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The influence of temperature and electric field history on the conductivity of the charge density wave system o-TaS_3

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Abstract

We have studied the dependence of the low electric field conductivity of the charge density wave (CDW) system o-TaS_3 on the temperature and applied electric field history. Without the application of high electric fields, the conductivity is higher on cooling than on heating in a wide temperature range below the CDW transition. With the application of increasing field at a given temperature the conductivity evolves towards a stable, history-independent value situated between the cooling and heating values. The evolution is gradual but most pronounced around the threshold field for nonlinear conductivity. The relative change of conductivity after the application of high field is asymmetric in respect of the temperature history. We discuss the results within the model of quantized changes of the CDW wavelength induced by the variation of the temperature and the electric field.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Various metallic systems with a quasi-one-dimensional Fermi surface undergo a transition to the new collective state [1] known as the charge density wave (CDW) at a finite, so-called Peierls transition temperature T_P. Anomalous polarizability of a 1d electron system enhances the electron–phonon coupling, stabilizing periodic electron modulation and corresponding lattice distortion with the wavevector Q of $2k_F$. Due to the new periodicity, an energy gap of 2Δ forms around the Fermi energy E_F.

CDW systems exhibit various unusual phenomena which include nonlinear conductivity (sliding) above a finite, although low electric field as well as slow dielectric relaxation with extremely high dielectric constant. Such complex low energy dynamics is due to the acoustic-like excitations of the phase of the complex order parameter coupled to random defects (vacancies, interstitials, impurities) and free carriers excited over the gap. Defects destroy the long range phase coherence, break CDW into domains and pin it to the preferential position, while free carriers screen the phase distortions and govern the degree of interaction between the domains.

While probably not as impressive as other phenomena, temperature hysteresis, observed so far in electrical conductivity [2–6], thermocurrent [7, 8], thermal expansion [9], IR transmission [10] and dielectric response [11] of several CDW systems, such as o-TaS_3 or $K_{0.3}MoO_3$, is still puzzling. Typically, temperature hysteresis can be observed in the narrow range around the first-order phase transition temperature or around the glass transition temperature, in which case it depends strongly on the rate of temperature variation. In this respect, the hysteresis in CDW systems is unique. It is independent of the rate of temperature variation, spans a very wide temperature range between two transition temperatures, T_P and the temperature of CDW freezing T_g [12], and it can be reverted from the heating to the cooling curve and vice versa within 10 K by reverting the temperature variation direction.

Such behaviour has been explained [13] by the temperature evolution of the CDW wavevector Q. On one hand, due to inherent electron–hole asymmetry [14], equilibrium CDW wavevector Q_{eq} changes by 2% from T_P down to T_g in CDW systems that exhibit temperature hysteresis [15, 16]. On the other hand, in any finite CDW system Q can change only discontinuously [13], corresponding to the addition/removal of at least a single wave front. This

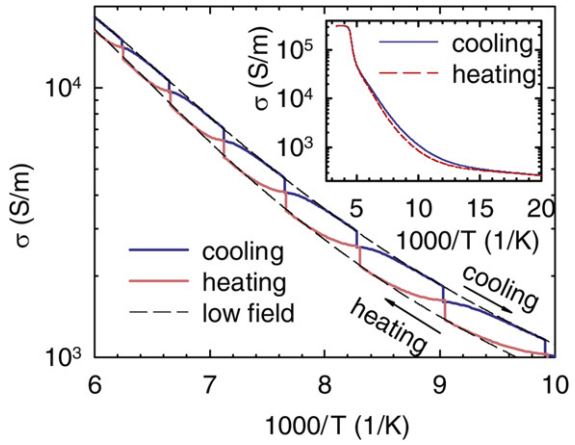

Figure 1. Low field conductivity of o-TaS_3 measured during heating and cooling. Data in the inset are obtained in low electric field only and are represented with dashed black lines in the main panel. Discontinuous lines in the main panel are obtained after the application of electric field exceeding several times E_T at selected temperatures.

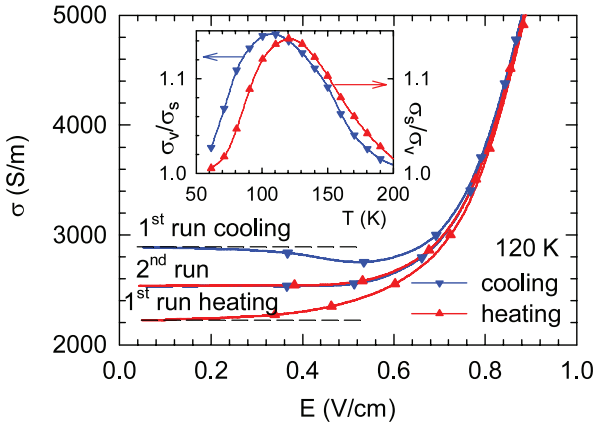

Figure 2. Electric field dependence of conductivity in the first two runs after heating and cooling to 120 K. Inset: temperature dependence of the ratio of low field conductivities between virgin (σ_v) and stable (σ_s) state values on cooling and heating.

process requires a finite energy [17] so Q will always lag behind Q_{eq} and its value at a given temperature will depend on the temperature history. As different Q values correspond to different free carrier densities [14] the history dependence of Q is seen as the hysteresis in conductivity.

However, this theory does not provide a clear picture of how the evolution of Q and the corresponding conversion between free and condensed carriers affects the low energy properties of CDW. Hysteresis has been observed in the low frequency dielectric response of the CDW system o-TaS_3 [11] with the value of low frequency dielectric constant $\epsilon(0)$ being higher on heating than on cooling. From the basic properties of the CDW pinning mechanism it follows that $\epsilon(0)$ should be inversely proportional to the value of threshold field E_T for CDW sliding [1], so hysteresis should affect the onset of nonlinear conductivity as well. In order to verify this prediction, we have investigated the effects of temperature history on the onset of nonlinear conductivity in o-TaS_3.

2. Experimental details

We have measured dc conductivity σ_{dc} of several o-TaS_3 samples in different regimes of temperature and electric field (E) evolution. We have used the standard four-contact configuration with a Keithley 220 current source and a Keithley 2182 nanovoltmeter. The samples, typically 5 mm long and with a cross-section of about 10^{-4} mm^2, have been attached with silver paint on four predefined copper contacts on a thin epoxy glass base. The inner contact separation was 2 mm. Typical resistance of an o-TaS_3 sample at room temperature is of the order of 10–100Ω, while E_T below the CDW transition at 220 K is of the order of 100 mV cm^{-1}.

We have measured linear σ_{dc} during temperature scans on heating and cooling. At selected temperatures several V–I characteristics have been recorded up to the fields several times exceeding E_T. During the temperature scans, electric field was typically 5 mV cm^{-1}, which is sufficiently below E_T to ensure linearity. We have investigated temperatures below 300 K down to about 20 K, where o-TaS_3 becomes insulating in low electric fields. V–I characteristics have been measured in the temperature range of the hysteresis between 60 and 200 K. At 110 K, where the temperature hysteresis is most pronounced, we have not observed any time evolution of low field σ_{dc} for a period of over 1 h.

3. Results and discussion

We present in figure 1 linear σ_{dc} measured on cooling and heating. The inset represents 'regular' σ_{dc} obtained between 300 and 50 K without application of high electric fields during the temperature scan. It features all characteristics of o-TaS_3 linear σ_{dc} [18]; the Peierls transition at 220 K is seen as a steep decrease of σ_{dc} followed by the activation regime down to 100 K and then a crossover to a plateau region down to 50 K. The well known hysteresis loop [2] is present in the wide temperature range below 200 down to 50 K.

In the main panel we present the temperature evolution of linear σ_{dc} after application of field exceeding several times E_T at selected temperatures. After the application of high electric field, σ_{dc} changes to the same value inside the hysteresis loop regardless of temperature history [5]. However, as temperature is changed further, σ_{dc} gradually merges with the corresponding branch of the hysteresis loop within 10 K.

In figure 2 we present the evolution of σ_{dc} as the electric field is gradually increased above E_T and then decreased again. In the first ('virgin') scan at a given temperature σ_{dc} is field dependent even at lowest fields and evolves slowly up to E_T where an abrupt increase of σ_{dc} starts. Depending on the temperature history, virgin σ_{dc} either increases (on heating) or decreases (on cooling). E_T is not well defined for the CDW state corresponding to σ_{dc} values on the hysteresis loop and therefore cannot be compared to the results of dielectric spectroscopy.

As the electric field is decreased from values exceeding E_T, σ_{dc} becomes constant ('stable') below E_T and the

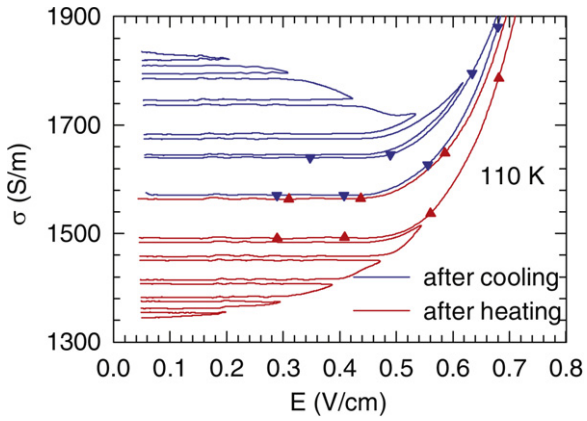

Figure 3. A partial relaxation process performed at 110 K with six field cycles after cooling and heating. Every subsequent cycle has been performed using a higher maximum electric field. Curves for each cycle are artificially shifted with respect to the previous one for clarity.

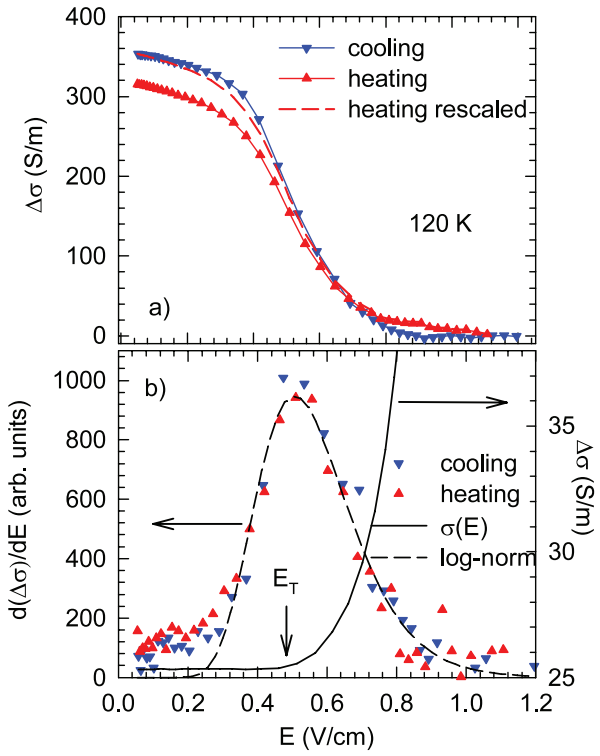

Figure 4. Upper panel: the difference between conductivity in the virgin and the stable state after cooling and heating, as well as the rescaled difference after heating. Lower panel: the derivative of conductivity difference in respect of electric field together with the conductivity of the stable state. The dashed line represents the fit to the log-normal distribution to be discussed later in the text.

same regardless of temperature history. Furthermore, any subsequent V–I characteristics give the same $\sigma_{dc}(E)$ dependence which now allows a proper estimate of E_T. However, as the application of sufficiently high field erases CDW memory of temperature history, E_T is the same for both cooling and heating.

The amount of σ_{dc} change between virgin and stable values depends significantly on temperature history. The inset of figure 2 presents ratios of low field σ_{dc} before (virgin state) and after (stable state) application of high field. The change of σ_{dc} is more pronounced at low temperatures on cooling and at high temperatures on heating.

In order to test the nature of transition from the virgin state of σ_{dc} to the stable one we have measured a series of V–I characteristics (cycles) with increasing maximum applied field in each cycle. Figure 3 shows corresponding $\sigma_{dc}(E)$ curves obtained at 110 K.

We have obtained that the maximum applied field uniquely defines a new intermediate value of σ_{dc} which remains stable until a field exceeding the previous maximum is applied. σ_{dc} reaches the stable state only after a field several times higher than E_T is applied.

In this respect we can deduce the evolution of low field σ_{dc} with applied field ($\Delta\sigma(E)$) from the difference between conductivity values in virgin and stable $\sigma_{dc}(E)$, as presented in figure 3.

Apart from the multiplicative factor, the $\Delta\sigma(E)$ dependence is the same after cooling and heating, upper panel of figure 4. As is shown in the lower panel of figure 4, the strongest change of $\Delta\sigma(E)$, seen as a maximum in its derivative against E, occurs at E_T.

We have demonstrated that the evolution of dc conductivity with increasing electric field from the low field virgin state at the hysteresis loop proceeds gradually, through the cascade of metastable states for a given field, until a truly history-independent stable state is reached for fields about two times higher than E_T. In this respect, no E_T for this initial $\sigma_{dc}(E)$ curve can be defined, but only for subsequent sweeps starting from the stable state. Also, at a given temperature the evolution of the virgin state with field shows the same functional dependence but with different amplitudes on cooling and on heating.

In order to explain these findings, we have to consider both the nature of metastable states of CDW and the mechanism by which the evolution of these metastable states with temperature and electric field affects σ_{dc}. In the first report of temperature hysteresis in σ_{dc} of o-TaS_3 [2], as well as in the numerical simulations of weakly pinned CDW [21] it has been suggested that the temperature evolution of Q is responsible for hysteresis. As Q is nearly commensurate, the ground state consists of sections of commensurate CDW separated by discommensurations (DC). Equilibrium distribution of DC changes as Q varies with temperature; however, CDW pinning prevents DC equilibrating at a given temperature which leads to hysteretic behaviour. Subsequently, temperature hysteresis has been attributed to the creation of CDW defects which either decrease or increase free carrier population with the variation of temperature [3] without any reference to the nature of these defects.

Eventually, it was recognized that the value of Q can change only through the condensation of free carriers in CDW [13, 14] which naturally affects σ_{dc}. This process, which in the presence of pinning creates locally a topological defect in CDW, requires finite energy and therefore leads to hysteresis when the equilibrium value of Q varies with temperature.

All these explanations require the creation or annihilation of topological defects in the CDW configuration. Therefore CDW would evolve towards a more disordered state with increased number of domains as temperature decreases. However, measurements of the low frequency dielectric response indicate that the distribution of low energy excitations does not change with temperature [12], nor does it depend on temperature history [11]. In the following we propose a model which is similar to [13] but does not require the formation of additional defects in order to account for phenomena we observe.

First we recount the model of temperature evolution of Q in CDW systems which relates the value of Q to the density of thermally excited carriers and therefore to the value of σ_{dc}. Opening of the gap at E_F below T_P makes the CDW system semiconducting, as only free carriers excited across the gap contribute to low field σ_{dc}. However, the nature of this semiconducting state depends on microscopic details of the electronic structure of the conduction band and on the band filling, i.e. on the position of E_F within the band [14].

For the simplest tight binding model, when the conduction band is less than half filled in the metallic state and a gap opens below the middle of the band, the density of hole states is higher than the density of electron states near the edges of new valence and conduction bands. Thus at a finite temperature thermally excited holes outnumber electrons and the conductivity in the CDW state is p-type, as observed in o-TaS_3, which has a $1/4$ filled band. If the band is more than half filled, as in $K_{0.3}MoO_3$ which has a $3/4$ filled band, then the conductivity is n-type.

The electron–hole asymmetry affects also the number of electrons condensed in the CDW which in turn determines the value of Q. Equilibrium Q value Q_{eq} at a finite temperature deviates from the zero temperature value $Q(0)$, proportionally to the difference in the density of thermally excited holes and electrons [14]. The change of Q changes also the position of the CDW gap in respect to E_F, so that the middle of the gap moves away from E_F.

If hole conductivity is dominant, as in o-TaS_3, the contribution of thermally excited electrons can be neglected and $Q_{eq}(T)$ will be larger than $Q(0)$ in proportion to the density of thermally excited holes p:

$$\frac{Q_{eq}(T)}{\pi} - \frac{Q(0)}{\pi} = p(T) \sim \sigma_{dc}. \tag{1}$$

Therefore the values of Q and σ_{dc} are mutually dependent.

The temperature dependence of p can be parametrized as

$$p(T) \sim \exp\left(-\frac{\Delta^*}{k_B T}\right), \qquad \Delta^* = \Delta - \Delta\mu \tag{2}$$

where Δ^* is the effective activation energy, Δ is the gap halfwidth and $\Delta\mu$ is the deviation of the chemical potential position from the middle of the gap. As temperature and consequently hole density decrease, $Q_{eq}(T)$ decreases towards $Q(0)$, which has been observed in o-TaS_3 [15]. On the other hand, in $K_{0.3}MoO_3$, where the electron conductivity is dominant, $Q_{eq}(T)$ is smaller than $Q(0)$ and approaches $Q(0)$ as temperature and electron density decrease [16].

Next we argue that, unlike Q_{eq}, the real value of Q cannot change continuously and lags therefore behind Q_{eq} which leads to the temperature hysteresis. If we consider the portion of CDW between two defects, Friedel oscillations around defects impose rather strict boundary conditions for CDW [19, 20]. Therefore, Q can assume only the values which are roughly the multiples of $2\pi/L$, where L is the domain length along the CDW direction, corresponding to the integral number of wavelengths within the domain. Correspondingly, Q can change within the domain only by a finite amount, through the addition/removal of one CDW wave front [13, 21]. As this requires the condensation of free carriers in CDW or their removal from CDW, the process of so-called phase slip has to occur [13, 21]. Phase slip corresponds to the local collapse of CDW which requires overcoming a finite energy barrier [17].

We consider first the ideal case of an isolated domain. If initially $Q = Q_{eq}$ at a given temperature, the change of temperature increases the difference between Q and Q_{eq} as Q remains constant and thus imposes stress on the CDW [22]. Chemical potential therefore shifts towards either the valence or the conducting band edge, as the density of free carriers remains unchanged [22]. This additional shift ξ can be parametrized as

$$p(T) \sim \exp\left(-\frac{\Delta^* - \xi(T)}{k_B T}\right). \tag{3}$$

Eventually, if the temperature change is sufficient, strain energy becomes high enough to overcome the barrier towards a new CDW configuration with Q within the domain closer to Q_{eq} at this new temperature [17]. According to (1) it results in the change of density of free carriers, and consequently of σ_{dc}.

In general, as Q cannot assume exactly the value of Q_{eq} due to the boundary conditions, the value of Q at low temperatures, where Q_{eq} does not differ significantly from $Q(0)$, will be fixed once it comes within $\pm\pi/L$ from $Q(0)$. In larger domains, in which the change of Q is smaller, this will happen at lower temperatures.

Maximum possible strain of uniform CDW corresponds to the shift of chemical potential almost all the way to the band edge [17] which destroys the CDW state and allows recondensation with a new Q. In this hypothetical case, critical shift of chemical potential ξ_{cr} on cooling is negative and equal to $-(\Delta - \Delta\mu)$ as chemical potential approaches the majority carrier band edge, while on heating it approaches the minority carrier band edge and ξ_{cr} is positive and equal to $\Delta + \Delta\mu$. As $|\xi_{cr}|$ is larger on heating, temperature should therefore change more than on cooling in order to induce the phase slip. We will discuss within this approach the asymmetry between cooling and heating which we observe in the temperature dependence of the σ_v/σ_s ratio (inset of figure 2) as well as in $\Delta\sigma(E)$ at a given temperature (figure 4).

Although in a real system fluctuations, non-uniformity, interaction between adjacent domains or creation of topological defects induce phase slip at lower strains, asymmetry between cooling and heating should persist. For instance, it has been demonstrated [23] that the creation of a charged soliton, which is a step in the phase slip process [24], requires less energy if

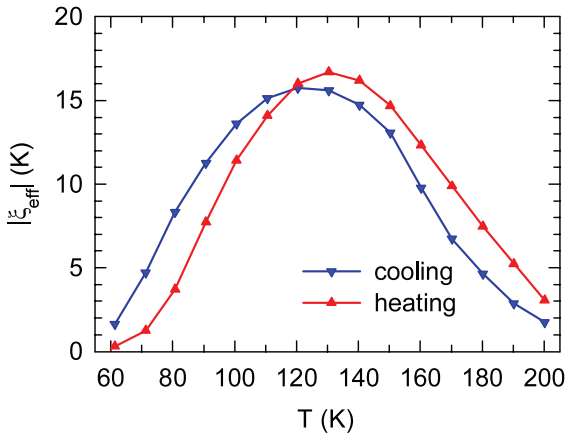

Figure 5. Absolute value of the shift of chemical potential $|\xi_{\rm eff}|$ due to CDW strain during cooling and heating of the sample, calculated from (4). In heating the shift is negative.

the charge of the soliton is the same as for majority carriers. So phase slip requires less energy on cooling, when majority carriers condense in the CDW.

Application of external electric field distorts the CDW, so that it becomes more compressed on one side of the domain and more expanded on the other [19, 22]. This corresponds to a local decrease or increase of Q, and therefore of ξ, near domain edges which facilitates a phase slip process on one side of the domain. If external field is sufficiently high, Q changes eventually to the value within π/L from $Q_{\rm eq}$ and then remains stable at a given temperature.

In a real system with a random distribution of defects domains are of various sizes, and so is the amount of possible change of Q. Phase slips do not occur simultaneously throughout the sample and both temperature and electric field evolution of $\sigma_{\rm dc}$ are quasicontinuous.

As the application of sufficiently high electric field puts the CDW in a stable, nearly strain-free state, we can calculate using (3) an effective, or averaged chemical potential shift $\xi_{\rm eff}$ from the ratio of virgin and stable state conductivities in the inset of figure 2 as

$$\xi_{\rm eff} = k_{\rm B}T \log\left(\frac{\sigma_{\rm v}}{\sigma_{\rm s}}\right). \tag{4}$$

The temperature dependence of $|\xi_{\rm eff}|$ is presented in figure 5, with $\xi_{\rm eff}$ negative on cooling.

At high temperatures phase slip is facilitated by thermal fluctuations and $|\xi_{\rm eff}|$ disappears at $T_{\rm P}$ because fluctuations destroy even the CDW order. As temperature decreases, fluctuations are suppressed and $|\xi_{\rm eff}|$ becomes larger. At even lower temperatures $Q_{\rm eq}$ comes so close to $Q(0)$ that in some of the domains Q is not able to change further, even in an external electric field. The electric field induces phase slip only in a fraction of domains and smaller corresponding change in conductivity results in a decrease of $|\xi_{\rm eff}|$. At low enough temperatures all domains are 'frozen' and $|\xi_{\rm eff}|$ drops to 0.

Asymmetry of $|\xi_{\rm eff}|$ on cooling and heating close to $T_{\rm P}$ is due to the asymmetry in $|\xi_{\rm cr}|$. If $|\xi_{\rm cr}|$ is smaller, as on cooling, ξ does not exceed this value in any domain, so the average $\xi_{\rm eff}$ is smaller as well. The origin of asymmetry at low temperatures is slightly different, but stems again from the asymmetry in $|\xi_{\rm cr}|$.

As temperature is lowered, in more and more domains Q is fixed to a value close to $Q(0)$, while $Q_{\rm eq}$ becomes equal to $Q(0)$. On heating, as $Q_{\rm eq}$ starts to deviate from $Q(0)$, phase slip occurs only after $Q_{\rm eq}$ first reaches a fixed value of Q in a particular domain and then deviates further, which means at a higher temperature than on cooling. This is why at low temperatures the $|\xi_{\rm eff}|$ curve on heating appears to be shifted to higher temperatures with respect to cooling.

In terms of ξ, it has first to decrease and then become negative in order to induce the 'inverse' phase slip which will take majority carriers out of the CDW. Unless ξ is negative, electric field does not initiate this process either, because chemical potential approaches first the majority carrier band edge and this starts the phase slip process contributing to nonlinear conductivity but not to the stable change of Q.

We have demonstrated that the evolution of $\sigma_{\rm dc}$ from the state on the hysteresis loop follows the same electric field dependence on cooling and heating, apart from the amplitude, figure 4. The difference in $\sigma_{\rm dc}$ between the virgin and the stable state $\Delta\sigma(E)$ changes most around $E_{\rm T}$ (the lower panel of figure 4). Based of the discussion so far, it follows that at a given electric field, all domains that come close to $\xi_{\rm cr}$ undergo a phase slip and their contribution to $\Delta\sigma$ abruptly vanishes. We can model it with the following expression:

$$\Delta\sigma(E) \sim \int_0^\infty P(E')\theta(E'-E)\,{\rm d}E'. \tag{5}$$

E' is the effective threshold field for phase slip in a given domain, corresponding to the difference between $\xi_{\rm cr}$ and ξ in the virgin state. θ is the step function and $P(E')$ is the distribution of E'. In this case $P(E')$ can be obtained as

$$P(E') = \frac{{\rm d}\left(\frac{\Delta\sigma(E)}{\Delta\sigma(0)}\right)}{{\rm d}E}. \tag{6}$$

We have been able to fit $P(E')$ to the log-normal distribution.

$$P(E') = \frac{1}{\sqrt{2\pi}bE'}\exp-\frac{\left(\ln\frac{E'}{E'_0}\right)^2}{2b^2} \tag{7}$$

where b is the standard deviation and E'_0 the median of distribution. The fit at 120 K is presented in the lower panel of figure 4. the steepest decrease of $\Delta\sigma$ is obtained at the position of maximum of log-normal distribution:

$$E'_{\rm m} = E'_0{\rm e}^{-b^2}. \tag{8}$$

The value of b and the ratio of $E'_{\rm m}$ and $E_{\rm T}$ at a given temperature are presented in figure 6.

In the temperature range down to 100 K $E'_{\rm m}$ corresponds well to $E_{\rm T}$, as already noted, but below 100 K it becomes increasingly lower than $E_{\rm T}$. At the same temperature b starts to increase and the distribution broadens. However, this is the temperature region below the maximum in $|\xi_{\rm eff}|$ where the Q of the domains starts to 'freeze', so $P(E')$ represents only the distribution of the sufficiently large domains in which Q can still change.

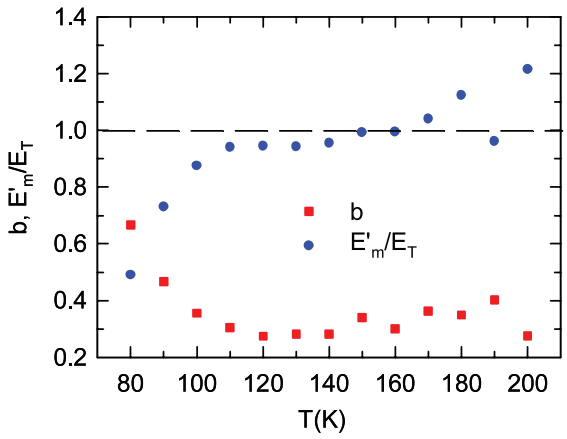

Figure 6. Standard deviation (b) of the distribution of effective threshold fields and the ratio of the maximum of distribution (E'_m) and the actual threshold field (E_T).

4. Conclusion

We have presented a thorough investigation of the effects of temperature and electric field history on the onset of nonlinear conductivity in the CDW system o-TaS_3. From the states on the hysteresis loop σ_{dc} evolves gradually with electric field even for lowest field values and this change can be either monotonic or nonmonotonic (after heating and cooling to the given temperature, respectively) before a sharper increase starts at a finite field. In such a case, the threshold field for nonlinear conductivity and its dependence on temperature history cannot be determined.

We argue that this effect corresponds to the local changes of CDW wavevector within the domains towards the equilibrium value at a given temperature. As a consequence, a unique stable state is reached after the application of electric field exceeding several times the threshold field, regardless of temperature history.

Asymmetry in hysteresis on cooling and heating is found from analysis of the changes of σ_{dc} in the field at selected temperatures. The origin of this asymmetry is the same as the origin of the hysteresis itself: the departure of chemical potential from the gap centre due to self-doping in the semiconducting CDW state.

We have found that the distribution of phase slip processes in domains can be modelled with the log-normal distribution of effective local threshold fields. The position of the distribution maximum corresponds well to the threshold field down to 100 K, but then becomes smaller, while distribution width increases. This might be a consequence of the freezing of Q in domains with high E'.

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