LA-UR-08- 6559

Approved for public release;
distribution is unlimited.

Title:	Low Temperature Thermal Conductivity of Bilayer Lanthanum-Strontium-Manganite (La_1.2Sr_1.8Mn_2O_7)
Author(s):	F. Ronning, N. Kurita, R. Movshovich, K.J. Mcclellan, N. Hur, I. Vekhter
Intended for:	Phys. Rev. Ltrs.

Los Alamos
NATIONAL LABORATORY
EST. 1943

" Bilayer Lanthanum-Strontium-Manganite "

Low Temperature Thermal Conductivity of $La_{1.2}Sr_{1.8}Mn_2O_7$

Z=108077

F. Ronning[1], N. Kurita[1], R. Movshovich[1], K. McClellan[1], N. Hur[2], I. Vekhter[3]

Z=195163

[1]*Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA*
[2]*Department of Physics, Inha University, Incheon 402-751, Korea*
[3]*Department of Physics and Astronomy, Louisiana State University, Baton Rouge, Louisiana, 070803, USA*

(Dated: September 29, 2008)

Thermal conductivity measurements were performed on bilayer manganite $La_{1.2}Sr_{1.8}Mn_2O_7$ down to 0.1 K and up to 7.5 T. Due to the fact that ferromagnetic magnons can be largely gapped with a magnetic field, we could separate the magnon contribution to the thermal conductivity as well as the specific heat. Consequently, we find evidence for the transport of heat by 2-dimensional ferromagnetic magnons which are scattered by electrons. Assuming that the Wiedemann-Franz law is obeyed we find a self consistent analysis which shows a phonon thermal conductivity at low temperatures proportional to $T^{1.7}$. This is evidence that structurally glassy dynamics persist down to very low temperatures in the manganites.

PACS numbers:

Intro.

Coupling of spin, charge, and lattice degrees of freedom is ubiquitous in strongly correlated materials. This is strikingly manifest in the manganites, where a system with strong electron-phonon coupling undergoes a metal-insulator transition which can be tuned by magnetic field. Thermal conductivity which can transport heat by electrons, phonons, and magnons, presents a unique opportunity to study the role and coupling between these various interactions.

The bilayer manganite $La_{1.2}Sr_{1.8}Mn_2O_7$ is a particlularly unique situation, which is a 2-D ferromagnet at low temperatures which is also metallic. Thermal conductivity has proven to be a very effective tool in studying the transport properties of ferromagnets [1–6], as magnetic spin excitations may either transport heat or act as a source of scattering. We examine low temperature because there we can extract the features of the data which are unique to the reduced dimensionality of the bilayered manganite we chose to study.

Here we demonstrate that up to 50% of the heat at 0.5 K is transported by 2-D ferromagnetic magnons. By comparing thermal conductivity with specific heat measurements we are able to extract mean free paths, which provides an additional length scale of 1 μm. We are also able to separate the phonon thermal conductivity, which shows rather remarkably, similar behavior to that found in metallic and insulating amorphous glasses.

Experimental Details

Single crystals of $La_{1.2}Sr_{1.8}Mn_2O_7$ were grown by the traveling solvent floating zone method. Crystal quality was confirmed by X-ray and in-plane resistivity was in good agreement with previously published data[7]. Heat capacity was measured in a Quantum Design Physical Properties Measurement System using a quasi-adiabatic relaxation method. Thermal conductivity was measured using the standard one heater, two thermometer setup in a dilution refrigerator. The data gave identical results whether the thermometer calibrations were performed in-situ or from a separate calibration run. Au contact pads were sputtered onto the sample, and Pt leads were attached with silver epoxy. The sample dimensions for the J||a and J||c sample were l = 1.33 mm, w = 0.17 mm, t = 0.067 mm and l = 1.10 mm, w = 1.56 mm, t = 1.34 mm, respectively, with an absolute uncertainly of 15% due to the uncertainty in measuring the geometric factor. The same leads were used for both the resistivity and thermal conductivity measurements.

Figure 1 presents the in-plane and out-of-plane thermal conductivity data for $La_{1.2}Sr_{1.8}Mn_2O_7$ for several magnetic fields. The total thermal conductivity, κ is a sum of heat carried by electrons, phonons, and magnons.

$$\kappa = \kappa_{el} + \kappa_{ph} + \kappa_{mag}$$

The most striking feature of the data is that upon application of a magnetic field the in-plane thermal conductivity is dramatically suppressed (a 50% decrease at 0.5 K). Below, we describe how we separate the various contributions of the thermal conductivity to identify this effect as the result of a suppression of magnon conductivity as a function of magnetic field.

The dashed curves in figure 1 provides an estimate of κ_{el} based on the Wiedemann-Franz law ($\kappa_{el} = L_0 T/\rho$ where L_0 is the Sommerfeld coefficient), which can be seen to be a fairly negligible contribution over the majority of the measured temperature range, and is also essentially independent of the magnetic field. To separate the phonon and magnon terms we utilize the fact that in a ferromagnet the magnon spectra will be gapped by a magnetic field. Consequently, the phonon thermal conductivity will rise as a function of magnetic field if magnon scattering is significant. Indeed, this is what occurs in $La_{1.2}Sr_{1.8}Mn_2O_7$ at high temperatures in the vicinity of the metal-insulator transition[12]. However, a field-dependent decrease in the thermal conductivity must be attributed to heat transport by magnons in zero field, which is subsequently suppressed as the number of magnons available to transport heat is reduced by the opening of a gap with magnetic field. This type of behavior has been documented in a variety of ferromagnets [1–5].

We are able to rule out the possibility of both the phonon conductivity increasing concomitant with an even greater decrease of the magnon conductivity, on the basis of our c-axis transport data. The electrical resistivity perpendicular to the MnO_2 is roughly 3 orders of magnitude larger than for conduction in the plane, and thus, the charge contribution can be safely ignored. From neutron scattering work we know that the magnon dispersion is roughly 100 times stronger in-plane than perpendicular to the planes[9], and consequently, one can estimate the ratio of magnon thermal conductivity $\kappa^{ab}_{mag}/\kappa^{c}_{mag}$ as roughly 100. However, phonon dispersions, even for layered structures, are much more isotropic. As can be seen in figure 1a, the magnitude for the c-axis thermal conductivity in zero field is comparable to the in-field thermal conductivity in the plane, which we are attributing to be largely dominated by the phonon conductivity. Furthermore, there is relatively little field dependence to the thermal conductivity when heat is transported perpendicular to the planes, which suggests that κ_{ph} is indeed field independent, and that magnons do not significantly scatter phonons below 3 K.

Consequently, in figure 2 we plot the temperature dependence of the in-plane thermal conductivity for each component of heat transport. κ_{el} is obtained using the Wiedemann-Franz law. By assuming that the entire magnon conductivity has been gapped out in high fields (valid up to 1.5 K) we obtain the phonon conductivity by subtracting the small charge conduction component from the high field data $\kappa_{ph} = \kappa(5\text{T}) - \kappa_{el}$. With the sum of the field-independent phonon and electron conductivity given by the in-field data the remainder of the zero field data gives the magnon contribution present in zero field $\kappa_{mag}(0\text{T}) = \kappa(0\text{T}) - \kappa(5\text{T})$.

This now permits us to discuss the various scattering mechanisms which are responsible for the observed behavior. The charge conductivity by the electrons is in good agreement with previous reports which demonstrate a $\sqrt{T}$ dependence due to weak localization effects in a quasi-2D material [13, 14].

To further analyze the thermal conductivity data we utilize the kinetic theory formulation for the thermal conductivity which may be expressed as a sum over all possible heat carrying modes with the form:

$$\kappa = \frac{1}{D}\int Cv\ell \, d\omega \qquad (1)$$

where D is the dimensionality of the system, C is the specific heat, $v = \nabla_k E$ is the velocity of the particles, and ℓ is their mean free path.

Analysis of our heat capacity data shown in (figure 4) find that the phononic specific heat is given by

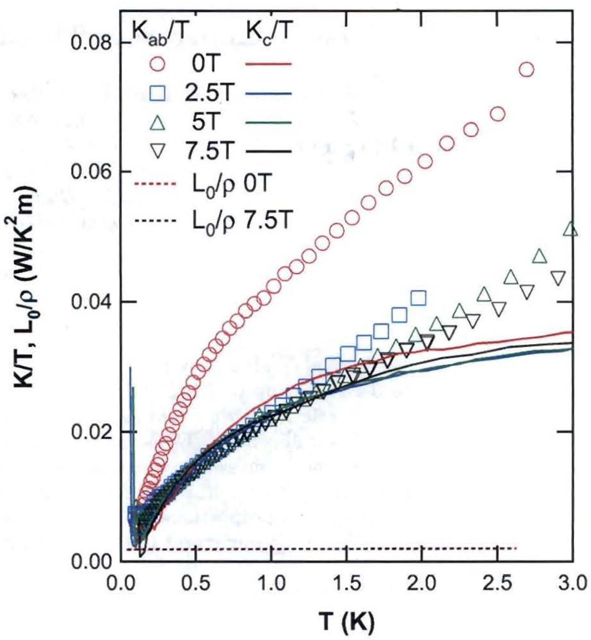

FIG. 1: (color online)(a)In-plane thermal conductivity of $La_{1.2}Sr_{1.8}Mn_2O_7$ as a function of magnetic field for J//a//H and c-axis thermal conductivity for J//c//H.

$C_{ph} = 0.28\text{mJ/molK}^4T^3$, in agreement with previous analysis[11]. In the case of phonons, there simply aren't enough phonons (or magnons) around for Umklapp scattering of phonons off of other bosonic modes to be relevant at temperatures below 3 K. Consequently, at low temperatures the mean free path is typically limited by the boundaries of the sample, in which case the familiar T^3 law for the specific heat translates into a T^3 dependence of the thermal conductivity. In our case, however, an additional scattering mechanism is involved as the low temperature thermal conductivity is more appropriately described by a different power law $\kappa_{ph} \sim T^{1.7}$. Phonons scattered by electrons produces a T^2 or T powerlaw behavior in κ_{ph} depending on whether one is in the clean ($q_{dom}\ell_e \gg 1$) or dirty limit, respectively. With an electronic mean free path of 20 Å [19] we may safely conclude that we are in the dirty limit which then does not give the correct temperature dependence. Both the magnitude and the temperature dependence found here has been observed previously in insulating and metallic glassy systems below 1 K [8], where it has been attributed to the accoustic phonons being scattered by two-level systems created by the structural glassy state. By assuming that the phonons are isotropic (an assumption supported by the c-axis thermal transport data), and using the specific heat data, we get a reasonable estimate for the mean free path as a function of temperature shown in figure 3. Note, that the presence of two-level systems should give an additional T linear contribution to the specific heat

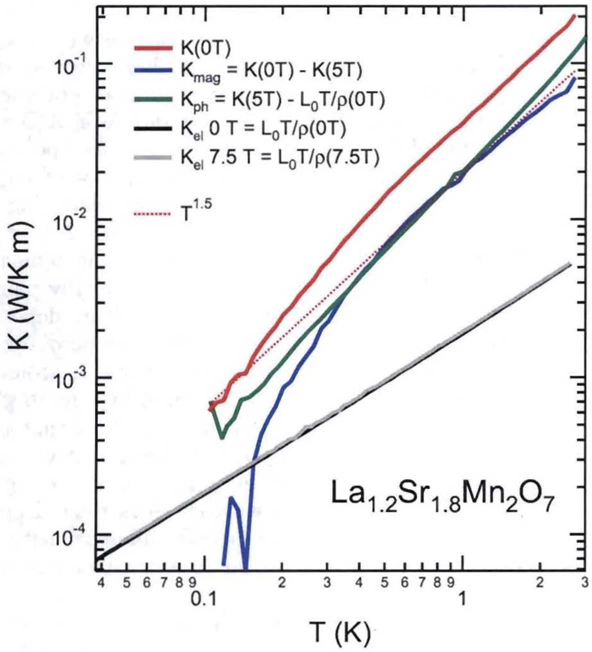

FIG. 2: (color online) Separation of the in-plane thermal conductivity of $La_{1.2}Sr_{1.8}Mn_2O_7$ into electronic, phononic, and magnon contributions.

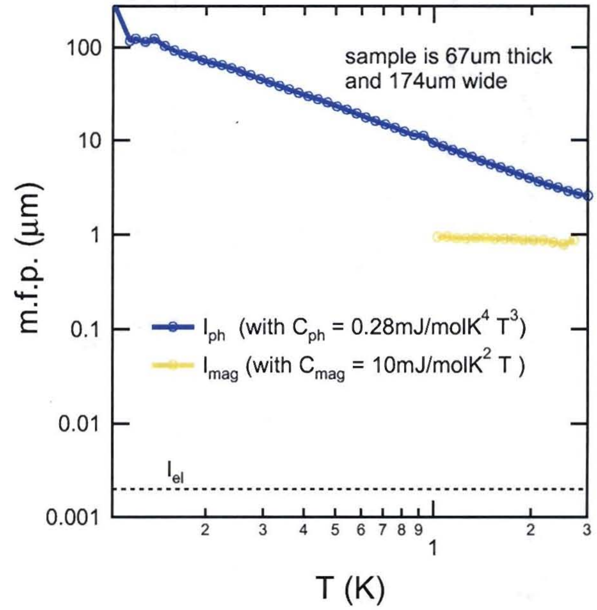

FIG. 3: (color online) mean free path of heat carrying collective excitations in $La_{1.2}Sr_{1.8}Mn_2O_7$. ℓ_{el} is obtained from ref [19].

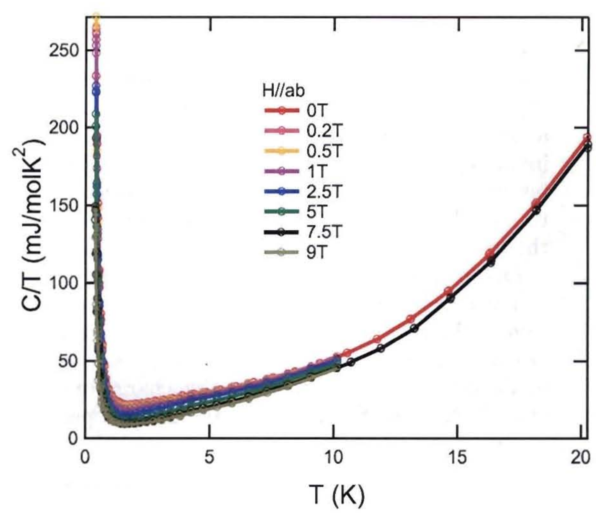

FIG. 4: (color online) raw heat capacity data of $La_{1.2}Sr_{1.8}Mn_2O_7$.

at low temperatures. However, this is estimated to be $\approx 2\mu J/gK^2T$ (cite Stephens), which is only 10% of the measured linear term which was previously attributed solely to an electronic contribution. Thus, we conlude that the phonon conductivity in $La_{1.2}Sr_{1.8}Mn_2O_7$ below 3 K is well described by phonons scattering off of two-level systems, expected for a strongly disordered lattice.

For magnons, the dispersion has been determined by neutron scattering to be $E_{mag} = \Delta + 2SJ_a(2 - cos(q_x) - cos(q_y)) + 4SJ'(1 - cos(q_z/2))$, with an effective spin S = 1.8, J_a = 4.8 meV, and J' = 0.026 meV [9, 10]. For a ferromagnet $\Delta = \Delta_0 + g\mu_B(H + 4\pi M)$, where $\Delta_0 = 0.04 \pm 0.02$ meV is a consequence of spin anisotropy[10], and magnetic field gaps the spin waves. Given this dispersion we correctly account for the heat capacity data shown in figures 4 and 5. Note that the low temperature upturn is nominally attributed to the expected nuclear Schottky contribution; however, we offer no explanation for the fact that this contribution decreases with increasing magnetic field.

With the measured magnon dispersion used as input for equation (1), and with the measured magnon thermal conductivity in zero field we can extract the mean free path, which is plotted in figure 3. Here we find a mean free path which is constant with a value of $\sim$1 μm. This value is much too small to be considered limited by the boundaries of the sample. Magnon-magnon scattering is expected to be irrelevant for temperatures below at least 0.35 J_a = 19 K[3]. Alternatively, in metallic ferromagnets which have observed magnon conductivity, the scattering appears to be well described by magnons scattering off of electrons in the dirty limit ($q\ell_e \ll 1$)[2]. From transport as well as ARPES measurements the electron mean free path is estimated to be less than 20 Å [19]. q_{dom} is obtained from the dispersion and is roughly $q_{dom} = \sqrt{(1.8k_BT/D)}$ which is approximately 1/28Å at 1 K providing evidence that we are indeed in

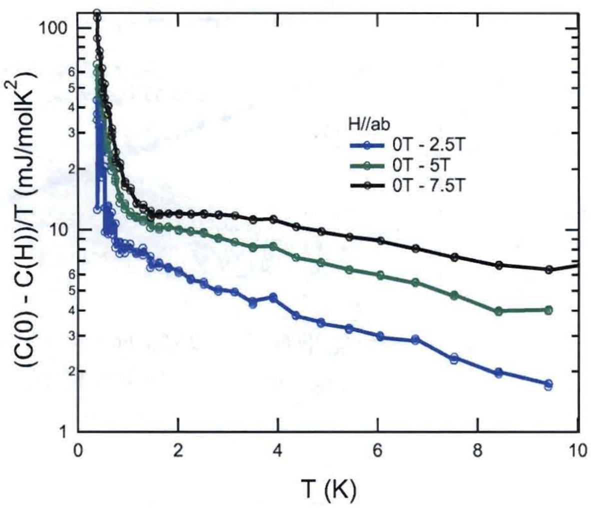

FIG. 5: (color online) Assuming that the electrons and phonons are field independent, then $C(H=0)-C(H)$ probes just the magnon and nuclear contributions. The low temperature limiting behavior is as expected for 2D magnons ($E = Dk^2$ with a spin stiffness D = 90 K. Note, naively the nuclear contribution should increase with increasing field, and thus there should be a low temperature *downturn* (not upturn).

the dirty limit. Note, that in the dirty limit, the size of the magnon conductivity is inversely related to the density of states, which is perhaps why we can observe such a sizeable contribution in this poor metal. Given the inhomogeneous nature of the manganites [18], it would be interesting to find out if the length scale of the electronic/magnetic/sturctural inhomogeneity is relevant for the heat conduction.

Discussion

The current results also have relevance for the discussion of thermal conductivity in cuprate materials, which are also layered materials often with strong amounts of disorder due to doping in the charge-reservoir layers. Both $La_{1.2}Sr_{1.8}Mn_2O_7$ and underdoped cuprates have been described as a nodal metal [17, 19], although subsequent work would seem to disagree with such an interpretation. In addition, the charge transport in $La_{1.2}Sr_{1.8}Mn_2O_7$ is very similar both in magnitude and anisotropy to underdoped cuprates, such as YBCO where a violation of the Wiedemann-Franz law was reported[17] It is argued that the phonon conductivity of cuprate materials can be explained by boundary scattering with specular reflection giving $\kappa_{ph} = bT^{\alpha}$ with $2 \leq \alpha \leq 3$ [17]. In our case the low temperature limiting behavior of $La_{1.2}Sr_{1.8}Mn_2O_7$ has $\alpha = 1.7$. The reason why phonons should possess an even stronger temperature dependent scattering rate here compared with other perovskites is unknown, but may stem from the strong polaronic nature of the manganites, which results in a more glassy like behavior than observed in cuprates. Note that glassy dynamics in $La_{1.2}Sr_{1.8}Mn_2O_7$ have been observed above the metal insulator transition by neutron scattering [20]. An interesting question is whether the strong electronic anisotropy coupled with electronic inhomogeneity (cite Davis STM) could produce an apparent violation of the Wiedemann-Franz law. In this study, we have found a self-consistent description of the data by assuming that the Wiedemann-Franz law holds over the measured temperature range. We hope that further systematic studies to lower temperatures on the manganites could address such a scenario.

In conclusion, we have demonstrated the first (to our knowledge) evidence of transport by 2-D ferromagnetic magnons in a metallic system, which is up to 50% of the total thermal conductivity at 0.5 K. Furthermore, from the phonon thermal conductivity we have presented evidence of glassy dynamics in the form of accoustic phonons scattering off of two-level systems, which supports the inhomogeneous nature of the manganites as well as the strong coupling of spin, lattice, and charge degrees of freedom.

We would like to thank M. Graf and A. Chernyshev for useful discussions. Work at Los Alamos was performed under the auspices of the US DOE.

[1] R.L. Douglass, Phys. Rev. **129**, 1132 (1963).
[2] Y. Hsu, L. Berger, Phys. Rev. B **14**, 4059 (1976).
[3] F.W. Gorter, L.J. Noordermeer, A.R. Kop, A.R. Miedema, Phys. Lett. **29A**, 331 (1969).
[4] H. Miike, J. Yoshinaga, K. Hirakawa, J. Phys. Soc. Japan **41**, 347 (1976).
[5] H.W. Gronert, D.M. Herlach, E.F. Wassermann, Europhys. Lett. **6**, 641 (1988).
[6] M. Sutherland, et. al., unpublished , (2008).
[7] Y. Moritomo, et. al., Nature **380**, 141 (1996).
[8] H.v. Löhneysen, Phys. Rep. **79**, 161 (1981).
[9] T. Chatterji, L.P. Renault, P. Thalmeier, R. Suryanarayanan, G. Dhalenne, A. Revcolevschi, Phys. Rev. B **60**, R6965 (1999).
[10] T. Chatterji, P. Thalmeier, G.J. McIntyre, R. van de Kamp, R. Suryanarayanan, G. Dhalenne, A. Revcolevschi, Europhys. Lett. **46**, 801 (1999).
[11] H. Martinho, C. Rettori, D.L. Huber, J.F. Mitchell, S.B. Oseroff, Phys. Rev. B **67**, 214428 (2003).
[12] M. Matsukawa, H. Ogasawara, R. Sato, M. Yoshizawa, R. Suryanarayanan, G. Dhalenne, A. Revcolevschi, K. Itoh, Phys. Rev. B **62**, 5327 (2000).
[13] T. Okuda, T. Kimura, Y. Tokura, Phys. Rev. B **60**, 3370 (1999).
[14] A.A. Abrikosov, Phys. Rev. B **61**, 7770 (2000).
[15] R.W. Hill, et. al., Nature **414**, 711 (2001).

[16] C. Proust, K. Behnia, R. Bel, D. Maude, S.I. Vedeneev, Phys. Rev. B, **72**, 214511 (2005).

[17] N. Doiron-Leyraud, M. Sutherland, S.Y. Li, L. Taillefer, R. Liang, D.A. Bonn, W.N. Hardy, Phys. Rev. Lett. **97**, 207001 (2006).

[18] M. Fäth, S. Freisem, A.A. Menovsky, Y. Tomioka, J. Aarts, J.A. Mydosh, Science **285**, 1540 (1999).

[19] N. Manella, W. Yang, X.J. Zhou, H. Zheng, J.F. Mitchell, J. Zaanen, T.P. Devereaux, N. Nagaosa, Z. Hussain, Z.-X. Shen, Nature **438**, 474 (2005).

[20] D.N.. Argyriou, J.W. Lynn, R. Osborn, B. Campbell, J.F. Mitchell, U. Ruett, H.N. Bordallo, A. Wildes, C.D. Ling, Phys. Rev. Lett. **89**, 036401 (2002).