THERMAL CONDUCTIVITY OF QUASI-ONE DIMENSIONAL CONDUCTORS *

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ABSTRACT

The thermal conductivity of $K_{0.3}MoO_3$, $(TaSe_4)_2I$ and TaS_3 have been measured by two different methods and are found to be independent, within experimental uncertainties, of applied electric field. The enhanced zero-field thermal conductivity observed at the Peierls transition is possibly from extra heat carried by the soft mode associated with the structural distortion.

Since the discovery of quasi-one dimensional materials[1] in the early 1970's and the observation of non-ohmic electrical conductivity[2] in $NbSe_3$, the charge transport properties of the incommensurate charge-density-wave (CDW) systems in $NbSe_3$ and related materials have been studied extensively[3]. It is also well known that interesting field-dependent behavior is observed for the thermopower and related Peltier heat coefficient[4]. These two observations have prompted experimental efforts to measure all the Onsager transport coefficients related to CDW motion. However, for various reasons, the change in the thermal conductivity from a sliding CDW remains unknown. The following is a brief report on the thermal conductivity of the CDW systems $K_{0.3}MoO_3$, $(TaSe_4)_2I$ and TaS_3, measured by two different methods with and without an applied electric field.

The zero-field thermal conductivity of $K_{0.3}MoO_3$ and $(TaSe_4)_2I$ have been measured using a steady-state linear heat-flow method described in detail elsewhere[5], and the results are shown in Figs.1(a) and (b), respectively. The overall temperature dependence of κ_T is similar for both materials, with the slightly stronger temperature dependence above T_P for $(TaSe_4)_2I$ probably related to the stronger fluctuations in this material[6]. A sharp peak is observed at T_P in all of the

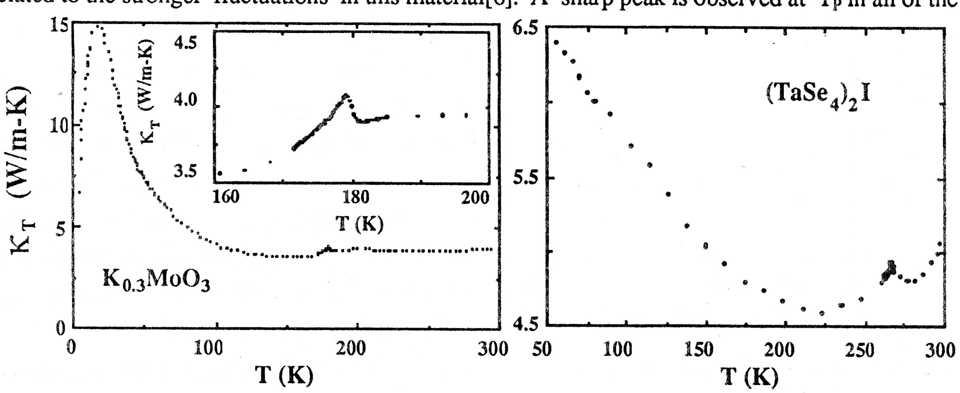

FIG.1 – Thermal conductivity of (a) $K_{0.3}MoO_3$ and (b) $(TaSe_4)_2I$ measured by a linear heat-flow method.

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blue bronze and $(TaSe_4)_2I$ samples that were measured, even though the details of the peak depend on the quality of the samples. We also observed that samples with a higher κ_T maximum at low temperature have a sharper and narrower anomaly at T_P. Since all samples measured have similar geometrical dimensions, it is likely that the correlation is from lattice defects.

In general, κ_T is modelled as a sum of the lattice κ_p and electronic κ_e contributions. For normal metals, κ_e can be estimated from the Wiedemann-Franz law. The situation here is complicated by the presence of a pseudo-gap and Peierls gap in the electronic spectrum above and below T_P, respectively. This will in general enhance the total thermal conductivity due to an extra contribution from recombination of electrons and holes[7]. As in the case of the new high-temperature superconductors[8], the opening of the energy gap at T_P decreases the phonon-electron scattering and therefore enhances the lattice contribution κ_p. On the other hand, enhanced fluctuations near the phase transition shorten the mean-free-path of the heat carrying phonons. As a consequence, a 'dip' is expected in the measured κ_T, as in the case of the antiferromagnets[9]. Such analyses have been carried out but could not explain the sharp peak observed in κ_T[10,11]. Recently, Deland et al.[12] also observed an extra 'bump' in the lattice thermal conductivity of blue bronze around T_P, although the anomaly is somewhat smeared out compare to what we report here. The authors suggested that the excess contribution was related to the fluctuation effects.

The thermal conductivity can also be measured by a steady-state self-heating technique[13] in which the specimen is heated directly by passage of an electric current. Such a method has been employed by Brill et al.[14] for measurement on $NbSe_3$. Using this technique, we have measured κ_T of orthorhombic TaS_3. Results for the zero-field limit are shown in Fig.2. The overall magnitude of κ_T is the same as previously reported[15]. However, the peak at T_P was not observed using the indirect method of Ref.[15]. The κ_T feature of TaS_3 around T_P is similar to, but significantly larger than that of blue bronze and $(TaSe_4)_2I$, indicating that the anomaly observed at T_P is an intrinsic property of CDW systems.

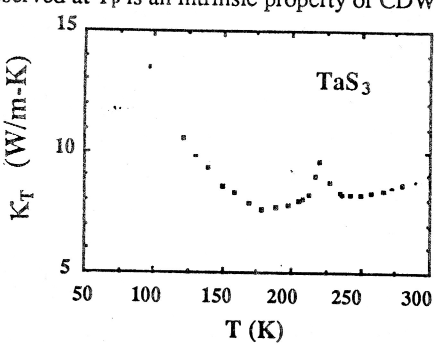

FIG.2 – Thermal conductivity of TaS_3 measured by a self-heating method.

The specific heat of blue bronze has been measured by a relaxation method[5]. A non-mean-field type of anomaly observed at T_P [10,16] indicated that the lattice contributed significantly to the specific heat anomaly[17]. Since the softening of phonons near $q=2k_F$ results in a non-zero group velocity $d\omega/dk$, it is possible that the increase in κ_T is a result of extra heat carried by the soft phonons.

To study the electric field dependence of κ_T of blue bronze, the linear heat-flow method was modified slightly[18]. The result is that, within the 2% uncertainty of our apparatus, κ_T of blue bronze is field independent up to 6 times the threshold field.

We were motivated to study the field dependence of the thermal conductivity of TaS_3 because the field dependence of the thermopower of that material is stronger than that of the blue bronze[4]. Due to the fragility of TaS_3, the linear heat-flow method is replaced by the self-heating technique described in Ref.[13]. By thermally anchoring the ends of the specimen to a base temperature T_o and measuring the mid-point temperature T_{max}, one can calculate κ_T (assuming κ_T and the electrical resistance R are weak functions of temperature) using :

$$\kappa_T = P\ell/8A\Delta T \tag{1}$$

where $\Delta T \equiv T_{max} - T_o$, $P=I^2R$ is the joule heat applied to the sample of cross-sectional area A and length ℓ. Also, the resistance of the sample is given by :

$$R = \frac{1}{A}\int_{-\ell/2}^{\ell/2} \rho(x)dx = R_o\left\{1 + \frac{I^2\ell}{12\kappa A}\frac{dR}{dT}\right\} \tag{2}$$

where ρ is the resistivity and R_o is the zero-field resistance of the sample.

Four representative curves of P vs. ΔT are shown in Fig.3 for four distinctive temperature ranges. For T>230K, κ_T and R are weak functions of temperature and Eq.(5) predicts ΔT to be linear with P. Around the Peierls transition at about 220K, the curvature of $\Delta T(P)$ is dominated by the temperature dependence of κ_T. The measurement at T_o=205K shown in Fig.3 follows the prediction of $\kappa_T(T)$ from Fig.2 as ΔT rises. For 150K<T<210K, κ_T is weakly temperature dependent as is the case for T > 230K. However, R is a stronger function of temperature and consequently, T_{max} is much smaller than that predicted by Eq.(1) because the mid-point resistivity is much lower than the average ρ. This is revealed in the slightly concave down $\Delta T(P)$ of the T_o=160K curve in Fig.3. A calculation of $\kappa(I_{max})$ at T_o=160K from Eq.(2), using the measured I-V curve and the appropriate dR/dT, gives the result that $\kappa(I_{max}) \approx \kappa(0)$ to within 5%. Finally, for T<150K, the upturn of $\Delta T(P)$, due to the rapid decrease of κ_T as T_{max} increases, is partly compensated by the concave down effect due to the fast dropping mid-point

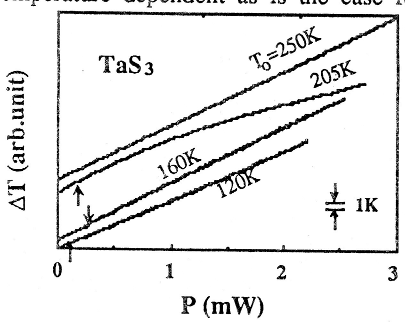

FIG.3 – Four typical ΔT vs. P curves for TaS_3 as discussed in text. Arrows show where the threshold field ($i_T V_T$) is from resistivity measurements.

resistivity. The net effect is a slight upward concavity of $\Delta T(P)$ as shown in the T_0=120K curve in Fig.3.

We have checked the previous result for $K_{0.3}MoO_3$ using the same method over the temperature range of 100-160K. ΔT is found to be linear with P, to within 1%, up to about 25mW or equivalently $\Delta T \approx 6K$. This corresponds to applied fields of about 8 times the threshold field. Combined with the earlier experiments, we conclude that κ_T is field independent for TaS_3 as well as for blue bronze to within 5 and 1%, respectively.

In summary, we observed a peak in κ_T of the CDW systems $K_{0.3}MoO_3$, $(TaSe_4)_2I$ and TaS_3. Together with the specific heat measurement on blue bronze, we suggested that the anomaly is related to the heat carried by phonons with q near $2k_F$. Furthermore, κ_T of blue bronze and TaS_3 are found to be field independent within the uncertainty of our apparatus, which is about 1 and 5%, respectively.

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