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Low-temperature thermal conductivity of $BaFe_2As_2$: A parent compound of iron arsenide superconductors

N. Kurita,[1] F. Ronning,[1] C. F. Miclea,[1] E. D. Bauer,[1] J. D. Thompson,[1] A. S. Sefat,[2] M. A. McGuire,[2] B. C. Sales,[2] D. Mandrus,[2] and R. Movshovich[1]
[1]*Condensed Matter and Thermal Physics, Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA*
[2]*Materials Science and Technology Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831, USA*

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We report low-temperature thermal conductivity down to 40 mK of the antiferromagnet $BaFe_2As_2$, which is the parent compound of recently discovered iron-based superconductors. In the investigated temperature range below 4 K, the thermal conductivity κ is well described by the expression $\kappa = aT + bT^{2.22}$. We attribute the "aT"-term to an electronic contribution which is found to satisfy the Wiedemann-Franz law in the $T \rightarrow 0$ K limit and the remaining thermal conductivity, $\sim T^{2.22}$, is attributed to phonon conductivity. A small influence on thermal conductivity by magnetic fields up to 8 T is well accounted by the observed magnetoresistance. The result is consistent with a fully gapped magnon spectrum, inferred previously from inelastic neutron scattering measurements.

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I. INTRODUCTION

As in the high-T_c cuprates, with electron or hole doping, or with application of external pressure, superconductivity in the iron pnictides emerges upon suppression of the antiferromagnetism found in the parent compounds.[1–4] As to their superconductivity, in spite of a large number of experiments trying to determine the structure of the superconducting gap, the question whether superconductivity in iron pnictides is conventional *s* wave, unconventional *s* wave, or *d* wave with node(s) remains a controversial issue.[5–8] The detailed knowledge of the antiferromagnetic state in the nonsuperconducting parent compound is a necessary ingredient to elucidate how superconductivity, with transition temperatures which can reach 55 K,[9] emerges in these compounds. Soon after the discovery of superconductivity in LaFeAs(O,F),[1] it was established that the ground state of the parent compounds was a metallic collinear antiferromagnetic state.[10–13] More recently the spin-wave spectrum of several related "122" compounds crystallizing in the tetragonal $ThCr_2Si_2$ structure have been explored by inelastic neutron scattering, revealing a steep spin-wave dispersion with a gap of 6–10 meV.[14–16]

Low-temperature thermal conductivity is a sensitive probe of spin, charge, and lattice degrees of freedom. In particular, magnetic excitations, such as spin waves mentioned above can both carry heat and scatter other excitations which transport heat, such as electrons and phonons.[17–21] Furthermore, low-temperature thermal conductivity can be a powerful probe of the superconducting order parameter as recently demonstrated in nodal superconductors Tl-2201[22] and $CePt_3Si$,[23] multiband MgB_2,[24] and fully gapped Ni-based arsenic superconductor $BaNi_2As_2$.[25] However, thermal conductivity studies also allow deduction of electronic, phononic, and magnetic contributions to the parent state out of which superconductivity emerges. In this work, we report on the low-temperature thermal conductivity of $BaFe_2As_2$, which is the nonsuperconducting parent compound of recently discovered hole-doped $(Ba,K)Fe_2As_2$ (Ref. 2) and electron-doped $Ba(Fe,Co)_2As_2$ (Ref. 26) superconductors. From the lack of magnetic-field dependence, we conclude that our measurements up to 4 K are consistent with a gapped magnon spectrum. We also extract the electronic and phononic thermal conductivity that will be useful for interpreting the data of the doped compounds which become superconducting.

II. EXPERIMENTAL DETAILS

Single crystalline $BaFe_2As_2$ was grown by a self-flux method described in Ref. 27, although not from the same batch as the one used in that investigation. Thermal conductivity was measured by a standard one-heater and two-thermometers technique on a platelike crystal with dimensions of $\sim 1.5 \times 0.6 \times 0.1$ mm^3, with a heat current $q \parallel [100]$. Pt wires spot welded to the sample provided a thermal link to heater, thermometers, and the bath. The heater and RuO_2 thermometers were thermally isolated from the support frame by superconducting NbTi filaments which have a small thermal conductance at low-temperature ($\sim 10^{-10}$ W/K at 0.1 K for each thermometer or heater). Electrical resistivity was measured for electrical current $J \parallel [100]$, using the same crystal with the same electrical contacts as in thermal conductivity measurement. Thermal conductivity measurements were performed down to 40 mK and in magnetic fields up to 8 T using a dilution refrigerator with a superconducting magnet. For resistivity measurements we used a Quantum Design Physical Property Measurement System. The field orientation was $H \parallel [001]$.

III. RESULTS AND DISCUSSIONS

Figure 1 shows the temperature dependence of electrical resistivity $\rho(T)$ of $BaFe_2As_2$ in zero field and 8 T. The data are consistent with previous reports.[2,27] An anomaly at $T \approx 140$ K appears due to the simultaneous occurrence of a structural and magnetic phase transition.[2] As seen in the inset, we fit the data at 0 T below 30 K to the Fermi-liquid form $\rho = \rho_0 + AT^2$, where ρ_0 is the residual resistivity and AT^2 is ascribed to electron-electron scattering. The fit, together

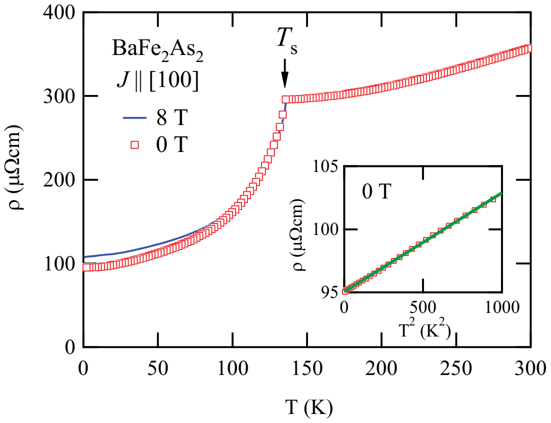

FIG. 1. (Color online) Temperature dependence of electrical resistivity $\rho(T)$ in zero field and 8 T with current direction parallel to [100]. The arrow indicates the Néel temperature T_N coincident with the temperature T_s of the structural transition. The inset shows ρ vs T^2. The straight line, which was used to estimate an electronic thermal conductivity at low-temperature, is a least-square fit to $\rho = \rho_0 + AT^2$

with the thermal conductivity data, is employed below to test the Wiedemann-Franz (WF) law. Using the value obtained $A = 0.0079\ \mu\Omega\text{cm}/\text{K}^2$ and the reported electronic specific heat coefficient $\gamma = 3\ \text{mJ}/\text{Fe-mol K}^2$,[2,27] we get a ratio $A/\gamma^2 = 8.8 \times 10^{-4}\ \mu\Omega\text{cm}/(\text{mJ}/\text{mol K})^2$ which is two order of magnitude larger than the value of 1.0×10^{-5} observed in several heavy fermion compounds. The origin of this enhanced ratio is unknown, but similar enhancement was also observed in other systems which possess strong electronic correlations.[28–30]

Next, we discuss the results of thermal conductivity measurements in BaFe$_2$As$_2$ down to 40 mK. The thermal conductivity κ can be expressed as the summation of multiple delocalized excitations which can carry heat. In this system, we anticipate possible contributions from electrons, phonons, and magnons and thus $\kappa = \kappa_{\text{el}} + \kappa_{\text{ph}} + \kappa_{\text{mag}}$. From the magnetic field dependence (shown below), we deduce that $\kappa_{\text{mag}} = 0$ in the investigated temperature range below 4 K. To separate the remaining electronic and phononic contributions, we rely on the fact that the temperature dependence of the thermal conductivity for fermions and bosons is well known in certain instances. In particular, when impurity scattering dominates the charge conduction, as Fig. 1 clearly shows to be the case for our BaFe$_2$As$_2$ sample below 20 K, it is known[31] that the thermal conductivity of electrons is proportional to T. The remaining thermal conductivity can be assigned to phonons.

Figure 2 shows the T^2 dependence of κ/T of BaFe$_2$As$_2$ in zero field. We use two different approaches to fitting the low-temperature thermal conductivity data to the form $\kappa/T = a + bT^{\alpha-1}$, where aT and bT^{α} are electronic and phonon contributions, respectively. For $\alpha = 3$, based on the conventional phonon scattering off the sample boundary (dotted curve), we fit below 0.1 K. In addition, we allow α to be a free parameter, (solid curve), which results in a good fit over the entire temperature range measured (see Fig. 2 inset). From these, we find $a = 0.0258\ \text{W}/\text{m K}^2$, $b = 0.163\ \text{W}/\text{m K}^4$ for the former and $a = 0.0252\ \text{W}/\text{m K}^2$, $b = 0.0396\ \text{W}/\text{m K}^{3.22}$, and $\alpha = 2.22$ for the latter. An electronic contribution to thermal transport is govern by the WF law, which relates charge and thermal conductivities by $\kappa/T = L_0/\rho$ with $L_0 = 2.44 \times 10^{-8}\ \text{W}\Omega/\text{K}^2$. The values of a for both fits to thermal conductivity above are in excellent agreement with the expectation for the electronic contribution $L_0/\rho = 0.0257\ \text{W}/\text{m}$ based on the value of ρ_0 obtained from the fit to the resistivity data as indicated by a dashed line. Thus, we confirm that the WF law holds in BaFe$_2$As$_2$. The remaining thermal conductivity is attributed to phonons and will be discussed in more detail below.

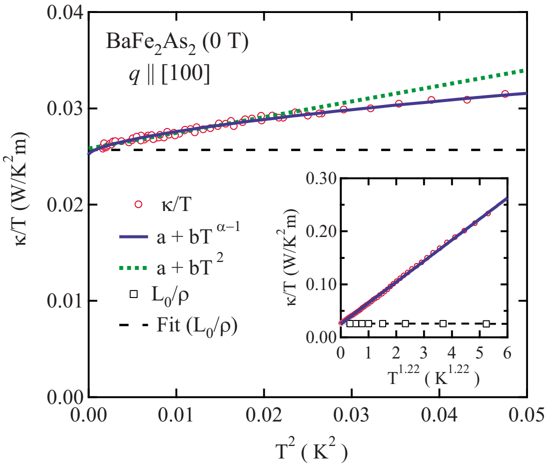

FIG. 2. (Color online) Low-temperature magnification of $\kappa(T)/T$ as a function of T^2 in BaFe$_2$As$_2$ for heat current $q \| [100]$ in zero field. The inset shows κ/T vs $T^{1.22}$ in the temperature range up to 4 K. Solid or dotted line represents a fit of the data to $\kappa = aT + bT^{\alpha}$ for $\alpha = 2.22$ or 3, respectively. Open squares in the inset correspond to the electronic thermal conductivity $\kappa_e = L_0 T/\rho$ with $L_0 = 2.44 \times 10^{-8}\ \text{W}\Omega/\text{K}^2$, derived from the resistivity data using the Wiedemann-Franz law. Dashed line is the fit from the inset of Fig. 1 in thermal units.

By applying a magnetic field up to 8 T, we find a small suppression of the thermal conductivity at low-temperatures as can be seen in Fig. 3. As shown in the inset, $\Delta\kappa/T = [\kappa/T(0) - \kappa/T(H)]$ exhibits almost constant negative values in the investigated temperature and field range. In fact, the drop in thermal conductivity is fully accounted for by a drop in electronic contribution, indicated by bold solid curves, due to magnetoresistance. The change in electronic contribution is estimated via Wiedemann-Franz analysis from the resistivity data in Fig. 1. The remaining thermal conductivity, due to a combination of phonons and magnons, is independent of magnetic field. Assuming that the phonon spectrum is field independent in BaFe$_2$As$_2$ at low-temperature, a nonsuperconducting material away from any structural or magnetic instabilities, leads to a conclusion that the magnon contribution is also field independent, i.e., magnetic excitations are gapped with $\Delta_{\text{mag}} > 4$ K. The fully gapped magnon spectrum we deduce is, hence, consistent with inelastic neutron-scattering measurements that reveal that the spin-wave spectrum of BaFe$_2$As$_2$ has a 9.8 meV gap.[14] Consequently, we would not anticipate an effect from the magnons on the thermal conductivity until roughly 100 K.

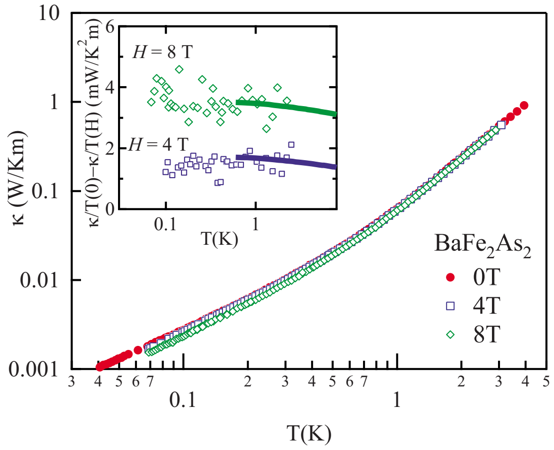

FIG. 3. (Color online) κ vs T at 0, 4, and 8 T for the field direction $H \| [001]$ and $H \perp q$. The inset shows temperature dependence of $\Delta\kappa/T = [\kappa/T(0) - \kappa/T(H)]$ for $H=4$ and 8 T. The bold solid curves represent corresponding electronic term $\Delta L_0/\rho = [L_0/\rho(0) - L_0/\rho(H)]$ derived from measured resistivity data in fields.

We return finally to the thermal conductivity remaining after subtraction of the electronic contribution, which we attribute to phonons $\kappa_{ph} = \kappa - aT$. It satisfies a single power law$=0.0396\ T^{2.22}$ over a large temperature range as can be seen in Fig. 4. At low-temperatures where these measurements are made, it is often found that a single scattering mechanism is dominant and consequently the phonon thermal conductivity obeys a simple power-law behavior $\kappa_{ph} = BT^{\alpha}$. For phonons scattering off the boundary of the crystal, one can calculate the expected thermal conductivity using the formula:[31]

$$\kappa_{ph}^{BS} = \frac{1}{3} C \langle v \rangle l_{ph}$$

where $C(\propto T^{\alpha})$ is the heat capacity of the phonons per volume, $\langle v \rangle$ their velocity (both obtained from the experimentally measured heat capacity), and l_{ph} is an effective mean-free path based on the crystal dimensions ($l_{ph} = \sqrt{4ab/\pi}$, where a and b are sample cross-section dimensions).[32] Since acoustic phonons have a low-temperature heat capacity proportional to T^3, one finds $\alpha=3$. If the faces of a crystal are smooth, one can anticipate specular reflection of the phonons[33] resulting in a lower power law typically with $\alpha \approx 2.7$. We are not aware of a single scattering mechanism, either theoretically or experimentally, over this temperature range which would give $\alpha \sim 2.22$. It should be noted that a similar magnitude of power law $\alpha \sim 2.4$ has been reported in LiF after reduction of dislocation density by annealing.[34]

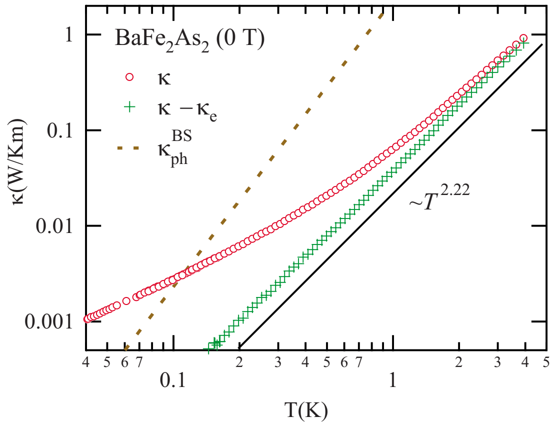

FIG. 4. (Color online) κ vs T in zero field and the extracted phonon term $\kappa_{ph} = \kappa - L_0 T/\rho$ which follows $T^{2.22}$. The phonon thermal conductivity based on boundary scattering κ_{ph}^{BS} is shown with a dashed line.

Phonons scattering off either grain boundaries or electrons are expected[31] to give $\alpha=2$ and so another possibility should be considered, that the observed power-law temperature dependence with $\alpha=2.22$ is not dominated by a single scattering mechanism, but is in fact a combination of boundary scattering, phonon, and electron scattering.

Interestingly, a similar situation occurs in the cuprates,[35] where the low-temperature phonon thermal conductivity can be fit to the form $\kappa_{ph} = BT^{\alpha}$ with α between two and three. Taillefer and co-workers[35] have argued that this empirical form provides for the most reliable determination of the low-temperature electron and magnon contributions to the thermal conductivity and suggest the origin of the T^{α} term is specular reflection off the smooth surfaces of the crystals. On the other hand, Ando and co-workers[36] maintain that the obtained mean-free path is typically of the same order as the crystal dimensions which is not consistent with specular reflection. Consequently, they attribute this strange power law simply to a crossover before the low-temperature boundary-limited scattering T^3 behavior is observed below ≈ 100 mK.

In our measurements of BaFe$_2$As$_2$, the expected boundary-scattering limit κ_{ph}^{BS} is shown as a dashed line in Fig. 4, using $C = 8.98\ \mathrm{J/K^4\ m^3} \times T^3$ from Ref. 27, $\langle v \rangle = 2400$ m/s and the $l_{ph} = \sqrt{4ab/\pi} = 323\ \mu$m, where $a = 630\ \mu$m and $b = 130\ \mu$m. The fact that it is larger than the measured κ_{ph} indicates our phonon mean-free path is less than the sample dimensions and continues to increase with decreasing temperature, consistent with the power-law exponent $\alpha < 3$. Thus, as in the cuprates, we find a phonon thermal conductivity which is well described by a single power law with $\alpha < 3$ over two decades in temperature (40 mK $\rightarrow$ 4 K) and a mean-free path slightly smaller than the crystal dimension. The utility of this parameterization will be tested in future comparisons between two families of high-temperature superconductors.

IV. CONCLUSION

In conclusion, we have performed magnetothermal conductivity experiments on the nonsuperconducting antiferromagnet BaFe$_2$As$_2$, which has a high superconducting transition temperature by electron or hole doping. The thermal conductivity κ follows $\kappa = aT + bT^{2.22}$ in a wide temperature range below 4 K. We attribute the aT-term to an electronic contribution which is consistent with the electronic conductivity expected on the basis of the Wiedemann-Franz law in the $T \rightarrow 0$ K limit and the remaining thermal conductivity

, $\sim T^{2.22}$, is attributed to phonon conductivity. A slight suppression of thermal conductivity by magnetic fields up to 8 T corresponds to the observed positive magnetoresistance and implies a fully gapped magnon spectrum in $BaFe_2As_2$. This is an important step in understanding the low-temperature thermal conductivity of the doped compounds which become superconducting.

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