasielastic scattering is present, justify these assumptions [Figs. 3(e)-(h)]: here, fixing the elastic scattering amplitude to the low temperature value and assuming that the phonon intensity is proportional to the Bose factor and $1/\hbar\omega_{phon}$ in the phonon cross section works very well in the entire temperature range.

IXS data at $\mathbf{Q} = (1.75, 2.25, 0)$ [Figs. 3(a)-(d)] unambiguously demonstrate the presence of QE scattering for $T \geq 350 \text{ K}$. On heating from 290 K to 350 K the total scattering intensity increases by 75% whereas the Bose factor for a 9 meV phonon gives an intensity increase of only 17%, which is already included in the DHO fit functions [28]. Thus, an additional QE scattering component is necessary at elevated temperatures and we added a Lorentzian centered at zero energy to the fits. Going to negative energy transfers in the IXS spectra allowed us to independently fit the amplitude and line width of the QE contribution. After removing the effect of the resolution, the energy width of the fitted Lorentzian at 350 K is 4.15 meV (half width at half maximum, $HWHM$), which corresponds to the lifetime of $t = (1.0 \pm 0.15) \text{ ps}$. It does not vary significantly with T compared to the uncertainty. Hence, we used this line width value also to analyze the neutron data, from which it cannot be determined independently. Results for the amplitude of QE scattering from inelastic neutron and x-ray scattering agree qualitatively very well [Fig. 4(b)] and highlight two temperatures: weak quasielastic scattering appears above 200 K and its

intensity peaks at $T = 350 \text{ K}$, which is the Curie temperature determined by magnetic neutron scattering [Fig. 4(a)].

The resulting phonon energies and line widths of the TA mode at $h = 0.25$ from neutron scattering show strong softening [Fig. 4(c)] and broadening [Fig. 4(d)]. The conventional effects of thermal expansion and thermal induced disorder are much smaller as can be seen in the results for the TA zone boundary mode at $h = 0.5$ plotted in the same panels. The phonon renormalization increases for $200 \text{ K} < T < 350 \text{ K}$, the same temperature range in which QE scattering develops, and levels off at higher temperatures.

Summarizing our wave vector dependent measurements, we find that the low temperature phonon dispersion agrees well with a shell model calculation based on the cubic structure [26] near the zone center and at the zone boundary but deviates from it half-way in between [Fig. 4(e)]. It is possible that this deviation is due to the rhombohedral distortion not included in the calculation (see [26]). The detailed wave vector dependences of the phonon softening [Fig. 4(f)], broadening [Fig. 4(g)] and QE scattering [Fig. 4(h)] show that both QE scattering and phonon renormalization are strongest close to \mathbf{q}_{CE} . Hence, we argue that the effects are linked to CE-type [4,5] short-range COO fluctuations. Indeed, the wave vector range of anomalous effects along the [110] direction is the same for phonon and quasielastic scattering and we can extract a correlation length for the COO fluctuations from quasielastic scattering of $\xi = (34 \pm 4) \text{ \AA}$.

IV. Discussion

Here, we want to comment on a previous neutron scattering report on dynamic correlated polarons in LSMO [22]. This study showed that constant energy scans at $E = 6$ meV along $\mathbf{Q} = (1.5 + h, 2 + h, 0)$ have a peak at $h = 0.25$ appearing above $T = 200 \text{ K}$. This is consistent with our data [Figs. 1(d)(e)]: At low temperatures such a scan probes only the (flat) background, whereas the softening of the TA phonon leads to increasing intensities near $h = 0.25$ at $T > 200 \text{ K}$. Our detailed analysis [Figs. 2(d)-(f)] shows, however, that this increase at $E = 6$ meV is largely due to the softening of the TA phonon and to a much lesser degree caused by QE, i.e., polaron scattering. Indeed, it was reported that no CE-type correlations could be identified at zero energy transfer [22]. Our experiments with improved momentum and energy resolution, in particular in IXS, clearly demonstrate the presence of such correlations with a strong maximum near $\mathbf{q} = (\frac{1}{4}, \frac{1}{4}, 0)$ [Fig. 4(h)]. However, the contributions from TA phonons have to be analysed carefully because they overlap and can mask the polaron scattering in a substantial part of the Brillouin zone.

Strong CE-type polarons that we found in LSMO, are not as obvious as in $\text{La}_{1.2}\text{Sr}_{1.8}\text{Mn}_2\text{O}_7$ or $\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$ because they are broad in energy (≈ 4 meV, $HWHM$) and thus have a lower peak intensity. However, the

energy-integrated intensity of polaron scattering in LSMO at $T = T_C$ appears to have the same order of magnitude as in $\text{La}_{1.2}\text{Sr}_{1.8}\text{Mn}_2\text{O}_7$ at $T = 128\text{ K}$ ($T_C = 115\text{ K}$), which we studied in a previous investigation [14]. A more quantitative comparison of polaron intensities in different CMR manganites would be highly desirable in order to see whether the polaron intensity scales with the strength of the CMR effect or not. However, this is outside the scope of this report and cannot be done using published literature because results from both neutrons and x-ray TAS experiments typically do not allow quantitative comparisons of intensities from different reports. We note that neutron time-of-flight measurements are regularly put on an absolute scale using vanadium standard measurements but this would be extremely time consuming for TAS measurements. Hence, only a focused measurement on several specimens of LSMO, $\text{La}_{1-x}\text{Ca}_x\text{MnO}_3$ and $\text{La}_{2-2x}\text{Sr}_{1+2x}\text{Mn}_2\text{O}_7$ using the same experimental setup could provide this information.

Although we cannot go beyond this limited analysis concerning the intensities of polaron scattering in CMR manganites, we note that we recently reported on the increase of the mean square displacement of oxygen atoms from their average atomic position in LSMO [20]. Theories based on local physics predict that the magnitude of the resistivity jump at T_C scales with the increase of the amplitude of lattice distortions from the average structure at the phase transition [1,2]. However, our neutron diffraction results showed that the jump at T_C in LSMO is close to those observed in CMR manganites with much larger CMR effects. This result combined with our inelastic scattering suggests that polaronic distortions are of similar size and character in CMR manganites.

A key difference between $\text{La}_{1.2}\text{Sr}_{1.8}\text{Mn}_2\text{O}_7$ and LSMO is that the former has a polaron-metallic ground state [29] characterized by an optic phonon anomaly already at $T = 10\text{ K}$ [14], whereas LSMO shows no measurable electron-lattice effects at low temperature at either low or high energies [25]. The polaronic metal character of $\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$ at low temperatures has not been established or ruled out and is a subject of future work.

LSMO shows signatures of a polaronic metal only above 200 K [Figs. 4(e)-(g)]. Above T_C $\text{La}_{1.2}\text{Sr}_{1.8}\text{Mn}_2\text{O}_7$ and $\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$ become paramagnetic polaronic insulators with *static* polarons [8,14], whereas LSMO features a paramagnetic polaronic metallic state with *mobile* polarons evidenced by their finite life time. This explains why the absolute strength of magnetoresistance near T_C in LSMO and $\text{La}_{1.2}\text{Sr}_{1.8}\text{Mn}_2\text{O}_7$ differs by more than an order of magnitude although lattice distortions are of similar size [20].

Phase separation between charge-ordered (CO) and ferromagnetic metallic regions is discussed in CMR manganites since more than 15 years [30] and, recently, CMR in undoped LaMnO_3 under an applied pressure of $p = 32 - 35\text{ GPa}$ was also explained in this scenario [31]. For this scenario in LSMO, the sharp increase of the QE intensity at T_C would indicate the breakdown of the

percolation path of ferromagnetic metallic domains and the sudden increase of CO ones while some of the latter already form shortly above $T = 200\text{ K}$.

V. Conclusion

Our results suggest that strong electron-phonon coupling and polarons are a generic feature of FM manganites. Thus, the model associating the value of T_C solely with stronger or weaker electron-phonon coupling is incomplete. Our results imply the following phase diagram for LSMO: The ground state is a FM-DE metal. For $200\text{ K} \leq T \leq 350\text{ K}$ ($= T_C$) LSMO is a FM polaronic metal, whose polaronic character becomes strongly enhanced in the PM polaronic metal phase above T_C . This reconciles the need for polarons to explain CMR quantitatively in LSMO at $T \approx T_C$ [1,2] with the observation of a DE metallic ground state, which has a much higher conductivity [17] than the metallic phase in the compounds with strong CMR. Our results suggest that the strength of magnetoresistance in manganites is primarily determined by polaron mobility. Thus, a realistic model of polaron dynamics is essential to understand CMR theoretically. We hope that our work will stimulate development of such theories.

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Figure 1 (color online): (a)-(c) Color-coded contour plots of raw neutron scattering data obtained for $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ on the thermal TAS 1T, LLB, for $100 \text{ K} \leq T \leq 375 \text{ K}$. Constant \mathbf{Q} scans at $\mathbf{Q} = (2 - h, 2 + h, 0)$, $0.1 \leq h \leq 0.5$ and $\Delta h = 0.05$ r.l.u., were performed with energy steps of $\Delta E = 0.25$ meV. For readability, the intensity scale was adjusted at different temperatures according to the Bose factor of a phonon at an energy of $E = 9$ meV. (d)(e) Color-coded contour plots of raw neutron scattering taken at (d) $T = 155 \text{ K}$ and (e) $T = 375 \text{ K}$ along an off-symmetry direction $\mathbf{Q} = (1.5 + h, 2 + h, 0)$, $0 \leq h \leq 0.5$, $\Delta h = 0.05$ r.l.u. and $\Delta E = 0.25$ meV. (f) Sketch of the scattering geometry in the $(HK0)$ plane in reciprocal space. Open squares denote various Brillouin zone centers (see labels). Dots mark the position in \mathbf{Q} , where energy scans were performed shown in panels (a)-(c) (full/red symbols) and panels (d)-(e) (open/blue symbols). The arrow connects the origin with the zone center at $\boldsymbol{\tau} = (2, 2, 0)$.

Figure 2 (color online): Representative inelastic neutron scattering spectra of $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$. Energy scans at $\mathbf{Q} = (2 - h, 2 + h, 0)$ with (a)-(c) $h = 0.15$, (d)-(f) $h = 0.25$ and (g)-(i) $h = 0.5$ at $T = 100 \text{ K}$ (left), $T = 300 \text{ K}$ (middle) and $T = 375 \text{ K}$ (right). Solid (red) lines are fits consisting of a damped harmonic oscillator for the TA mode (solid & blue), a Lorentzian for quasielastic scattering described in the text (solid & black) and the estimated background (dashed, see text). The small Gaussian component (thin black line) in (a)-(f) denotes a spurious signal.

Figure 3 (color online): Representative inelastic x-ray scattering spectra of $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$. Energy scans at $\mathbf{Q} = (2 - h, 2 + h, 0)$ with (a)-(d) $h = 0.25$ and (e)-(h) $h = 0.5$ at $200 \text{ K} \leq T \leq 400 \text{ K}$. Solid (red) lines are fits consisting of a damped harmonic oscillator for the TA mode (solid & blue), a Lorentzian for quasielastic scattering (solid & black), an estimated experimental background (dashed) and a Gaussian for a small spurious signal (thin). The amplitude of the elastic scattering was fixed to the value observed at low temperatures. Phonon intensities were set as well by the low temperature observations and only allowed to vary according to the Bose factor and the factor $1/\hbar\omega_{phon}$ in the scattering cross section (for more details see text).

Figure 4 (color online): Wave vector and temperature dependences of phonon renormalization and quasielastic scattering in $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$. Temperature dependences of (a) the ferromagnetic intensity of the (100) Bragg position and (b) the quasielastic scattering intensity, (c) the observed phonon softening $\Delta E = E_{phon,5K} - E_{phon}(T)$, (d) the phonon line width Γ_{phon} at $h = 0.25$ (dots) and 0.5 (open squares). INS results for the quasielastic intensity in (b) (circles) are scaled to match the high temperature x-ray results (dots). (e) Dispersion of the TA phonon along the [110] direction at 100 K and 375 K obtained from inelastic neutron scattering. Lines represent a shell model calculation for a cubic unit cell of $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ reproduced from Ref. [27]. (f)-(h) Wave vector dependences at temperatures $100 \text{ K} \leq T \leq 375 \text{ K}$ of (f) $\Delta E(q)$, (g) $\Gamma_{phon}(q)$ (HWHM) and (h) the quasielastic intensities associated with CE-type polarons. As the character of the TA phonon is transferred to the nominal first TO near the zone boundary, we plot results of the latter for $0.4 \leq h \leq 0.5$. Data in (h) and at $T = 350 \text{ K}$ in (f) and (g) were obtained by IXS. All other results are based on INS. Solid lines are Gaussian fits to the data. Data are offset for clarity and the corresponding zeros are shown as color-coded dashed lines on the right side of the panel. Vertical dashed lines denote the position of the ordering wave vector of the CE-type COO $\mathbf{q}_{CE} = (\frac{1}{4}, \frac{1}{4}, 0)$.