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Field dependence of the non-linear conduction of charge-density waves in thallium blue bronze $Tl_{0.3}MoO_3$

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Abstract. The non-linear transport properties of sliding charge-density waves (CDWs) in pure $Tl_{0.3}MoO_3$ and impurity-doped $(Tl_{0.2}K_{0.8})_{0.3}MoO_3$ blue bronzes are measured in the temperature range 80–150 K. It is found that the analytical form of non-linear current in a moderate field range fits the power-law relation, i.e. $I_{CDW} \propto (V/V_t - 1)^{\alpha}$, where the exponent α is temperature depdendent and not a constant 1.5. Such a relation seems to be universal for blue bronzes over the temperature range below the Peierls transition temperature and does not depend on the quality of samples. The results are in agreement with those observed by Mihaly *et al* for pure $K_{0.3}MoO_3$ blue bronze in the low-temperature range and suggest that the dominant damping of the collective mode arises from the dissipative normal current induced by dynamical deformation of the CDWs, not only in the low-temperature range, but also in the high-temperature range.

1. Introduction

Since the mid-1970s a great deal of interest has been demonstrated in charge-density-wave (CDW) materials, where collective charge transport of the CDWs results in non-linear conduction below the Peierls transition temperature (see [1–5] for a review of CDW phenomena). A variety of theoretical models [6–10] have been proposed to account for the main characteristics of the non-linear transport in quasi-one-dimensional metals due to the depinning of the CDWs in the presence of the electric field. Up to now, there has still been no consensus as to the correct approach for describing these systems [11]. The experimentalists found that none of these predictions is universal over the wide non-linear range nor fitted the data well for different CDW materials. It seems that the tests of most models resulted from the data on $NbSe_3$ and TaS_3 [12–15]. The experiments aimed to determine the field dependence of CDW transport were fitted by different analytical forms with different physical meanings. However, for blue bronzes, relatively little work was available such that these theoretical models might fit the experimental data well over the wide temperature or field range.

Mihaly *et al* [16] have however, investigated the non-linear characteristics of high-quality $K_{0.3}MoO_3$ single crystals displaying a sharp threshold in the temperature range 30–100 K and found that the CDW conductivity follows an empirical form $\sigma_{CDW} = k\sigma_n(E_T/E)(E/E_T - 1)^{\alpha}$, where α is the temperature-dependent exponent and σ_n the conductivity of normal carriers. However, the physical meaning of such an empirical

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form seen experimentally in the K bronze is not clear, as we know none of the present microscopic theories of CDW damping suggests such a power-law relation.

Thallium blue bronze $Tl_{0.3}MoO_3$ was considered to be an isostructural compound [17, 18] with potassium blue bronze $K_{0.3}MoO_3$ and reveals the existence of CDW [19]. However, the investigations of low-frequency conductivity [20] have shown that both the mean relaxation time and the static dielectric constant are much smaller in $Tl_{0.3}MoO_3$ than they are in the corresponding isostructural alkali bronzes. Recently, our transport measurements [21] found that an increase in substitution of the alkali metal K by Tl in $K_{0.3}MoO_3$, the voltage–current (V–I) characteristics exhibit a large electrical hysteresis, and negative or near-zero-differential resistance phenomena (i.e. second threshold behaviour) in the high-field region above 77 K. Clearly, all these physical properties suggested quantitative differences of CDW dynamics compared with the alkali-metal blue bronzes. Therefore, in order to examine whether the present theories can be used to describe accurately the non-linear transport properties of these materials, we present in this paper a detailed measurement of the field dependence of sliding CDW conduction for pure Tl bronze $Tl_{0.3}MoO_3$ in the high-temperature range 77–150 K, and for impurity-doped blue bronze $(Tl_{0.2}K_{0.8})_{0.3}MoO_3$. The latter introduces a weak disorder in the CDW state, leading to a small shift in the Peierls transition temperature towards low temperatures.

2. Experimental method

The single-crystal $Ti_{0.3}MoO_3$ and $(Tl_{0.2}K_{0.8})_{0.3}MoO_3$ used in this work were grown by the standard electrolytic reduction method from melts of Tl_2CO_3–MoO_3 and K_2CO_3–Tl_2CO_3–MoO_3, respectively, with an appropriate mole ratio [22]. The structures of the crystals were verified by four-circle x-ray diffraction and are in agreement with that reported by Ganne *et al* [17]. The electrical contacts were made by evaporating an Au film onto a freshly cleaved surface. Gold wires were attached to the Au-film areas by silver paint. In these cases the contact resistance was negligible in the temperature range measured.

All measurements reported here were performed on crystal A (pure) and B (impurity doped) with dimensions of about 1.0 mm×1.0 mm×0.1 mm and 1.2 mm×1.0 mm×0.5 mm, respectively. The voltage-current (V–I) curves at different temperatures were recorded by a continuous method with a quick sweeping constant current in the low-field range and pulsed experiments in the high-field range in order to avoid the Joule-heating effect. The recorded value of voltage for each rectangular current pulse is determined from the leading edge of the response curve.

3. Experimental results

Figures 1, curves a and b, show the V–I characteristics of pure and impurity-doped blue bronzes respectively, at 90 K. It is seen that for both samples the first threshold region of the V–I characteristics is smooth, and at higher field strengths both V–I characteristics present a second threshold V_{T2}, when the current increases greatly, the voltage across the sample does not increase accordingly but rather decreases. This novel phenomenon seen in Tl- or impurity-doped blue bronzes will be discussed elsewhere. In this paper, the interesting point for these two samples with an unusual second threshold is whether the non-linear transport behaviour of CDWs below V_{T2} can be qualitatively described by the present theory or others. Such a non-linear V–I curve with a smooth first threshold, to a great extent,

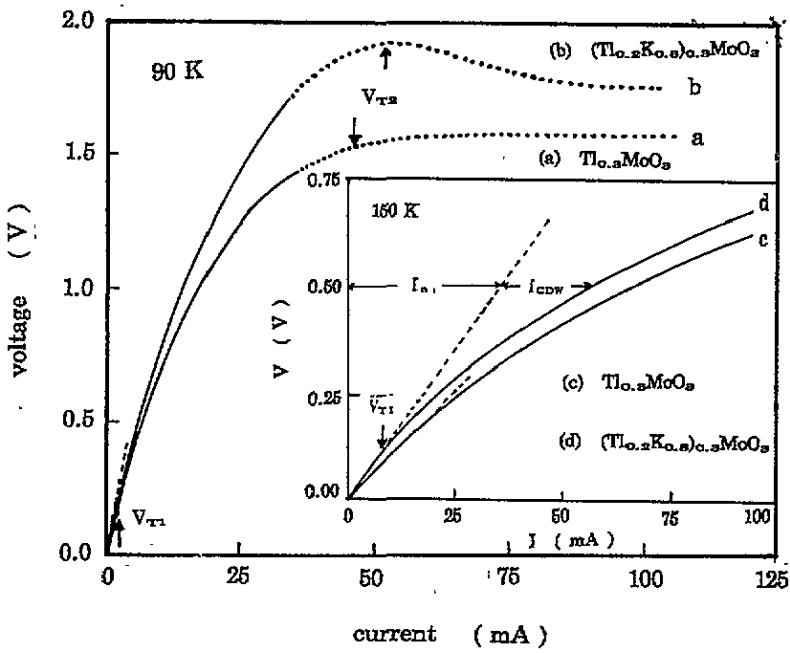

Figure 1. Voltage–current (V–I) characteristics of pure blue bronze $Tl_{0.3}MoO_3$ at 90 K (curve a) and for impurity-doped blue bronze $(Tl_{0.2}K_{0.8})_{0.3}MoO_3$ (curve b); the arrow denotes the threshold point; – – –, portion of the curve measured by the current pulse method. The inset shows the V–I curves at 150 K for $Tl_{0.3}MoO_3$ (curve c) and $(Tl_{0.2}K_{0.8})_{0.3}MoO_3$ (curve d).

reflects the sliding properties of CDWs pinned by randomly distributed weak pinning centres as suggested by Lee and Rice [23]. This case is different from that reported by Mihaly *et al* [16] at low temperatures where the threshold is very sharp. The two inset curves c and d show the transport feature for the CDWs at 150 K, for pure Tl bronze or impurity-doped bronze, respectively. It is seen that at 150 K over the measured field range no second threshold was observed (the second threshold disappears at 100 K and 110 K for $Tl_{0.3}MoO_3$ and $(Tl_{0.2}K_{0.8})_{0.3}MoO_3$ blue bronzes, respectively).

Figures 2(a) and 2(b) show the CDW current as a function of $V/V_T - 1$ at various temperatures for pure $Tl_{0.3}MoO_3$ and impurity-doped $(Tl_{0.2}K_{0.8})_{0.3}MoO_3$ blue bronzes, respectively, on double-logarithmic scales. The CDW current I_{CDW} at different fixed fields is determined from the V–I characteristics, where the normal-current component $I_n = V/R_n$ is subtracted using the relation $I_{CDW} = I_{total} - I_n$, shown in the inset of figure 1. It is seen that the curves of I_{CDW} versus $V/V_T - 1$ on double-logarithmic scales approximately give straight lines in a moderate field range, i.e. they can be described by the following power-law relation:

$$I_{CDW} \propto (E/E_T - 1)^{\alpha} \tag{1}$$

where the exponent α can be determined from the slope of the straight line. We found that at 78 K for the pure $Tl_{0.3}MoO_3$ sample the curve gives a satisfactory description of the field dependence from about $2V_{T1}$ up to $13V_{T1}$; above $13V_{T1}$ the curve shows a clear deviation from the straight line. When the temperature increases from below, the same analytical form occurs although the non-linear V–I characteristics are different for the various temperatures.

At $T = 150$ K, for the pure blue bronze, the range of validity of the power-law relation is from about 20% above threshold voltage V_{T1} up to $5V_{T1}$. The temperature dependence of the exponent $\alpha(T)$ for both kinds of blue bronze are shown in figure 3, and it is seen that for both samples the exponent α increases with increase in temperature; for pure Tl bronze the values of the exponent α in the temperature range 78–150 K increases from 1.9 ± 0.1 to 2.3 ± 0.1, but for impurity-doped bronze $\alpha = (1.45 \sim 2.1) \pm 0.1$. This behaviour does not depend on the crystal quality. We have found a similar temperature dependence of α for all the samples investigated, including the samples without the second threshold field in the temperature range measured. Hence, on the basis of the above analysis, the CDW conductivity at different temperatures can be written

$$\sigma_{CDW} = k\sigma_n(E_T/E)(E/E_T - 1)^{\alpha} \tag{2}$$

where σ_n is the conductivity of normal carriers and k is a temperature-independent constant. This form is in agreement with that found by Mihaly *et al* [16] in the K bronze $K_{0.3}MoO_3$ although our studied samples are quite different from those used previously.

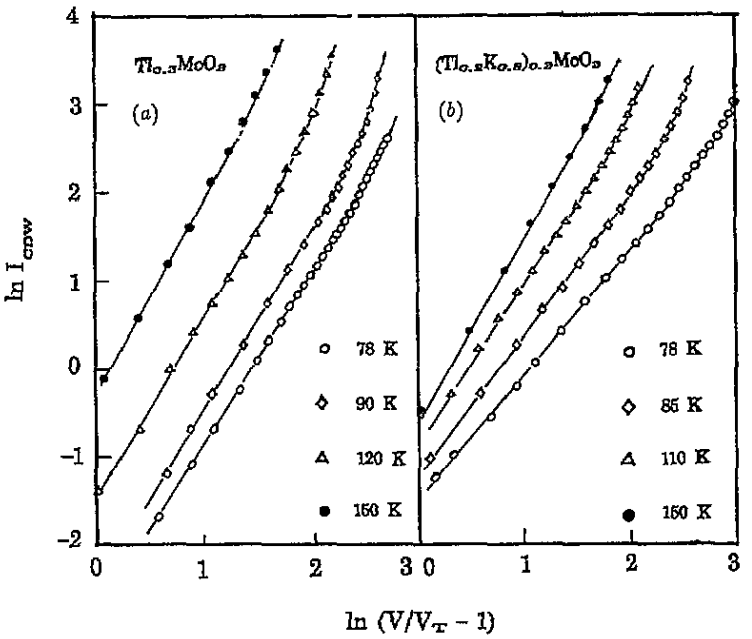

Figure 2. The non-linear CDW current I_{CDW} versus $V/V_T - 1$ on a double-logarithmic scale at *several temperatures for (a) pure blue bronze* $Tl_{0.3}MoO_3$, *and (b) impurity-doped blue bronze* $(Tl_{0.2}K_{0.8})_{0.3}MoO_3$.

4. Discussion

As seen above, for pure $Tl_{0.3}MoO_3$ or impurity-doped blue bronzes, the field dependence of the CDW collective mode transport over the measured temperature range can be described by a power-law relation at a moderate-field range, similar to that found by Mihaly *et al* for pure

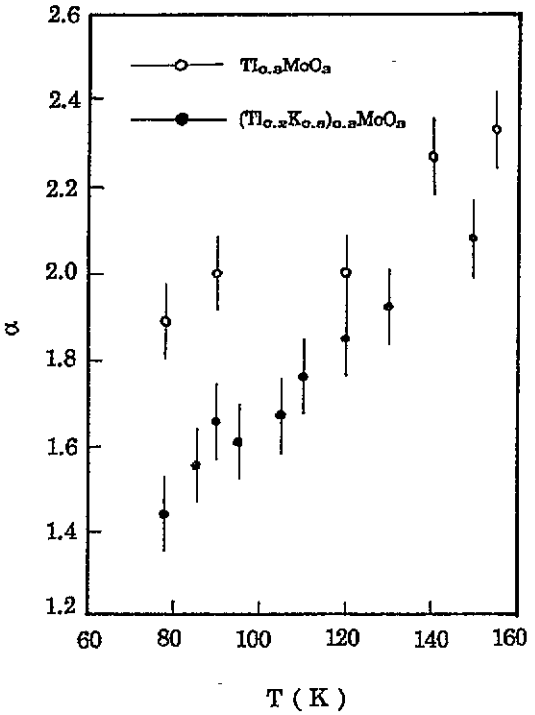

Figure 3. The curves of exponent α versus temperature for pure blue bronze $Tl_{0.3}MoO_3$ and impurity-doped $(Tl_{0.2}K_{0.8})_{0.3}MoO_3$.

$K_{0.3}MoO_3$, although the samples used here exhibit a dramatic second threshold at a higher electric field. We believe that equations (1) and (2) are the general expression for j_{CDW} and σ_{CDW} in blue bronze compounds in the moderate-field range. Therefore, although we do not exclude the possible validity of the quantum tunnelling theoretical models [8, 11–14] used for the $NbSe_3$ and TaS_3 system, at least for blue bronzes this model is not appropriate for treating the non-linear behaviour of CDW transport under low- and high-field conditions within the measured temperature range for all the samples investigated.

Mihaly *et al* [16] thought that one of the most important conclusions of equation (2) is that in wide field and temperature ranges the collective response of CDWs is determined by the single-particle conductivity, and therefore the dissipative normal currents induced by dynamic deformations cause the damped nature of the collective mode. This statement favours the theoretical predictions of Sneddon *et al* [9] but, owing to the clear deviation between the predicted and experimentally observed field-dependence forms, they speculated that it might arise from corrections due to high-frequency terms in σ_n in equation (2). If this speculation is correct, by comparing with our measurements in the high-temperature range (78–150 K), a similar power-law analytical form of CDW collective mode transport might imply that, at least for Tl bronze, the dominant dissipation involved in CDW motion over the measured temperature range below T_P also originates from the screening of the fluctuation produced by local CDW deformations by back-flow currents of normal carriers [9]. The viscous damping arising from the phason–phason or phason–phonon scattering may be very small, even at higher temperatures. In other words, we would expect the polarization and distortion of the CDW condensate in blue bronzes to be appreciably larger in the fields over the temperature range below T_P than in other CDW materials. Such a high polarizability

might be related to its crystal structure and probably results from a weak interlayer coupling due to additional screening of the direct interlayer Coulomb coupling. Another possibility could be that there are more effective impurities in blue bronzes owing to a weak coupling by alkali or thallium atoms between the interlayers. The size effect of Tl atoms might be able to cause a more intensive deformation and stress for $Tl_{0.3}MoO_3$ and $(Tl_{0.2}K_{0.8})_{0.3}MoO_3$ than for $K_{0.3}MoO_3$ and might be associated with the large hysteresis and second threshold at high temperatures, but they do not change the damping nature in the low-field range below V_{T2}. Tucker and Lyons [24] thought that such a slightly superlinear form ($\alpha = 1.2$–1.4 for $K_{0.3}MoO_3$) at low temperatures may represent a smeared-out version of the predicted non-linearity, $\exp(-E_0/E)$, because of inhomogeneous electric field distributions at low temperatures. Therefore, at high temperatures the inhomogeneity of the electric field will be expected to be greatly decreased on increase in the conductivity of the samples. In fact, from our measurements such a power-law relation does not seem to be strongly affected by the inhomogeneity of the electric field. As far as we know, none of the present microscopic theories of CDW damping suggests such a power-law relation. A further theoretical study is needed to clarify the clear physical meaning of such a power-law relation.

From figure 2, the power-law relation for CDW transport fits only the moderate-field range below V_{T2}; at high fields a deviation can be clearly observed. These deviations at high fields for Tl bronze samples may imply a different damping mechanism of CDW motion, which might be associated with the second-threshold phenomenon. Although several theoretical models [24–26] have been proposed to explain the second-threshold phenomenon, up to now the real origin is still unclear and remains to be further clarified.

In addition, the power-law form is quite similar to that suggested by Fisher [10], where the exponent α is a constant 1.5. However, Fisher's theoretical model should be limited only to quite near the critical threshold region, where the phase–phase correlation length is smaller than the dimensions of the specimens. The development of the current-carrying state is treated as a dynamical critical phenomenon, with several critical exponents that characterize the dynamical system. Hence, in principle it does not characterize the non-linear behaviour in the moderate-field range. Whether such a surprising similarity has some relation to the theoretical prediction form near E_T and the empirical form at moderate fields is still unclear.

In summary, we have investigated the field-dependent CDW non-linear transport of thallium bronze and impurity-doped bronze. The results showed that the analytical forms of the CDW non-linear transport in the moderate-field range above V_{T1} ($2V_{T1} < V < V_{T2}$ for the samples having a dramatic second threshold V_{T2}) also fit the empirical form of the power-law relation in the temperature range measured; the exponent α of the empirical form is not a constant value 1.5 but depends upon temperatures. It seems that the power-law form of the CDW non-linear transport is universal for blue bronzes in the moderate-field condition and almost independent of the quality of the samples. Such a similarity suggests that for Tl bronze the dominant damping nature of CDW motion in the moderate-field range arises from the screening of the fluctuation charges produced by local CDW deformations by back-flow currents of normal carriers over the measured temperature below T_P. At higher fields the non-linear transport deviates from the power-law form, which might mean that a different damping mechanism is induced in higher electric fields.

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