Classical-to-Quantum Crossover in Charge-Density Wave Creep at Low Temperatures

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It is found that the thermally activated behavior of nonlinear conduction below the threshold field in thin samples of orthorhombic TaS_3 vanishes at temperatures below 10 K. Current noise appearing in the low-temperature region indicates that the electric current is transferred by portions of charge which correspond to displacement of the charge-density waves (CDW) by a distance of the order of their wavelength. This phenomenon can be accounted for as a transition from thermally activated to quantum creep of the CDW.

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Quasi-one-dimensional conductors, such as TaS_3 and $K_{0.3}MoO_3$, are well known physical systems which exhibit nonlinear conduction due to collective transport of charge-density waves (CDWs) [1]. Below the threshold electric field, E_T, the CDW is pinned by impurities and the electric current is transported by normal carriers (electrons and holes) thermally excited above the Peierls gap. When the electric field E exceeds E_T, the CDW slides along the crystal, giving a nonlinear contribution to the electric current.

It is generally accepted that the kinetic properties of CDWs are temperature dependent down to zero temperature. It has been found, in particular, that in both orthorhombic and monoclinic TaS_3 $E_T(T)$ follows an exponential dependence, $E_T \propto \exp(-T/T_0)$ (T_0 = 20–25 K), at least down to 4.2 K [2], and that the origin of this dependence may be associated with thermal fluctuations of the order parameter [3]. A large number of both theoretical and experimental results have led to the conclusion that temperature-dependent screening by normal carriers also plays an essential role in the kinetics of CDWs (see [1] and references therein). At relatively high temperatures, this leads to a scaling relation between the temperature dependences of the linear conductivity and that of the viscosity inhibiting CDW motion. At liquid-helium temperatures, the low-temperature divergence of the viscosity in semiconducting CDW compounds (i.e., all materials except $NbSe_3$) leads to a transition into another regime of sliding (so-called "Fröhlich superconductivity" [4, 5]). In this regime an increase of the current by up to 7 orders of magnitude is observed within a few percentage variation of the electric field.

Freezing out of the normal carriers also reveals nonlinear conduction for fields less than the threshold field, $E < E_T$ [2, 4, 6]. The temperature dependences of both the linear and nonlinear conduction, as well as the magnitudes and forms of the low-temperature I-V curves, depend critically on the crystal quality [2]. In the particular case of o-TaS_3, lowering the temperature causes a reduction of the activation energy for linear conduction from 800 K ($T > 100$ K) down to 250 K (20 K $< T <$ 80 K), and an even further reduction at $T < 20$ K [2, 6]. Features of the temperature-dependent conductivity suggesting variable-range hopping, $I/V \propto V^{0.7-1.5}$ and $I(T) \propto \exp[(-T/T_0)^{1/2}]$, were found in some crystals of o-TaS_3 below 20 K (see [2] and references therein).

The present paper reports the results of an investigation of the electrical conduction of thin samples of o-TaS_3 at low temperatures down to 2 K. It is found that such samples exhibit a crossover from temperature-dependent to temperature-independent nonlinear conduction (for $E < E_T$) at $T \simeq 10$ K, below which the current is transported by large portions of charge corresponding to displacement of the CDW by a distance of the order of its wavelength. The low-temperature behavior can be accounted for as a transition from thermally activated to quantum creep of the CDW.

Measurements were carried out on o-TaS_3 crystals of cross-sectional area $s \simeq 10^{-2}$ μm^2 and with a contact separation, l = 0.1–0.5 mm. Conduction was measured in the voltage-controlled regime in the two-probe configuration, which is reliable for low-temperature measurements of o-TaS_3 [2, 5]. The quality of the contacts was ascertained by measuring the resistance ratio $R_{78\,K}/R_{300\,K}$. The lengths of the crystals were chosen to be as long as possible to reduce the probable influence of contact phenomena. Cross-sectional areas were estimated from the samples' resistances at room temperature by using the room-temperature resistivity 4×10^{-4} Ω cm. The results are illustrated by the data obtained for a representative specimen of o-TaS_3 having $l = 0.32$ mm, $R_{300\,K} = 110$ kΩ, and $s = 1.2 \times 10^{-2}$ μm^2.

Figure 1 shows a sequence of I-V curves obtained for the specimen at several different temperatures. In the temperature region 70 K $\leq T \leq$ 90 K, the nonlinear conduction becomes apparent at $E > 10$ V/cm. Upon lowering the temperature below 60 K, E_T rises beyond 100 V/cm, whereas the weak nonlinearity which develops at $E < E_T$ becomes more pronounced. Qualitatively similar behavior is observed for usual-size samples [2, 6]. However, the nonlinear conduction of thin crystals is found to be much higher. For example, for a fixed field of $E = 100$ V/cm, the resistance ratios are given by $R_{4.2\,K}/R_{300\,K} = 3 \times 10^4$, 1.4×10^4, $> 1.5 \times 10^{10}$, and $> 2.5 \times 10^{10}$ for samples whose cross-sectional areas are given by $s = 8 \times 10^{-3}$, 1.2×10^{-2}, 0.2, and 3 μm^2, re-

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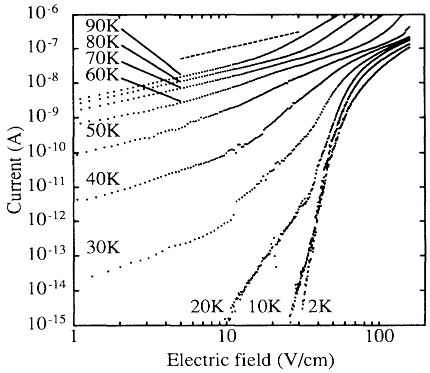

FIG. 1. Current-voltage characteristics of a thin specimen of o-TaS_3 obtained at different temperatures. Dashed line corresponds to Ohmic behavior.

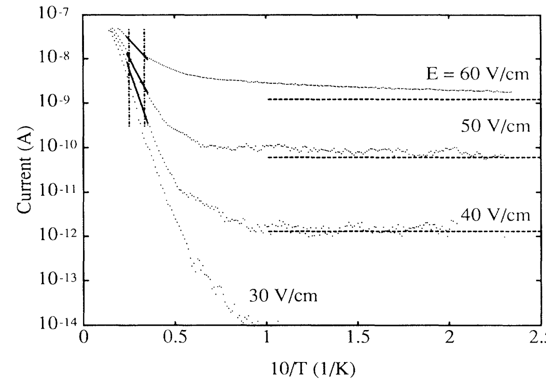

FIG. 2. Temperature dependences of the nonlinear current of the thin specimen of o-TaS_3 at different electric fields. Dashed lines show $I(2\text{ K})$. The respective slopes of activated conduction required according to Eq. (3) are shown by solid lines. Dot-dashed lines denote the temperature interval 30 K $\leq T \leq$ 40 K used for evaluation of $W(E)$ (see text).

spectively. The increasing nonlinear conductivity with decreasing sample thickness proves the existence of appreciable finite-size effects in the low-temperature nonlinear conduction of o-TaS_3. Thus, thin samples, with small cross-section areas, enable one to study nonlinear conduction at low temperatures in significantly greater detail than thick ones.

When reducing the temperature down to 20 K, the Ohmic contribution falls below the resolution of the measurements, $\sim 3 \times 10^{-15}$ A, whereas the nonlinear current continues to be detectable at $E > 10$ V/cm. Further reducing the temperature stops the temperature variation of the current at the largest fields first, and subsequently at smaller and smaller fields. The form of the limiting I-V curve is not as vertical as that observed for Fröhlich superconductivity [4, 5] and exhibits no hysteresis, in contrast to the results of more recent measurements of $K_{0.3}MoO_3$ [7].

Figure 2 shows the temperature-dependent current, $I(T)$, measured at fixed E. In the temperature range 80 K $> T >$ 20 K the form of $I(T)$ can be approximated by an activation law

$$I = I_0 \exp(-W/T), \tag{1}$$

where the "barrier height" W is a function of the field E. Thus, there is no scaling between the temperature dependences of linear and nonlinear conduction in the low-temperature region. Moreover, for $T < 10$ K the activated behavior vanishes, and the current approaches its limiting nonzero value. Such a temperature-independent nonlinear conduction has not been reported previously for TaS_3 and other CDW conductors.

In general, the relative contribution of the contacts to the measured voltage, in the two-probe configuration, may be uncertain. In the present study, both the spatial nonuniformity of the electric field in the contact region and other contact phenomena are not expected to play a significant role for the following reasons. First, no correlation was observed between the form of $I(E)$, where $E = V/l$, and the contact separation ($l = 0.1 - 0.5$ mm). Moreover, the crossover in the temperature-independent conduction is more pronounced in thinner samples of comparable lengths, where the spatial nonuniformity of the electric field plays a smaller role. In addition, a Schottky-type barrier which can arise at the metal-TaS_3 interface gives only a minor voltage correction, $\delta V < 2\Delta/e$, where 2Δ is the low-temperature Peierls gap ($2\Delta = 0.14$ eV [8]) and e is the electron charge, in the limiting I-V curve, $\delta V \ll V = 1 - 5$ V. Thus the limiting I-V curve appears mostly as a bulk phenomenon.

The phenomenon reported here resembles the recent finding of Mihály and co-workers [9] who observed the crossover in a temperature-independent nonlinear conduction in the spin-density waves (SDW) material $(TMTSF)_2PF_6$. They have found that below 1 K the I-V curve follows a Zener-type expression

$$I = I_0 \exp(-E_0/E). \tag{2}$$

The phenomenon was attributed to a novel type of collective transport, presumably due to macroscopic quantum tunneling in the SDW.

The hypothesis of macroscopic quantum tunneling in density-wave systems was introduced initially for explanation of nonlinear conduction in CDW systems at high temperatures, and was widely explored as the alternative to the classical models (see, e.g., [1] and references therein). The inset in Fig. 3 shows the current I as a function of $1/E$. One can see that a part of the I-V curve ($I < 10^{-10}$ A) can really be fitted by Eq. (2) predicted by the tunneling models [10]. However, the value of E_0 was found to vary within a factor 10 even between thin samples. Such a large variation can be hardly accounted by the tunneling models [10].

There is, however, an alternative description of the lim-

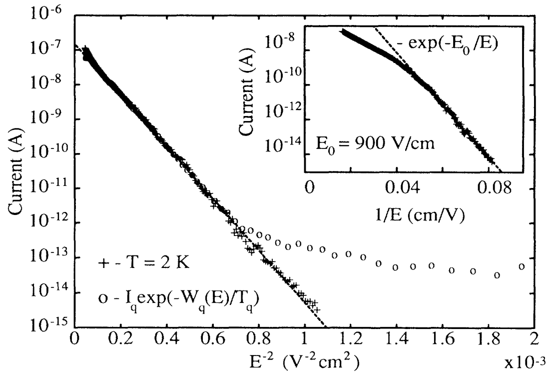

FIG. 3. The current as a function of $1/E^2$. See the text for details. The dashed line shows $I = I_q \exp(-W_q/T_q)$, where $I_q = 1.5 \times 10^{-7}$ A, $W_q = (2.6 \times 10^5 \text{ K V}^2 \text{ cm}^{-2})/E^2$, $T_q = 15$ K. Inset shows the current vs $1/E$ at $T = 2$ K. The dashed line corresponds to $I = I_0 \exp(-E_0/E)$, where $E_0 = 900$ V/cm and $I_0 = 0.015$ A.

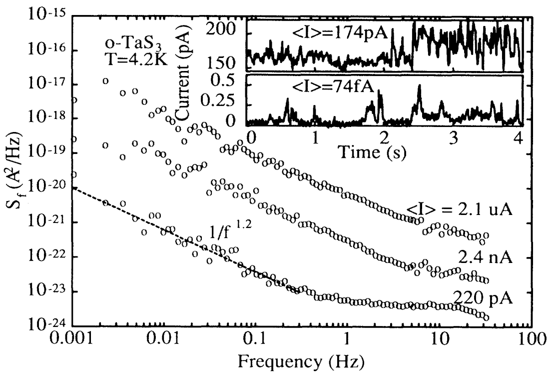

FIG. 4. Spectral power density of the current noise at different mean values of the current, I. The spectra were obtained by the Fourier transforming of time domains of the current. Insets show fragments of time domains taken at different mean values of the current. The observed duration of spikes is limited by the time constant of the electrometric amplifier used (12 ms).

iting I-V curve. First, the low-temperature activated conduction is considered to be due to thermally assisted creep of the CDW. Creep of the CDW near the transition temperature was observed recently in thin crystals of $NbSe_3$ [11]. The observed finite-size effect corresponds qualitatively to the expected size variation of the pinning energy per phase-correlation volume. In the creep regime, the kinetics of the CDW is governed by the time required to overcome pinning barriers, rather than by the energy dissipation, as in the case of viscous motion. Thus, the absence of a scaling relation between the temperature dependences of the linear and nonlinear conduction is a signature of creep of the CDW.

The activation energy of creep, $W(E)$, can be estimated from Eq. (1) with I-V curves measured at slightly different temperatures. It has been found that, at $T > 20$ K and $E > 1$ V/cm, $W(E)$ can be fitted by a power law, $W(E) \propto E^{-\alpha}$ [12], where α depends on temperature, e.g., $\alpha = 1.9$ at $T = 45$ K and $\alpha = 2.3$ at $T = 35$ K. The weak temperature dependence of α indicates a smooth temperature modification of $W(E)$. The equivalent power law for the pinning energy is known also for flux creep in superconductors, $W \propto I^{-\alpha}$, with a temperature-dependent α $(0.5 \leq \alpha \leq 2)$ [13].

Furthermore, in a majority of quasi-one-dimensional conductors the lattice zero-point motion is comparable to the lattice distortion corresponding to the CDW [14]. One can expect, therefore, that quantum fluctuations may also contribute to the CDW transport even at relatively high temperatures. If one introduces the quantum fluctuation scale, T_q, then at $T < T_q$ one can expect the I-V curve to obey an equation of the form

$$I(E) = I_q \exp[-W_q(E)/T_q], \tag{3}$$

where $W_q(E) \propto W(E)^\beta$, β depending on the details of a pinning potential. The crosses in Fig. 3 show the I-V curve obtained at $T = 2$ K and plotted vs $1/E^2$. The circles show $I(E)$ calculated according to Eq. (3) by using $W_q(E) = T_q[W(E)/T_q]^\beta$, where $W(E)$ is calculated from Eq. (1) using the I-V curves measured at 40 K and 30 K, and choosing $\beta = 0.85$, and $T_q = 15$ K, $I_q = 1.5 \times 10^{-7}$ A as fitting parameters [15]. One can see that both curves practically coincide for 5 orders of magnitude of current variation. Some discrepancy at $I < 10^{-13}$ A is due to the underestimate of $W(E)$ caused by the switching at $E \simeq 10$ V/cm at $T = 30$ K (see Fig. 1), and by the Ohmic contribution as well. Figure 2 shows the regions of thermally activated conduction (solid lines) related with the low-temperature conduction (dashed lines) according to Eq. (3): the agreement with the experimental results is also good. Thus, the limiting I-V curve can be expressed phenomenologically in terms of quantum fluctuation-assisted motion of the CDW through pinning barriers. This is one of the main conclusions of the present study. Quantum creep has also been reported recently for vortex motion in superconductors (see [16] and references therein).

The hypothesis of quantum creep agrees with the results of current fluctuation measurements. The inset in Fig. 4 shows typical plots of current vs time. One can see that the current exhibits very strong fluctuations and is provided by spikes, as in large crystals [4, 17]. The spectral power density, S_f, of the shot noise allows one to estimate the mean charge, Q, related to a spike, $S_f = 2IQ$. The phenomenon is actually complicated by a steplike irregular variation of the mean frequency of the appearance of spikes (see inset in Fig. 4) [17]. Consequently, the spectrum of fluctuations may contain Lorentzians, in addition to the shot-noise component. Figure 4 gives a sequence of the noise power spectra obtained at different mean currents. At relatively low frequencies the $1/f^\alpha$ contribution $(\alpha > 1)$ dominates in the power spectrum at any current explored. However, at sufficiently high fre-

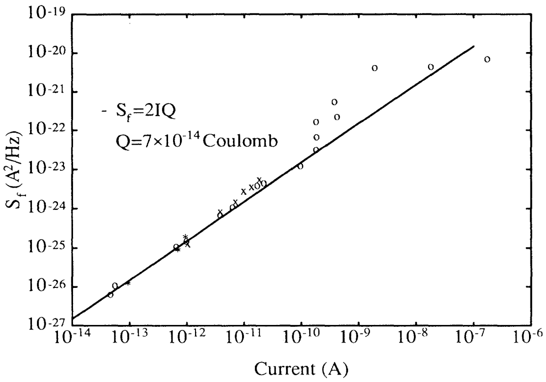

FIG. 5. Spectral power density at the frequency 1 Hz as a function of the mean current. Reproducibility of results is demonstrated by the data (marked by $*$ and $\times$) measured one week later than the main set (marked by $\circ$). $T = 4.2$ K.

quencies this contribution vanishes, and there is a range of the current ($I < 2 \times 10^{-10}$ A for $f = 1$ Hz) where the noise power is determined by the shot noise (the plateau in the spectrum corresponding to $I = 220$ pA). Figure 5 shows the spectral power density, S_f, at $f = 1$ Hz as a function of the mean current I. The most essential feature is the unit slope of $S_f(I)$ in the shot-noise region: at $I < 2 \times 10^{-10}$ A, $S_f \propto I$. The solid line in Fig. 5 is $S_f = 2IQ$ with $Q = 4 \times 10^5 e$. $Q \gg e$ indicates the collective origin of the phenomenon. The value of Q is consistent with the mean charge accumulated by a spike on the time domain (Fig. 4), and is close to the number of electrons, N, per one period of the CDW across the sample's cross section s, $Q/e \sim N = n_e \lambda s = 1.6 \times 10^5$ ($n_e = 10^{22}$ cm^{-3} is the concentration of electrons [18], $\lambda = 13.6$ Å is the wavelength of the CDW). Thus, spikes correspond to the shift of the CDW by a distance of the order of one wavelength. This type of step-by-step motion of the CDW corresponds to creep and, as was argued above, at $T < 10$ K may be due to quantum fluctuations.

A mechanism of CDW creep depends on a type of pinning. Pinning of the CDW in o-TaS$_3$ at $T \geq 100$ K was found [19] to be consistent with the modified strong-pinning model [20] when the strong-pinning contribution is ineffective due to thermal fluctuations. Lowering the temperature, however, should restore the strong-pinning contribution. If so, the creep rate is controlled by phase slip at strong pinning centers (impurity sites or, probably, structural defects). Quantum tunneling of small fragments of the CDW through respective barriers (or, alternatively, quantum fluctuations of those barriers themselves) looks more probable than tunneling of large fragments of the CDW in the tunneling model [10].

In conclusion, a crossover from temperature-dependent to temperature-independent collective CDW transport has been observed for the first time in CDW conductors. It is shown that this phenomenon can be described phenomenologically as a transition from thermally activated to quantum creep of the CDW.

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