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THERMAL CONDUCTIVITY OF POTASSIUM BLUE BRONZE IN THE NON-OHMIC REGIME

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The thermal conductivity of the charge-density-wave (CDW) system potassium blue bronze has been measured and found to be, within 2% uncertainty, independent of applied electric fields (E) up to 6 times the threshold field. The thermopower S(E) has also been evaluated and its field dependence is significantly weaker than that of orthorhombic TaS_3.

One of the interesting features of some charge-density-wave (CDW) systems is the non-ohmicity of their electrical conductivity σ_{dc} below the Peierls transition temperature.[1] Such a non-linearity is often described in terms of a two-fluid model which involves the normal electrons excited across a semi-conducting gap and the condensed CDW electrons. For small applied field ($E < E_T$), the CDW is pinned to the impurities or to the lattice itself and σ_{dc} is characterized by that of a semi-conductor. Above the threshold field ($E > E_T$), σ_{dc} is enhanced by the depinned CDW sliding over the pinning centers.

Other transport properties also show similar features in the non-ohmic regime. For example, a field dependent thermopower has been found in some CDW systems, although it is not clear if these observations arise directly from the sliding CDW. Stokes et.al.[2] proposed that the CDW electrons "drag" the phonon gas and therefore alter the total heat current and also the thermopower. This seems to explain the observations on $NbSe_3$[3] as well as TaS_3. On the other hand, Mihaly et.al.[4] measured the Onsager coefficient L_{12} on TaS_3 and $K_{0.3}MoO_3$ and concluded that the CDW does carry entropy.

To our knowledge, there has been no direct measurement of the field dependent thermal conductivity of blue bronze. However, Brill et.al.[5] measured the thermal conductivity of $NbSe_3$ using a self-heating method and observed that the temperature gradient across the sample remained proportional to the input power, even when the applied electric field was above E_T. The authors then concluded that the thermal conductivity is field independent.

We have used a unique technique to investigate directly the electric field dependence of the thermal conductivity κ in the CDW system $K_{0.3}MoO_3$ which undergoes a Peierls transition at T_P = 180K. The sample under study was from the same batch as the one previously used for zero field measurements[6] and was polished to a dimension of 2 x 1 x 11 mm^3. The experiment employed basically a steady-state linear heat flow method described in detail elsewhere.[7] Radiation heat losses were minimized using a stainless steel heat shield with a controlling heater on one end configured in such a way that the temperature gradient along the shield was nearly the same as that along the sample. For the field dependent electrical and thermal conductivity measurements, four gold pads were evaporated on the sample before thin copper wires were attached to it with silver paint. Direct current was swept slowly (about 6 hours per cycle) throughout the experiment so that the system was always in "steady-state". The temperature difference ΔT between two points on the sample was measured using a differential AuFe-chromel thermocouple. By switching on and off the heater at the hot end of the sample, two ΔT curves were recorded by a computer for each temperature. The set of ΔT curves at 100K is shown in Fig.1 as curves (a) and (b). A negative ΔT simply indicates that the temperature of the sample at the free end is higher than that at the base end. To assure the applied field was above E_T, the differential resistance dV/dI, shown in Fig.1 as curve (c), was measured simultaneously using a common method in which a small alternating current was superimposed on the sweeping direct current. Because of the absence of exchange gas in the sample chamber, the dV/dI curve is somewhat rounded due to the following reasons: the small amount of self-heating changes the shape of the I-V curve slightly because both the sample resistance and E_T are strongly temperature dependent; the Seebeck effect associated with the temperature gradient on the sample and leads is proportional to I^2 because the source is self-heating of the

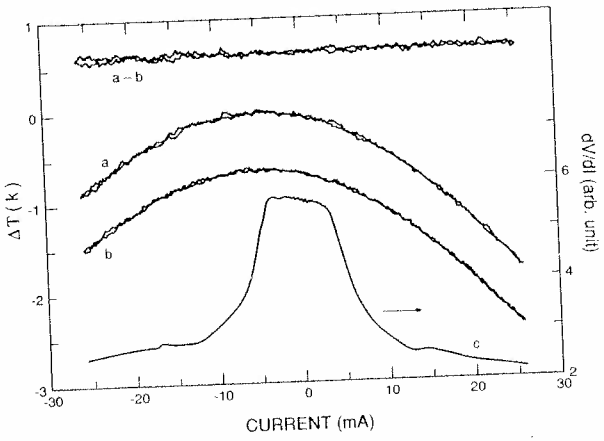

Fig. 1 A set of raw data at 100K. (a) ΔT between two points on the sample with the heater off and (b) with the heater on. A flat (a-b) curve indicates that κ is field independent. Curve (c) is the differential resistance of the sample measured simultaneously.

contacts. The rounding is not a result of the asymmetry of electrical contacts, because the sample was carefully polished on the ends and a sharp transition was observed at E_T when helium gas was introduced into the sample chamber. Nevertheless, the threshold field is clearly visible from the dV/dI curve and of a similar magnitude as commonly reported in the literature[8] ($E_T \simeq 80$, 150 and 100 mV/cm at T = 54, 100 and 147K respectively; no threshold field is detectable at T = 174K, which is near the CDW transition temperature). The maximum dc sweep is typically 3 to 6 times E_T so that the base temperature remains constant to within about 0.5%.

In the presence of an electric field, the total heat current of the system can be expressed as :

$$j^u_{total} = j^u_{ext} + j^u_{joule} + \Pi_1 j^q_n + \Pi_2 j^q_{CDW},$$

where j^u and j^q are the heat and electrical current density respectively, and Π_i are the Peltier coefficients of the normal and CDW carriers.

In terms of measurable quantities, the above equation can be rewritten as:

$$K\,\Delta T = P_0 + P_{joule} + \Pi_1 I_n + \Pi_2 I_{CDW}, \quad (1)$$

or more explicitly,

$$K\,\Delta T = P_0 + P_{joule} + \Pi_1 V/R_0 + \Pi_2 (I - V/R_0), \quad (2)$$

where K is the total thermal conductance, P_0 is the heat applied through the external heater, $I_n = V/R_0$ is the normal current, V is the voltage across the sample, R_0 is the zero-field sample resistance, $I_{CDW} = (I - I_n)$ is CDW current, and P_{joule} is the self-heating from the contact and sample resistance.

The difference between the measured ΔT with and without the external heat would then simply be :

$$K\,\delta(\Delta T) = P_0. \quad (3)$$

We have made no assumption on the field dependence of K nor on the nature of P_{joule} and Π_2. A field independent $\delta(\Delta T)$, as indicated by the constant value for curve (a-b) in Fig.1, implies that κ is field independent within the resolution of our experiment of about 2%. In fact, the value of κ has been evaluated using equation (3), together with the known value of P_0 and measured $\delta(\Delta T)$. The result is plotted in Fig.2 along with the zero field measurement reported earlier.[6] The excellent agreement shown in Fig.2 supports our conclusion.

Below the threshold field, equation (1) becomes $K\,\Delta T = P_0 + I^2 R_C + \Pi_1 I$, where R_C is a fitting parameter associated with the effective resistance. By fitting the ΔT curves to a quadratic function of I, one gets the Peltier coefficient in addition to the thermal conductivity. Further, since $\Pi_1 = TS$ in this small-field limit, one can evaluate the thermopower S as well. The calculated S(T) is plotted in Fig.3 along with the data measured by conventional methods.[9,10] The deviation at 54K is primarily due to a relatively small signal to noise ratio arising from the experimental necessity for a small (± 50 μA) swept direct current.

The fitting parameter R_C is dominated by the contact resistance at the heater end of the sample. The contact resistance at the other end of the sample does not contribute to the joule heating because it is thermally anchored to the copper heat reservoir. Also, the sample resistance does not contribute significantly to ΔT since the power dissipates rather uniformly along the sample except close to the end where it connects to the thermal reservoir. Using the known contact and sample resistances along with the previously measured thermal conductivity of blue bronze[6], we have estimated that the sample resistance accounts for <10% of the measured ΔT. This is consistent with the experimental results. For example, at

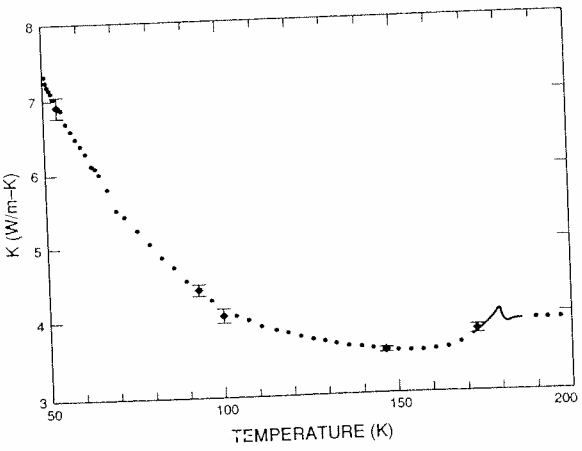

Fig. 2 The zero field κ reported previously [dots] and the calculated κ using eqn.(3) [diamond]. The error bar reflects the noise to signal ratio.

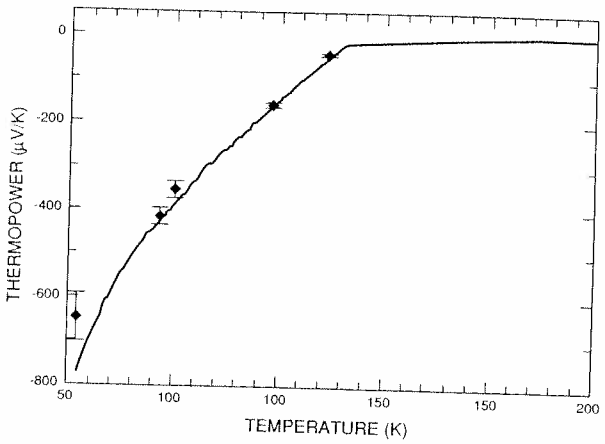

Fig. 3 Thermopower measured by a conventional method [solid curve] and by fitting the data as discussed in text [diamond]. The error bar shows the deviation of the parameter from different fits.

T = 100K, the contact resistance between the current lead and the hot end of the sample was estimated to be 4.2 ohms by measuring the resistance of different combinations of the four leads. The agreement of this measured R_C to the fitting value of 4.5 ohms confirmed that the contact resistance was indeed the major contribution to the joule heating term in equation (1).

The contact resistance is observed to scale approximately like the sample resistance in small applied field. Assuming R_C scales like the sample resistance below and above E_T, we fit our data to equation (3) with $\Pi_2=0$ and $\Pi_2/T=C_1+C_2E$ as reported in ref.(4). Neither of these fits well to our data, suggesting that R_C is field independent[11]. Therefore, it is reasonable to treat R_C as a constant throughout the fit.

Taking R_C as a constant and $\Pi_2=0$ in equation (3) also works well above E_T. In fact, the Peltier coefficient obtained from fitting the whole curve is within a few percent deviation from that of the small-field fit. This suggests that blue bronze might have a rather weak field dependent thermopower. For instance, if the field dependence were as strong for blue bronze as for TaS_3, we estimate from ref.(3) that at 100K (T/T_P= 0.56), a change of ΔT (between the high-field data and the low-field fit), due to the excess heat current at $E \simeq 6E_T$, would be about 0.7K and asymmetric about zero current. Such a deviation has not been observed in our experiment. Note that we have taken into account that the Peierls transition is at 215K and $E_T(0.56T_P) \simeq 450$mV/cm for TaS_3[12] while those for $K_{0.3}MoO_3$ are 180K and 150mV/cm respectively.

In arriving at equation (3), we have made use of the fact that neither K, R_C nor Π_i vary significantly over a small temperature interval of about 0.5K. This is justified by calculating the first order corrections to equation (3) which are found to be negligible. For example, at T=100K, the term $I_n[d\Pi_1/dT]\delta T$ only gives rise to $\leq$ 0.1% increase of the total power or equivalently, an increase in temperature of $\leq$ 0.6mK. The term Rc decreases by about 0.5% estimated from the fitting parameters and K is rather weakly temperature dependent as shown in Fig. 2. The total change in ΔT is about 2 to 3mK and is undetectable within the sensitivity of our apparatus.

In summary, the thermal conductivity of potassium blue bronze is measured directly up to 6 times E_T and found to be field independent within our 2% uncertainty. However, our data cannot exclude the observed empirical expression[4] $\Pi_2/T=C_1(T)+C_2(T)E$. Although our experiment was not aimed to resolve the controversy on the magnitude of Π_2, it suggests the thermopower of $K_{0.3}MoO_3$ is weakly field dependent relative to other CDW systems and is consistent with ref.(4) in which the phonon-drag contribution to the thermopower of the blue bronze is shown to be about two orders of magnitude smaller than that of TaS_3 at 80K. Such a difference implies either that the CDW couples stronger to the heat-carrying phonons in TaS_3 than in blue bronze or that the thermal conductivity of TaS_3 is at least an order of magnitude higher than that of blue bronze. The latter will be tested by similiar experiment.

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