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Non-linear conductivity of quasi-one-dimensional TaS₃ at low temperatures

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Abstract. The non-linear conductivity in orthorhombic and monoclinic TaS₃ has been measured as a function of temperature from the Peierls transition temperature down to 4.2 K in the electric field range 10^{-2} -500 V cm⁻¹. At liquid-helium temperature a sharp increase in conductivity is measured in a very limited variation range of voltages. A general description based on phase-slip processes is given to explain these non-linear properties in the whole temperature range.

1. Introduction

In the last few years there has been considerable interest in the study of non-linear conductivity of quasi-one-dimensional conductors at liquid-helium temperature. One of the reasons for this interest was the observation, below 20 K for blue bronze $K_{0.3}MoO_3$, of an abrupt increase in the current by several orders of magnitude at almost constant voltage. At 4.2 K above a threshold field a switching appears between the insulating state at low voltages to a highly conducting state in which the current apparently increases with a zero differential resistivity [1–5]. The low damping of the charge-density-wave (CDW) motion and the fast current response (about 10⁻⁸ s) exhibited in this CDW compound yield the possibility that the physical mechanism of CDW motion at low temperatures is radically different from that at higher temperatures and may be associated with the so-called Fröhlich superconductivity [3, 4]. Therefore, such properties should be generic in other CDW systems which are insulating at liquid-helium temperature.

In this paper we report the results of a systematic study of non-linear conductivity in orthorhombic TaS₃ (o-TaS₃) and monoclinic TaS₃ (m-TaS₃) in the wide temperature T range 300–4.2 K and the wide electric field E range 10^{-2} –500 V cm⁻¹. On the basis of these results, previous results and available theories, we attempted to explain the experimental dependences of the electrical conductivity as a function of T and E from a general point of view based on phase-slip processes.

2. Experiment

We investigated ten samples of $o-TaS_3$ and $m-TaS_3$ with different impurity contents and degrees of structural perfection. Some of the results, essentially for samples of $o-TaS_3$



Figure 1. Temperature dependence of the conductivity of TaS₃ samples: \bigcirc , orthorhombic sample O-2; \triangle , orthorhombic sample O-3; \bigcirc , monoclinic sample. The numbers indicate the activation energy on the appropriate regions of the curve. The inset shows the dependence of σ_1 versus $(10^3/T)^{1/2}$ at $T \le 20$ K: _____, $\sigma_1 \sim \exp[-(T_1/T)^{1/2}]$.



Figure 2. Dependence of conductivity normalized to its room-temperature value as a function of the electric field for $o-TaS_3$ sample O-2. The temperatures are indicated on the curves.

with a large impurity concentration, were published earlier [6, 7]. The conductivity measurements were usually carried out in a two-terminal configuration, but in several cases we used four terminals. The present results were obtained on thin samples which were placed on insulating substrates. Electrical contacts were made by gold paint or by vacuum deposition of In strips after appropriate thermal treatment of samples in vacuum [8].

For conductivity and I-V curve measurements the whole range of electric fields was divided into several slightly overlapping regions. For low electric fields at low temperatures we used electrometers with a high input impedance (about $10^{16} \Omega$). For high electric fields at low temperatures we used a pulse method in the regime of controlled voltage with pulse duration down to about $0.1 \,\mu$ s. The I-V curves were recorded using a dual-channel boxcar integrator and slowly sweeping the pulse amplitude. The highest electric field available was governed by the beginning of sample heating which was regularly controlled by observing the pulse form on an oscilloscope.

Figure 1 shows the temperature dependence of the conductivity σ_1 of o-TaS₃ and m-TaS₃ samples in the linear region of the *I*-V curves. For o-TaS₃ samples at temperatures below $T_P = 220$ K (T_P is the Peierls transition temperature where threedimensional ordering occurs) down to $T \approx 100$ K the dependence of log σ_1 is close to linear as a function of 1/T with an activation energy Δ of about 800 K. Below 100 K in some samples a smooth transition to another regime was observed. This region is also close to linear with an activation energy Δ_5 of about 220–240 K. Then below 20 K the log $\sigma_1(1/T)$ dependence becomes flatter and the σ -values become strongly dependent on the sample quality and vary within several orders of magnitude. In this temperature



Figure 3. Dependence of conductivity normalized to its room-temperature value as a function of the electric field for $m-TaS_3$ sample M-1. The temperatures are indicated on the curves.



Figure 4. Dependence of the threshold field E_T as a function of temperature for various TaS₃ samples: \triangle , sample O-2; \bigcirc , sample O-3; +, sample O-4; \blacksquare , sample M-1. The inset shows the dependence of E'_T on temperature for sample M-1 (\Box).

range the experimental dependence of $\log \sigma_1 (1/T)$ for sample O-2 can be approximated by $\log \sigma_1 (1/T) = \log \sigma_0 - (T_1/T)^{1/2}$ with $T \approx 400$ K (see inset in figure 1). This type of approximation was also correct for a number of other o-TaS₃ samples [6, 7]. It is necessary to note that below 100 K the conductivity perpendicular to the chain c axis continues to decrease with an activation energy close to the gap value $\Delta \approx 800$ K [6] and, consequently, the anisotropy of conductivity when T is reduced increases sharply [9]. On the other hand, for m-TaS₃, $\log \sigma_1$ is almost a linear function of 1/T with an activation energy Δ of 950 K down to 25 K, the lowest temperature at which a linear resistivity (ten orders of magnitude larger than at room temperature) can be measured.

Figure 2 shows the dependence of conductivity on electric field E at different temperatures for o-TaS₃ and figure 3 shows the same for m-TaS₃. As can be seen from figures 2 and 3 these dependences have a qualitatively similar form. Dependences of $\sigma(E, T)$ for K_{0.3}MoO₃ samples obtained in [3] and in some of our experiments are also similar. For T > 100 K the form of I-V curves and their variation with temperature have been well investigated previously [10, 11]. After the electric field exceeds the threshold value $E_{\rm T}$, the non-linear conductivity $\sigma_{\rm n} = \sigma - \sigma_{\rm 1}$ appears, which can be approximated by a $\sigma_{\rm n} \sim \exp[-E_0/(E-E_{\rm T})]$ dependence [12], where E_0 is some characteristic field.

When T is reduced further, the variation in $\sigma(E)$ begins to change. Near $T \simeq 70-80$ K

the $\sigma(E)$ curves with upward convexity at $E > E_{\rm T}$ become more linear on a logarithmic scale and approximately follow a $\sigma_n \sim E^{\alpha}$ dependence, with $\alpha \simeq \frac{3}{2}$. The transition from ohmic to non-ohmic conductivity broadens and we can see two threshold fields $E'_{\rm T}$ and $E_{\rm T}$ in the $\sigma(E)$ dependence (figures 2 and 3). At the field $E \ge E'_{\rm T}$ we observe a slight deviation from a linear *I–V* curve and a gradual increase in σ_n between E'_T and E_T . With further temperature reduction (T < 40-50 K) this increase in σ_n becomes significant and the region in which it exists is broadened; the $E'_{\rm T}$ -value decreases (inset in figure 4) but $E_{\rm T}$ continues to increase. For electric fields in the range $0.1E_{\rm T} < E < E_{\rm T}$ the $\sigma(E)$ dependence follows a low $\sigma \sim E^{\nu}$ where, for o-TaS₃ (figure 2), $\nu = 0.7$ at T = 20 K and increases up to about 1.5 at 4.2 K. Below $0.1E_{\rm T}$ the $\sigma(E)$ variation shows some sample dependence. Thus in figure 2 at T = 4.2 K it seems that there is another characteristic field between $E_{\rm T}$ where the conductivity increases sharply and $E'_{\rm T}$ below which the conductivity is linear. Such a bend in the curve does not occur for every sample and disappears at a slightly higher temperature. Although this behaviour is not totally understood, it may reflect peculiarities of the hopping conductivity with electric field in this sample. It should also be noted that, if at 4.2 K a linear state can be measured in o- TaS_3 (figure 2), the conductivity is non-linear at the smallest voltage applied in m-TaS₃ (figure 3, sample M-1, T < 25 K).

At $E \ge E_T$ we observe a sharp increase in conductivity, which is enhanced with decreasing temperature (figures 2 and 3). For T < 70 K many $\sigma_n(E)$ dependences are close to a power-law-type dependence: $\sigma_n \sim (E - E_T)^{\alpha}$, where the value of α increases significantly from $\alpha \simeq \frac{3}{2}$ ($T \simeq 70$ K) up to $\alpha \simeq 15$ ($T \simeq 4.2$ K) for sample O-2. Preliminary measurements with a four-terminal configuration show a dependence of $\sigma(E)$ for $E > E_T$ which is even more vertical (i.e. in this case the same values of the conductivity correspond to electric field values smaller than in two-terminal configuration).

Figure 4 shows the temperature dependence of E_T obtained from several samples of o-TaS₃ with various purities and also for m-TaS₃. In the case of o-TaS₃ for T > 100 K, E_T depends slightly on T with some peculiarities observed near T_P and $T \le 150$ K [6, 13]. E_T starts to increase below $T \approx 100$ K and continues to grow when T is reduced to the liquid-helium temperature. In the case of m-TaS₃ the E_T increase below $T_{min} \approx 145$ K, which corresponds to the minimal value of E_T and continues also down to 4.2 K. For all samples, in this temperature range, the log E_T dependence on T turns out to be very close to linear, which agrees with the exponential growth of E_T with decreasing temperature:

$$E_{\rm T}(T) = E_{\rm T}(0) \exp(-T/T_0). \tag{1}$$

A similar $E_{\rm T}(T)$ dependence for o-TaS₃ has been measured earlier but only in the temperature range above 40 K [6, 13]. As can be seen from figure 4 the temperature dependence of $E_{\rm T}$ and the value of T_0 are almost independent of the quality of the o-TaS₃ samples. Equation (1) is characteristic of various o-TaS₃ samples with $E_{\rm T}$ which varies by two orders of magnitude from 0.3 to 30 V cm⁻¹ (T = 100 K). For all o-TaS₃ samples the characteristic temperature T_0 is approximately equal to 20 K and $T_0 \approx 25$ K for m-TaS₃.

3. Discussion

To explain these results, one needs to take into account the opening of the gap below the Peierls transition temperature and the formation of the CDW superlattice which is close to fourfold commensurability in both TaS_3 with respect to the initial (pristine) lattice. However, local deformations and defects can develop very easily in the soft CDW superlattice; dislocations, jumps in the CDW phase and the CDW amplitude near strong pinning centres, contacts or surfaces of the sample and phase shifts needed for compensation of the near commensurability with the initial lattice can be such types of excitation. In general these defects of the CDW condensate can be developed not only under the influence of the above-mentioned factors but also under the action of thermal fluctuations and of the applied electric field [6, 9, 10, 14–16]. Following this general framework, the discussion hereafter will be divided into several parts related to the behaviour of the conductivity

- (i) in the linear regime for the full temperature range and at low temperatures,
- (ii) for applied fields approximately equal to $E'_{\rm T}$ and
- (iii) approximately equal to $E_{\rm T}$.

3.1. Conductivity in the linear state

Below T_p the linear log $\sigma(1/T)$ dependence in o-TaS₃ and m-TaS₃ is commonly attributed to the thermal activation of free carriers—electrons and holes—across the Peierls gap $\Delta_P(\Delta_P = 800 \text{ K} \text{ for o-TaS}_3; \Delta_P = 950 \text{ K} \text{ for m-TaS}_3)$. In o-TaS₃ the behaviour is slightly more complicated owing to hysteresis effects in temperature cycling between T_P and 100 K [14] which is very probably consecutive to the temperature dependence of the longitudinal component of the *q*-vector. However, good agreement between the values of the thermal and optical gaps has been found for o-TaS₃ [17].

The linear region with a log $\sigma(1/T)$ dependence in the range 100 K < T < 30 Kfor o-TaS₃ with an activation energy $\Delta_s < \Delta_P (\Delta_s \simeq T_P \simeq 200 \text{ K}; \text{ see figure 1})$ has been attributed to the thermal excitation of soliton-like excitations [6]. Theories [18-21] and experimental data [6, 7, 9, 14-16] show that in CDW quasi-one-dimensional conductors, close to commensurability with $n \ge 3$, defects in the CDW superstructure can be defined in terms of static and dynamic jumps of 2π in the CDW phase. At the places where these phase jumps occur, local variations in the CDW amplitude and in the energy gap also develop. The nucleation of such excitations destroys the order in the CDW superlattice only within small regions of phase and amplitude variations. In the other regions of the crystal the change in CDW phase of 2π between one part and another part does not modify the total energy of the system. This means that in the case of a phase jump of 2π the energy growth is minimal and it only occurs at the small local region of the jump itself. The minimum energy needed for developing phase shift between two parts in the CDW lattice is determined by the interchain interaction and is of the magnitude of the interchain energy, approximately $kT_{\rm P}$ [6, 9, 18]. Being related to the quasi-onedimensional nature of the investigated materials, these excitations make a contribution mainly to the conductivity σ_{\parallel} parallel to the chain axis while the transverse conductivity σ_{\perp} decreases with the same activation energy of about $\Delta_{\rm P}$ as determined for free electrons as before.

When the temperature T is much less than Δ_s (beginning from $T \approx 20-30$ K for o-TaS₃), the thermal activation of solitons becomes very weak. However, deformations in the CDW superstructure near defects and impurities, which are considered in terms of soliton-like excitations, remain at these low temperatures. Naturally they are localized in the CDW superlattice in a random way. In these conditions the jumps of solitons between these localized states become the main mechanism of the current transport in quasi-one-dimensional conductors [6, 7, 19]. The large difference between the activation energies of σ_{\parallel} and σ_{\perp} and also the difference between values of the dielectric constants ε_{\parallel} ($\approx 10^6$) and ε_{\perp} ($< 10^2$) [7] provide strong evidence that the longitudinal conductivity and the polarization arise because of the motion of CDW deformation solitons than because of the free-electron motion [6, 7, 9, 18]. It seems that these processes are similar to hopping conductivity in disordered systems [22–24]. It was shown earlier [6] that in this temperature range the temperature dependence of the ohmic conductivity of o-TaS₃ samples is well approximated by the equation $\sigma_1 = \sigma_0 \exp[-(T_1/T)^{1/2}]$ characteristic for variable-range hopping in the one-dimensional case [22]. As can be seen from figure 1 the same dependence was observed in sample O-2. For similar o-TaS₃ samples it was also shown that the frequency-dependent conductivity $\sigma(\omega)$ was proportional to ω^{ν} , where $\nu \simeq 0.8[6, 7]$, and $\sigma(\omega)$ increases in direct proportion to T with saturation at $T \simeq$ 20 K [7] and that the ratio of the imaginary to the real part of the conductivity $\sigma(\omega)$ is in quantitative agreement with the value corresponding to the case of variable-range hopping [7, 22]. Many other pieces of evidence for the existence of disordered states in CDW at low temperatures were also obtained in studies of the frequency-dependent conductivity over a wide frequency range [25], in investigations of the temperature dependence of the specific heat of o-TaS₃ [26], from electron paramagnetic resonance spectra in blue bronzes [27] etc.

3.2. Electric-field-dependent conductivity: $E \simeq E'_T$, T < 100 K

The variation in the conductivity with the electric field for T < 100 K can also be explained on the basis of the mechanism of hopping motion of solitons. The experimental dependence shows good agreement with the theoretical predictions obtained in electricfield-dependent hopping conductivity of disordered systems [24]. In particular, it has been shown that the $\sigma(E, T)$ dependence is governed by the parameter $\beta = eEl/2kT$ where l is some length which in our case seems to be equal to the average distance between impurities. When $E \rightarrow 0$, the conductivity remains finite. When E is increased, a transition between the linear and the non-linear regime occurs at $\beta = 0.1$ [24], i.e. at $E \simeq 0.1(2kT/el)$ which can be considered as the first threshold $E'_{\rm T}$. Following this interpretation, E'_{T} decreases linearly with T. Experimentally as shown in the inset of figure 4, E'_{T} determined from the curves in figure 2 varies linearly with T. Substituting the experimental data in the equation for β we obtain $l \simeq 1 \,\mu m$ which is the order of the value for the average distance between impurities in our samples. For $E > E_T^{\prime}$, σ increases; this is associated with the increase in hopping mobility of solitons without motion of the CDW as a whole. The fact that the dielectric constant does not change for $E < E'_{\rm T}$ and for $E'_{\rm T} < E < E_{\rm T}$ provides evidence for this [7]. The conductivity is determined by the joint action of temperature and electric field and in principle, for $\beta \ge 10$, σ should not depend on temperature and be weakly dependent on the electric field [24]. However, in these quasi-one-dimensional conductors, we observe a sharp increase in σ for $E \ge E_T$ (figures 2 and 3).

3.3. Electric-field-dependent conductivity: $E \ge E_T$

When E is increased, the CDW pinned at the contacts and at the strong pinning centres sustains stronger deformations. For $E > E_T$ the CDW deformation in some local region exceeds a critical value with local suppression of the order parameter (energy gap), destruction of the CDW phase coherence and removal of the accumulated phase difference by 2π [14, 15, 20, 21, 27–34]. As a result, 2π solitons nucleate in this local region [15, 20, 21, 34]. Further the process is periodically repeated. Such a phase-slip process seems to be the unique mechanism capable of providing the pinning of the CDW from contacts and strong pinning centres, to allow conversion of the electron current to CDW

current [15, 20, 21, 34]. The sharp increase in σ at $E > E_T$ seems to be associated with phase-slip processes and nucleation of 2π solitons under the influence of E. The phase slips in actual crystals do not occur on a single chain but in some minimal volume which seems to correspond to a unit cell of the CDW superstructure. After a phase slip the number of CDW periods and the value of the CDW wavevector in a domain change, which is equivalent to the development of a dislocation in the superstructure [19, 21]. These dislocations move and grow in the longitudinal and transverse directions, providing a decrease in the phase difference through the whole cross section. The threshold field E_T can be associated with the energy F_0 which is needed for the nucleation of a dislocation in the CDW condensate. Various versions of the estimation of this energy exist, but the most realistic estimation associates F_0 with the condensation energy of about kT_P [14, 16]; for one electron the condensation energy is $f_0 \simeq (kT_P)^2/E_F \simeq \Delta_P^2/E_F$. In o-TaS₃ E_F is about 2400 K [15] and then f_0 is about 20 K which corresponds to a voltage V_0 of about 2 mV. In other words it is necessary to apply such a voltage over some length in order that the CDW deformation approaches the critical value.

This physical picture of CDW depinning has been previously proposed in the hightemperature range (T > 100 K) and for NbSe₃ in the cases where free electrons are able to screen the CDW deformations and to provide local electroneutrality in the sample. However, the application of the same picture at low temperatures is problematic [5, 35]. It was pointed out that, at low temperatures, the screening length and the CDW time response should be greatly increased. Indeed, in the absence of free electrons the propagation of long-wave and small-energy CDW excitations becomes impossible [3, 4]. However, it is possible that short waves and high-energy CDW excitations develop, which correspond to a fast response time at $E > E_{T}$. The removal of such deformations at low temperatures will occur in the same manner as at high temperatures through the phaseslip mechanism. The pulse duration memory effect observed in $K_{0,3}MOO_3$ [3] becomes clearer with such an interpretation. The length over which the CDW can retain deformations can be very small at low temperatures. According to the results obtained in [34], this length has been estimated to be about 500 Å for TaS₃ at liquid-helium temperature. Then the threshold field $E_0(0) = V_0/l \approx 400 \text{ V cm}^{-1}$ (figure 4; see also [4]). It should also be noted that, in samples with a coherent CDW motion the phase slips and the additional V_0 develop mainly near contacts [14–16]. It seems that the more vertical $\sigma(E)$ dependence at $E > E_T$ observed in the four-terminal configuration can be explained on this basis.

In some sense, the phase-slip process can be considered as a transition between two potential minima in the energy dependence of the CDW phase which differ by $\Delta \varphi \simeq 2\pi$. Then the energy barrier for one electron will be approximately $f_0 \simeq 20$ K. Beyond this average energy the transition of 2π solitons across the barrier at $E < E_T$ and the CDW depinning at $E > E_T$ will be significantly enhanced. At low temperatures (T < 20 K) and low electric fields $E < E_T$, the motion of solitons is probably through a tunnel mechanism. During a phase slip, the CDW order parameter vanishes in a single unit cell of the CDW superstructure which contains about a hundred electrons [15]. Therefore one deals with the tunnelling of many particles, i.e. probably with macroscopic quantum tunnelling [36, 37]. However, the question of the application of this concept to CDW motion needs further experimental and theoretical studies.

3.4. Temperature dependence of E_T

As can be seen from figure 4, the threshold field $E_{\rm T}$ decreases exponentially with increasing temperature according to equation (1) in good agreement with theory [38].

The effective barrier height V_0 decreases in the same manner and becomes very small, about $10^{-4}-10^{-5}$ V at 100 K. The appropriate value of the depinning field due to the CDW deformation over a length of about 0.1 μ m is about 1 V cm⁻¹. The phase-slip processes for o-TaS₃ have been investigated at length in [14, 15] for T > 100 K. The small barrier height does not prevent the spontaneous CDW motion due to thermal fluctuations. Then the CDW pinning and the threshold value for T > 100 K are provided by the pinning of weak impurities in the sample volume. As a first approach it can be suggested that the pinning force due to strong and weak pinning centres is additional. The $E_T(T)$ dependences for samples with various impurity contents (figure 4) provide some evidence in favour of this suggestion; the increase below about 100 K is added to the intrinsic value of E_T above 100 K, which depends on the defect concentration.

Finally, above we have mainly discussed the case of a single domain with CDW coherence through its volume. Actual samples contain many domains. The beginning of the CDW motion and the dislocation motion in one of them can lead to a redistribution of CDW strains and deformations within the sample which may yield an avalanche growth of the current. Moreover the cross sectional area of the conductivity channel provided by CDW motion can be smaller than the cross sectional area of the sample, because the weak transverse interaction between chains (especially for TaS_3 and $K_{0.3}MoO_3$) puts a limitation on the transverse dislocation motion. This picture is experimentally observed for $K_{0,3}MOO_{3}$ [3]. In this case the increase in the total current can be provided by the broadening of the conducting channel almost at constant current density within it. Taking this suggestion into account, it is possible to explain the existence of a vertical region in I-V curves observed for K_{0.3}MoO₃ at low temperatures and the proportionality of the oscillation frequency to the total current I. In these conditions the oscillations can have a relaxation character and they seem to be provided by non-stable regions in the I-V curves. The oscillation frequency depends on the impedance of the external circuit and the sample impedance which changes with the current variation. The oscillation frequency ω may be proportional to the value $(RC)^{-1} \sim \sigma C^{-1} \sim IC^{-1}$, which agrees with experiment (*R* is the real part and *C* is the capacitive part of the impedance).

4. Conclusions

We have measured the electrical conductivity of m-TaS₃ and o-TaS₃ with different impurity concentrations as a function of electric field and of temperature down to liquidhelium temperature. We have shown that the threshold field E_T above which the conductivity increases sharply is for both varieties of TaS₃ exponentially dependent on temperature (from about 100 to 4.2 K) and independent of the purity, suggesting two pinning mechanisms: weak impurities in the volume are essentially efficient above about 100 K and intrinsic strong pinning at low temperatures. At lower electric field and low temperatures, non-linearity occurs at a threshold $E'_T < E_T$ which varies linearly with T. These results provide evidence for a significant contribution of phase-slip processes to the non-linear conductivity and to the CDW depinning. This mechanism is common for CDW depinning at high temperatures as at low temperatures; it is in some sense the trigger mechanism. The description of the experimental properties that we have presented gives the microscopic physical picture of the CDW motion in crystals with strong pinning effects.

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