Critical current in charge-density wave transport

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We report transport measurements under very high current densities $j$, up to $\sim 10^8$ A/cm$^2$, of quasi-one-dimensional charge-density wave (CDW) conductors NbSe$_3$ and TaS$_3$. Joule heating has been minimized by using a point-contact configuration or by measuring samples with extremely small cross-sections. Above $j_c \approx 10^7$ A/cm$^2$ we find evidence for suppression of the Peierls gap and development of the metallic state. The critical CDW velocity corresponding with $j_0$ is comparable with the sound velocity, and with $\Delta/hk_F$ ($k_F$ is the Fermi wave vector), which corresponds to the depairing current. Possible scenarios of the Peierls state destruction are discussed.

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The discovery of non-Ohmic conductivity in quasi-one-dimensional charge-density wave (CDW) materials has opened great expectations concerning the so-called Fröhlich superconductivity [1]. However, the experiments up to now have showed that the CDW conductivity under the highest electric fields asymptotically approaches a value close to the "normal-state conductivity" and never exceeds it. This conductivity is estimated from the extrapolation of the temperature dependent conductivity above the Peierls transition [2]. The hypothesis which can be naturally risen, namely that the CDW state is suppressed at high currents was rejected by x-rays measurements of the CDW satellite profiles in the sliding state [3], [4] and by narrow-band noise measurements at high current densities $j$ (up to $3 \cdot 10^4$ A/cm$^2$ for TaS$_3$ [5]): in the latter case, it was shown that the main part of the current is carried by the coherently sliding CDW with no reduction of its charge density. On the other hand, the CDW velocity cannot grow up to infinity. So, the question concerning the existence of an upper limit for the velocity of the sliding CDW is still undecided up to now.

To our knowledge, it has been reported only one indication in favour of the local suppression of the Peierls energy gap $2\Delta$, and that in K$_{0.3}$MoO$_3$ [6]. This effect, attributed to a high current density near the metal – CDW surface was observed at a high degree of injection of normal carriers through a Au–K$_{0.3}$MoO$_3$ boundary. Estimation of the critical current density gave a value of $j_c = 4.8 \cdot 10^7$ A/cm$^2$. However, it is not clear if the gap suppression reported in Ref. [6] is associated with the high CDW velocity, or is induced by the injected quasiparticles. For a clarification of the situation it would be highly desirable to observe this phenomenon directly. Evidently, Joule heating in the previous experimental configurations will not allow to achieve high enough values of $j$.

Hereafter we propose two approaches for overcoming this problem. The first one is the point contact configuration formed between two thin whiskers of NbSe$_3$. It is well known that the electric field is localized near a point contact in a small region with a characteristic size of the order of the point contact diameter, $d$. In the case $d \ll l$ ($l$ is the mean free path) the length for energy relaxation is much longer than that corresponding to the formation of the point contact resistance and the heating effect is strongly suppressed [7]. Thus, a point contact is a very convenient configuration for high current measurements. We have used the configuration proposed in Ref. [8]. The electric contact between two NbSe$_3$ stripe–like single crystals with perpendicular $bc$ – planes is formed at low temperatures by means of a precise mechanical motion transfer system, so that the current through the contacts flows along the $b$-axis. The samples selected for the experiment had typical dimensions: along the $b$-axes $L_b \approx 1$ mm, along the $c$-axes $L_c \approx 10 \div 50 \mu$m and along the $a$- axes $L_a \approx 1 \mu$m.

Disadvantages of the point contact configuration are a probability for having a non-predicted barrier at the boundary between the samples and a non-uniform current distribution. So we have performed high–current measurements of extremely thin samples of TaS$_3$ with a cross-section area $\lesssim 10^{-3} \mu$m$^2$ and a typical length $5 \div 10 \mu$m. The cross-section area is estimated from the value of the room-temperature resistance of the samples [9]. Due to the very good thermal contact with the sapphire substrates such samples can sustain extremely high current densities with a relatively small Joule heating. The main results presented below are insensitive to the exact value of heating.
Let first analyse our results with the point contact configuration. Figure 1 shows typical dependencies of the differential resistance $R_d(I)$ for three different NbSe$_3$-NbSe$_3$ point contacts obtained at $T=77$ K. $R_d(I)$ monotonously decreases with the increase of the injected current revealing the CDW sliding. The threshold electric field for initiation of the CDW sliding, $E_T$, is achieved at a very small current, so the plateau in the curves corresponding to the pinned state of the CDW is indistinguishable in this scale. At very high current $R_d$ is close to the resistance values which would be observed in the absence of the Peierls transition [2]. In this region, practically for all investigated contacts we clearly observed a sharp decrease of the resistance at the current $I_0$ (indicated by arrows in Fig.1). Above $I_0$ the resistance is slightly growing with current, evidently reflecting the Joule heating. The growth of $R$ with heating should be the case for a metallic state. We could not measure the $R_d(I)$ dependence far above $I_0$ because the contacts are very unstable in this region and often burn.

The exact determination of the CDW current density corresponding to $I_0$ is complicated in the case of NbSe$_3$. The unit cell of NbSe$_3$ contains three different types of chains. In the temperature range from 59 K to 145 K two types of chains are in the normal state and one is in the CDW state [2], but at high electric field we can assume that the injected current is homogeneously distributed over the chains, because in this case the CDW conductivity is close to the normal-state value. So $I \approx j \pi d^2/4$, where $j$ is the current density, and the point contact diameter may be estimated from the well known Sharvin formula [11]: $R_{ds} = l\rho/d^2$, where $\rho$ is the normal state resistivity and $R_{ds}$ is the saturation value of $R_d$ at high current. Fig. 2 shows the variation of $I_0$ as a function of $1/R_{ds}$, which is proportional to the point–contact area, for more than 20 different point contacts. The dependence is close to be linear. The proportionality between $I_0$ and the conductivity means that the current density is constant for all contacts and is evidently determined by fundamental properties of the material. For $\rho = 3 \cdot 10^{-4}$ $\Omega$cm (Ref [11]) and for $l=100$ nm, we estimate the mean value for the CDW current density $j_0 = 5.9 \cdot 10^7$ A/cm$^2$.

For making clear if the abrupt change in conductivity of NbSe$_3$ at $j \sim 10^7$ A/cm$^2$ really reveals a transition into a metallic state, i.e. a critical (depairing) current for the CDW, or is associated with contact phenomena under inhomogeneous conditions, we performed transport measurements of extremely fine samples of TaS$_3$. We selected this material, as it demonstrates a purely dielectric behavior below $T_P \approx 220$ K. So, a transition into a metallic state should be more obvious for it. Besides, TaS$_3$ crystals can be easily splitted and make possible the preparation of samples with a cross section area below $10^{-3} \mu m^2$.

Typical results of transport measurements are shown in Figure 3, which illustrates “raw” dependencies of the conductivity of a representative sample at different temperatures as a function of the current density (open symbols). These data have to be corrected of Joule heating. We have estimated its value by two ways. First, we noticed, that at temperatures well above $T_P$ (typically $T > 250$ K) the non-linear conductivity due to CDW fluctuations becomes negligible; therefore, the slight change of resistance with voltage, $\delta R \equiv R(V) - R(0) \propto V^2$, is only provided by heating. The comparison of $\delta R$ with $dR/dT$ at several temperatures yields the estimate of heating as a function of Joule power, $W$. The second estimate is based on the dependencies $R(j)$ obtained at low temperatures. We noticed that at the highest currents the temperature of the samples exceeds $T_P$, and that at a certain current density each curve $R(j)$ (and $R_d(j)$) demonstrates
a minimum corresponding with the minimum of $R(T)$, which is observed around $T_M = 250 \pm 300$ K. Thus, the power $W_M$ at the minimum of $R(j)$ provides the estimate of heating $\delta T = T_M - T$, where $T$ is the ambient temperature. The dependence $W_M$ vs. $T$ is approximately linear, its slope giving the required value $dT/dW$. Both ways give $dT/dW \approx 10^5$ K/W for samples with dimensions $5 \mu\text{m} \times 10^{-3} \mu\text{m}^2$. Knowing the value of $W$ at each $V$ and $dT/dW$ we thus know the temperature for each point of each curve. Interpolating the set of I-V curves we got isothermal dependencies and thus the correction for heating. The curves obtained after this temperature correction are shown in Fig.3 (solid lines). One can see that the curves for $T < T_P$ intersect at $j = 10^7$ A/cm$^2$. For $j > 10^7$ A/cm$^2$ the growth of conductivity tends to saturate value. However, we cannot determine exactly the asymptotic behavior of the curves, because our results for high currents are very sensitive to the value of $dT/dW$ used for the temperature correction.

To ascertain if TaS$_3$ really exhibits a transition into a metallic state, we have plotted the conductivity measured at a given current as a function of temperature. Fig.4 shows such a set of dependencies. For comparison we also show the temperature dependence of the conductivity at a low field $E \ll E_T$ ($E_T \approx 25$ V/cm). For $E \lesssim 3$ kV/cm the temperature dependence of the conductivity demonstrates a dielectric behaviour ($dR/dT < 0$). While for $E \lesssim 500$ V/cm, the high-field conductivity approximately follows the behaviour of the conductivity measured at low-field [12], at higher $E$ the activation energy decreases, and for $E_0 = 3.3$ kV/cm ($j_0 = 10^5$ A/cm$^2$) the conductivity is nearly independent of temperature. Increasing the current further results in the metallic behaviour of conductivity, i.e. $dR/dT > 0$. We emphasize that this result and the value of the current $j_0$ are quite insensitive to the value of $dT/dW$ taken for the Joule heat correction [13].

The value of the non-linear contribution to $j_0$ as a function of temperature is presented in Fig.5 for TaS$_3$ (black circles) and for a stable point-contact NbSe$_3$-NbSe$_3$ for which we succeeded in obtaining the temperature dependence of the current $I_0$ (open circles). Both temperature and current scales are normalized by the value of $T_p$ (145 K for NbSe$_3$ and 208 K for TaS$_3$) and the value of $j_0$ at $T = 0.62T_p$ respectively. As seen in the figure, the value of $j_0$ for both materials has a little tendency to decrease with increasing temperature. The growth of $j_0$ for TaS$_3$ at $T \to T_P$ is probably due to the growth of non-collective contributions to the current. In fact, at the highest temperatures (Fig.5) the linear contribution to $j_0$, i.e. $j_0 \propto R/R(0)$, is comparable with the non-linear part of the current.

![Graph](image-url)
Analyzing our experimental results with the point contact technique we are led to the assumption that the observed anomalies at \( j = j_0 \) are related to the suppression of the CDW state at high current density. Indeed, the conductivity of the CDW is less than the normal state conductivity at any current. So, the sharp drop of the resistance of the NbSe\(_3\)-NbSe\(_3\) point contacts reveals the transition from the CDW conductivity to the normal state conductivity. Similarly, a metallic behaviour is demonstrated in very small cross-section TaS\(_3\) samples above \( j_0 \). The fact that the transition into the metallic state in the latter case is more gradual can be ascribed to the non-uniform distribution of the CDW velocities in the volume of the sample, while the point contact probes only several wavelengths. Evidently, the transition of TaS\(_3\) into a metallic state is not complete, and in the vicinity of \( T = T_0 \) the dielectric behaviour is observed up to the highest fields (Fig.5). By analogy with superconductors this may be associated with the development of a kind of mixed state in the CDW at high currents.

The magnitude of the critical current densities for different CDW materials are approximately the same: \( j_0 = 5.9 \cdot 10^7 \, \text{A/cm}^2 \) for NbSe\(_3\), \( 10^7 \, \text{A/cm}^2 \) for TaS\(_3\) and \( 4.8 \cdot 10^7 \, \text{A/cm}^2 \) for K\(_0\)MoO\(_3\). It has to be noted that, for the latter compound, the suppression of the gap has been observed directly.

What could be the physical mechanism of this transition? The theoretical description or even the consideration of this phenomenon does not exist up to now. There-fore, we can only propose qualitative explanations of the effects we have observed. First of all, let estimate the velocity of the CDW motion corresponding to the critical current density, \( j_0 \). The usual formula \( v = j_0/n \), where \( n \) is the density of condensed carriers, yields \( v = 2.7 \cdot 10^5 \, \text{cm/s} \) for NbSe\(_3\), \( 0.2 \cdot 10^5 \, \text{cm/s} \) for TaS\(_3\) and \( 0.6 \cdot 10^5 \, \text{cm/s} \) for K\(_0\)MoO\(_3\). These velocities, \( v_s \), in these materials \([14]\). As the CDW results from an electron-phonon interaction, it is unclear how the CDW could survive at \( v > v_s \). So, anomalies in the Peierls gap behaviour, presumably its suppression, is expected at \( v \sim v_s \).

The critical current can also reveal the electron-hole depairing, by analogy with superconductors. In the sliding state, the Fermi distribution \( \varepsilon(k) \) is shifted by a quantity \( 6k = k_F v_F / v \) and distorted by \( \delta \varepsilon = +v q/2 \) at \( k = \pm k_F \), where \( q = 2k_F \) is the CDW wave vector. One may consider that the CDW gap will be suppressed when the distortion \( \delta \varepsilon \) will reach a value comparable to \( \Delta : v q \sim \Delta \). This gives the estimate of depairing current density \( \lesssim 10^8 \, \text{A/cm}^2 \). The same estimate arises from quite another consideration. Note, that the frequency of the narrow-band noise generation (the so-called fundamental frequency) \( f \approx v q / 2 \pi r \) for the depairing current density is \( \sim \Delta / h \). Suppose, at some point (say at a pinning center) the gap is suppressed, \( i.e. \) a phase slip (PS) act occurs. From the principle of uncertainty, the lowest possible time for the gap suppression or recovering is \( \sim h / \Delta \), which could be treated as the lowest possible time of a PS act. If the PS frequency exceeds the critical value, the gap has no time to restore before another wavelength comes, and the area of the gap suppression expands throughout the sample. So, the PS time limitation from below gives the same value of critical current. Nucleation of the metallic state resulting from a continuous increase of PS processes was considered earlier as an approach to the Peierls transition from low temperatures \([10]\).

In conclusion, we have for the first time considered the question about the critical velocity of the CDW and proposed an experimental answer to it. For TaS\(_3\) and NbSe\(_3\) we clearly observed a Peierls state – normal metal transition at \( j_0 \sim 10^7 \, \text{A/cm}^2 \). The critical velocity, \( v_0 \), of the sliding CDW corresponding to the value of \( j_0 \) approaches the speed of sound, and is comparable with \( \Delta / q \).

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