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THERMAL CONDUCTIVITY OF SINGLE CRYSTAL LANTHANUM CUPRATES AT VERY LOW TEMPERATURE

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We have undertaken a study of the thermal conductivity of single crystals of pure and strontium doped lanthanum copper oxide in the temperature range 0.1-6 K. For in plane conduction, all samples show approximately a $T^{2.7}$ dependence, whereas for conduction across the planes for x = 0 we observe $\kappa \sim T^2$. This suggests that dimensionality may play an important role in the lattice heat conduction properties of these materials at very low temperature.

Some three years have passed since the discovery of superconductivity at elevated temperatures in copper-oxide perovskite systems[1]. Through a massive experimental and theoretical effort, much progress has been made in the understanding of these unusual and fascinating materials. However, a number of important questions regarding their behavior remain as yet unanswered. This situation has been in some sense abetted by the wide variety of sample fabrication techniques used to make high-T_c superconductors, which has led to the observation that many of the properties of a given sample depend on how it was produced. Quite clearly, what is needed are high quality single crystals which will exhibit more convincingly the intrinsic behavior of these systems.

Among many of the puzzling properties of these compounds is the behavior of the thermal conductivity κ at very low temperatures, i.e., below 2 K. At these temperatures, far below the superconducting critical temperature, one expects the heat conduction in these materials to be similar to that of a crystalline dielectric, namely $\kappa \sim T^3$. Experimentally, this has not been observed. In the case of sintered compounds of $YBa_2Cu_3O_7$ (hereafter referred to as the 1-2-3 compound) and $La_{2-x}Sr_xCuO_4$ (called the 2-1-4 compound), one observes a quasiquadratic or cubic temperature dependence below 10 K which evolves into a linear dependence of κ on temperature below 0.3 K[2-5]. It has been suggested[2,4,5] that this linear dependence could be due to electronic conduction either in an impurity phase of normal material or a pocket of uncondensed carriers due to anisotropic Fermi surface effects. Some measurements on single crystal 1-2-3 and $Bi_2Sr_2CaCu_2O_8$ (hereafter called the 2-2-1-2 compound), reveal an essentially T^2 behavior down to the lowest temperatures[3,6]. The similarity of this temperature dependence with that of an amorphous insulator has given rise to the suggestion[6,7] that thermal transport in these systems is essentially a tunneling process, similar to the behavior of ordinary glasses at low temperatures. A recent investigation of the thermal conductivity of 1-2-3 and 2-2-1-2 single crystals[8] interprets the temperature dependence

of κ as a combination of cubic and linear terms. Thus there is considerable variance in results and their interpretation both in sintered and single crystal specimens.

Single crystal studies such as those described above have been very limited in scope due to the difficulty in growing single crystals of sufficiently large size to perform thermal transport measurements. Recently, we have succeeded in fabricating large single crystals of the 2-1-4 compound for use in thermal conductivity measurements, and used them to measure the anisotropy of heat conduction in this system above 10 K[9]. In this report, we present the results of our measurements of the thermal conductivity made on these samples down to 0.1 K.

Samples were fabricated at the MIT Crystal Physics Laboratory; details of the technique have been given in an earlier publication[10]. We studied samples of the 2-1-4 family with nominal Sr concentrations of x = 0, 0.05, and 0.10. Electrical resistivity measurements indicated that the undoped sample was semiconducting, and the x = 0.05 and 0.10 samples were superconductors with T_c (ρ = 0) of 9 K and 33 K, respectively. Thermal conductivity measurements were performed in a dilution refrigerator. We used a doped Ge thermometer traceable to an NBS standard and metal film resistors as heaters, all of which were fastened to the samples using conductive silver paint or epoxy. We measured each sample for heat flow parallel to the Cu-O planes using a standard steady-state, two heater, one thermometer method. For the measurement of the x = 0 sample perpendicular to the Cu-O planes the sample was sandwiched between the thermal sink and a copper plate housing a heater and thermometer. The copper piece was heated, and the temperature rise was measured. Given the relatively large width of the sample (~ 1 mm) in comparison to that of the adhesive (silver paint) and the poor thermal conductivity of the sample, we assume that the thermal contact resistance is negligible compared to the thermal resistance of the sample. We thus calculate κ using the known power applied and the measured temperature gradient.

Figure 1 shows the thermal conductivity of our samples from 0.1 to 6 K. For in-plane conduction, we find that the x = 0 and x = 0.05 samples have nearly the same magnitude and temperature dependence of the conductivity; the x = 0.10 sample has a somewhat smaller conductivity but its temperature dependence ($\kappa \sim T^{2.7}$) is very similar to the other two samples. In particular, we do not observe a linear term as in the case of 2-1-4 sinters[5]. The results also differ from single crystal 1-2-3 and 2-2-1-2 compounds, where a much slower temperature dependence has been observed and interpreted either as a strictly quadratic term[3,6], or a combination of linear and cubic terms[8]. For heat conduction across the planes, we observe a much lower conductivity at 6 K (which is consistent with our earlier measurements at higher temperature), but the temperature dependence is weaker, with $\kappa \sim T^2$ over much of the range studied. The absence of a linear term

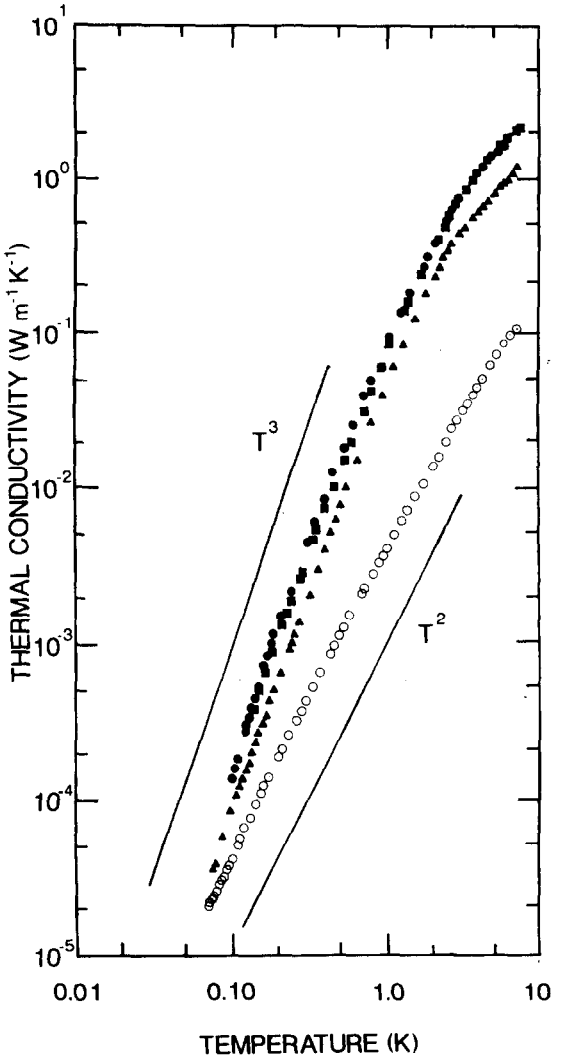

Fig. 1. Thermal conductivity of single crystal $La_{2-x}Sr_xCuO_4$. Upper three curves are for heat flow parallel to the Cu-O planes. Sample designation: ●, x = 0; ■, x = 0.05; ▲, x = 0.10. Lower curve is for heat flow perpendicular to the Cu-O planes for x = 0.

in the thermal conductivity at very low temperatures may suggest that sintered samples contain trace impurity phases which remain normal down to very low temperature whereas single crystal specimens, which are grown under more controlled conditions, do not. The nearly T^3 behavior which we observe is typical of a traditional superconductor far below T_c[11], and contrasts rather strongly with the quadratic dependences in single crystal 1-2-3 and 2-2-1-2 samples[3,6]. As evidenced by our out-of-plane results for x = 0, the phonon conduction is quite anisotropic in both magnitude and temperature dependence.

In fact, the present data for in-plane and out-of-plane conduction are strikingly similar to those of another layered system, graphite. In that case, the in-plane lattice conductivity between 1 and 10 K behaves as $\kappa \sim T^{2.7}$, while across the planes, the temperature dependence is quadratic above 1 K and cubic below this temperature[12,13]. These results have been quite successfully interpreted using the Komatsu model[14] in which both in-plane and out-of-plane phonons contribute to the conduction process and do so with differing temperature dependences. In this model, both types of modes cross over from a two-dimensional (T^2 dependence) to a three-dimensional (T^3 dependence) behavior, with a crossover temperature which is mode-dependent. Thus a possible interpretation of the different temperature dependence of the thermal conductivity in 1-2-3, 2-2-1-2, and 2-1-4 compounds is that the former two systems behave more two-dimensionally, with respect to phonon transport, than the present one, a conjecture which is qualitatively consistent with the crystal structures of these compounds. This in principle could be tested quantitatively by studying the anisotropy of κ in 1-2-3 and 2-2-1-2 samples also, and by comparing the results with a Komatsu-type model for high T_c compounds. However, the Komatsu model requires detailed knowledge of the elastic constants, which unfortunately is not yet available for any of these systems.

In conclusion, we have measured the thermal conductivity of $La_{2-x}Sr_xCuO_4$ single crystals with nominal Sr concentrations of x = 0, 0.05, and 0.10. We find that the in-plane conductivity is similar for all three concentrations, with $\kappa \sim T^{2.7}$ down to 0.1 K. Across the planes, we find $\kappa \sim T^2$ for x = 0. The nearly Debye-like behavior of the in-plane conduction differs from results on $YBa_2Cu_3O_7$ and $Bi_2Sr_2CaCu_2O_8$ single crystals. The anisotropy in the temperature dependence of the system studied here suggests that dimensional effects may play a role in the lattice conduction process. Finally, we observe no linear term in the thermal conductivity of our single crystals, as has been observed in sintered compounds of the same material. Clearly, a more comprehensive study of the anisotropy and temperature dependence of the thermal conductivity of single crystals across the entire series of high-T_c materials is essential before any firm conclusions can be drawn with regard to thermal transport in these systems.

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