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Low-temperature charge-density-wave dynamics and switching in $NbSe_3$

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We have studied charge-density-wave (CDW) transport in $NbSe_3$ at temperatures below 45 K. Between the threshold electric field E_T and a second characteristic field E_T^*, the CDW conductivity varies as $\sigma_{\rm CDW} \propto \exp(-\Delta/k_B T)$, with an activation energy Δ comparable to the CDW gap. Above E_T^*, the CDW conductivity increases, in many crystals via an abrupt "switch," to a much larger, only weakly temperature-dependent value. Contrary to previous suggestions, we find that E_T^* is determined by bulk CDW pinning and dynamics, not by isolated defects. We show that this behavior has strong analogs in the semiconducting CDW materials, and compare it with predictions based upon a model of CDW–normal-carrier interactions proposed by Littlewood.

For applied electric fields greater than a threshold field E_T, charge-density waves (CDW's) can depin from impurities and transport charge.[1] In $NbSe_3$, two independent CDW's form at temperatures of $T_{P_1}=145$ K and $T_{P_2}=59$ K. For the T_{P_1} CDW at temperatures between T_{P_1} and T_{P_2} and for the T_{P_2} CDW at temperatures down to ~ 40 K, the onset of CDW conduction at E_T is smooth, and the CDW current increases continuously as the field is increased. Below $T \approx 40$ K, the I-V characteristic changes qualitatively. Many crystals exhibit "switching,"[2] where the CDW current jumps discontinuously from zero to some finite value at a threshold E_T^s.

Previous studies[2–6] of switching in $NbSe_3$ have indicated that (1) E_T^s is approximately independent of temperature;[3] (2) switching at E_T^s is often hysteric and is often accompanied by large-amplitude low-frequency noise;[2–4] (3) at fields above E_T^s, the differential resistance dV/dI is approximately constant;[3] (4) switching is observed in only a fraction of $NbSe_3$ crystals, and this fraction can be increased by iron doping[4] or quenching;[5] and (5) in some crystals, regions having different switching thresholds E_T^s are arranged serially, with sharply defined boundaries between these regions.[3,6]

Motivated by observation (5) above, Hall and coworkers[3,6] proposed that switching in $NbSe_3$ is due to localized, ultrastrong pinning centers which necessitate phase slip at the pinning site; detailed models based upon this idea have been given by Inui *et al.* and Marcus, Strogatz, and Westervelt.[7] Other models invoking phase slip and/or serial coupling of CDW domains have also been proposed.[8]

Here we present an alternative view of low-temperature transport in $NbSe_3$. We show that switching is an intrinsic effect and thus that isolated defects are not its primary cause. More importantly, we find that the general characteristics of low-temperature CDW conduction in both switching and nonswitching $NbSe_3$ crystals are highly analogous to those observed in semiconducting CDW materials such as $K_{0.3}MoO_3$. We show that these characteristics are qualitatively consistent with predictions based upon a model of CDW–normal-carrier interactions proposed by Littlewood,[9] and we discuss the applicability of this model to $NbSe_3$.

We begin by describing experiments to investigate the origin of switching. Figure 1 shows four-probe data for the normal depinning threshold E_T at $T=50$ K and the switching threshold E_T^s at $T=25$ K versus inverse crystal thickness, for $NbSe_3$ crystals from a single high-purity ($r_R > 300$) growth. Both thresholds vary approximately linearly with inverse crystal thickness for thicknesses up to ~ 10 μm, and the average ratio $E_T^s(25\text{ K})/E_T(50\text{ K})$ is approximately 16.

The normal depinning threshold E_T is determined by CDW pinning by impurities distributed throughout the crystal volume. The increase in E_T with decreasing thickness results because the bulk [three-dimensional (3D)] CDW correlation length in the thickness direction is comparable to or larger than the thickness of most un-

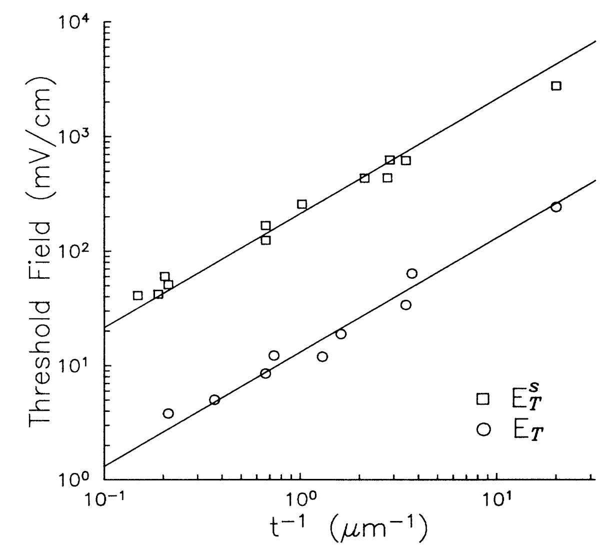

FIG. 1. Normal depinning electric field E_T at $T=50$ K and switching field E_T^s at $T=25$ K vs inverse crystal thickness for undoped $NbSe_3$. Both fields vary inversely with thickness (solid lines).

doped $NbSe_3$ crystals;[10] CDW pinning thus has a 2D character,[11] for which $E_T \propto t^{-1}$ is predicted. Since E_T^s and E_T show a similar thickness dependence and a similar scatter from the average behavior, Fig. 1 indicates that E_T^s is also determined by CDW pinning and dynamics throughout the crystal volume.

Figure 2 shows the electric field versus total current at $T=23$ K for three different segments of the same crystal. The measurements were performed with the current leads interior to the voltage leads to ensure that each segment was depinned independently. All segments show a single switch at nearly identical currents and electric fields E_T^s. Thickness-related E_T^s variations indicated in Fig. 1 make the odds of randomly selecting two crystals with comparably close E_T^s values much less than 1 in 1000. Similar measurements have been performed on 47 segments from 13 crystals. Of these, 36 segments showed a single switch. For 21 of the 33 pairs of adjacent segments, E_T^s values agreed to within 2.5%, and only two pairs had E_T^s values differing by more than 25%. These results, together with those of Fig. 1, provide strong evidence that switching is an intrinsic effect, and that the magnitude of E_T^s is determined within the crystal volume.

The results of Figs. 1 and 2 are inconsistent with models[3,6,7] in which switching arises from phase slip at isolated strong-pinning defects. Such models predict large crystal-to-crystal and segment-to-segment variations in E_T^s, and cannot explain the simple scaling of E_T and E_T^s with crystal thickness. Moreover, such models are implausible quantitatively. Measurements by Gill, Monceau *et al.*, and Maher *et al.*[12] and the theory of Ramakrishna *et al.*[13] indicate that the voltage required to produce phase slip at electrical contacts is independent of contact separation, impurity concentration, and (to first approximation) crystal thickness, but is strongly temperature dependent. For millimeter-long samples, the measured phase-slip voltages correspond to electric fields one to four orders of magnitude smaller than typical E_T^s values. Phase slip at isolated defects should, if anything, require even smaller voltages. While it is clear from previous work[3,5,6] that some $NbSe_3$ crystals are segmented into regions with different E_T^s values and different CDW current densities,[14] the phase slip which must occur between these regions cannot be the underlying cause of switching, and cannot determine the characteristic magnitude of E_T^s.

Switching is not observed in many $NbSe_3$ crystals. Therefore, more fundamental to understanding low-temperature CDW transport are those features which are observed in all crystals. Figure 3 shows typical differential resistance versus electric field data at temperatures between 41 and 23 K, for a single undoped $NbSe_3$ crystal. For $T<40$ K, CDW conduction above E_T separates into two regions: (1) Between E_T and a second characteristic field E_T^*, the CDW conductivity is strongly temperature dependent and vanishes at low temperatures; and (2) above E_T^*, the CDW conductivity is large and only weakly temperature dependent. E_T^* is nearly independent of temperature, while E_T increases gradually with decreasing temperature. At low temperatures where the CDW conductivity is very small, E_T is indicated by a change in the normal-carrier resistivity due to CDW polarizations. This general behavior is observed in *all* undoped $NbSe_3$ crystals, including those which do not switch. In crystals which do switch, E_T^* evolves into E_T^s at low temperatures.

Figure 4 shows the CDW conductivity at $E \approx 0.95 E_T^*$ vs $1/T$ for three $NbSe_3$ crystals. As indicated by the solid lines, the CDW conductivity is activated, i.e., $\sigma_{CDW} \sim \exp(-\Delta/k_B T)$, with an activation energy

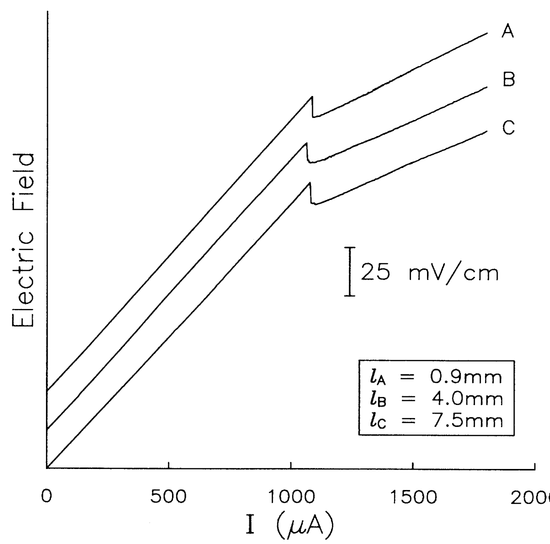

FIG. 2. Electric field vs total current at $T=23$ K for three different segments of the same undoped $NbSe_3$ crystal. Switching is observed at nearly identical fields and currents in all three segments.

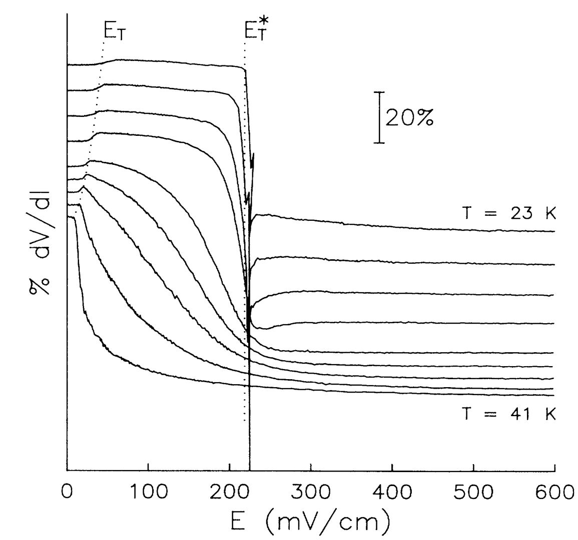

FIG. 3. Differential resistance dV/dI vs electric field for a single undoped $NbSe_3$ crystal at temperatures of 41, 38, 35, 33, 31, 28, 26, 25, and 23 K. CDW conduction between E_T and E_T^* decreases rapidly with decreasing temperature, whereas conduction above E_T^* is nearly independent of temperature.

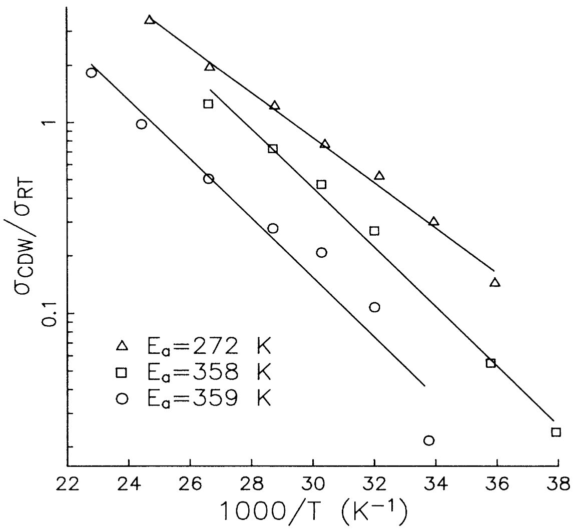

FIG. 4. CDW conductivity (normalized by the room-temperature conductivity) measured at $E \approx 0.95E_T^*$ for three different undoped $NbSe_3$ crystals. The solid lines represent fits of the form $\sigma_{CDW} \propto \exp(-E_a/k_B T)$.

$\Delta \approx 330$ K. This energy is comparable to half the T_{P_2} CDW gap energy determined from tunneling measurements.[15].

The behavior shown in Figs. 3 and 4 is strikingly similar to that observed in the semiconducting CDW materials $K_{0.3}MoO_3$, TaS_3, and $(TaSe_4)_2I$.[16] In these materials, the CDW conductivity σ_{CDW} above E_T scales with the normal conductivity σ_n of the thermally excited quasiparticles. Both are activated, $\sigma_{CDW} \propto \sigma_n \sim \exp(-\Delta/k_B T)$, with an activation energy Δ comparable to half the CDW gap. At temperatures below $\sim T_P/3$, the CDW conductivity is observed to increase dramatically above a characteristic field E_T^* roughly two orders of magnitude larger than E_T. In many crystals at sufficiently low temperatures, the transition from the low- to high-conductivity state occurs via switching.

Littlewood[9] has investigated low-temperature CDW conduction in the semiconducting materials by including the effects of CDW–normal-carrier interactions within the Fukuyama-Lee-Rice model. In this model, charge fluctuations associated with CDW deformations drive normal-carrier screening currents, and dissipation associated with these currents leads to a velocity (frequency) -dependent damping coefficient for the CDW. For CDW velocities corresponding to drift frequencies $\omega_d < \omega_d^* \propto \sigma_n$, the damping is inversely proportional to the normal-carrier conductance σ_n, resulting in a scaling between the CDW and normal conductivities. For drift frequencies $\omega_d > \omega_d^*$, normal-carrier screening is no longer effective, the damping decreases to a value determined by other processes, and the CDW conductivity increases. At sufficiently low temperatures, the decrease in damping at ω_d^* is dramatic and leads to bistability and switching in the I-V characteristic. Thus, this model predicts two characteristic fields E_T and $E_T^* \propto \omega_d^*/\sigma_{CDW}$; a scaling $\sigma_{CDW} \propto \sigma_n$ for $E_T < E < E_T^*$; a much larger, weakly temperature-dependent CDW conductance above E_T^*; switching at E_T^* at low temperatures; and since ω_d^* and $\sigma_{CDW}(E < E_T^*)$ are both proportional to σ_n, a weakly temperature dependent E_T^*. These predictions are all qualitatively consistent with observations in the semiconducting materials.[17]

These qualitative predictions are also consistent with most observations in $NbSe_3$, with one serious exception: the normal-carrier conductance σ_n in $NbSe_3$ increases with decreasing temperature in the temperature range where the CDW conductance below E_T^* is rapidly decreasing. Unlike the semiconducting materials, a small part ($\sim 10^{-4}$) of $NbSe_3$'s Fermi surface remains ungapped at low temperatures.[18] σ_n increases at low temperatures, in spite of a dramatic decrease in the normal-carrier density, because of an equally dramatic increase in the mobility of the remaining carriers. Littlewood's model of low-temperature transport might still apply to $NbSe_3$ if thermally excited quasiparticles on gapped portions of the Fermi surface were primarily responsible for CDW damping. Why this should be the case is unclear, but the activated form of the CDW conductance below E_T^* and the magnitude of the activation energy are consistent with this idea.

Littlewood's approximate analytic treatment also makes a quantitative prediction: that the crossover in CDW damping should occur at a CDW drift frequency equal to the dielectric relaxation frequency, i.e., at $\omega_d^* = \omega_r \equiv \sigma_n/\epsilon$. This prediction is in dramatic disagreement with experiment. From our data for $NbSe_3$, the measured σ_n values together with the assumption $\epsilon = 10\epsilon_0$ yield $\omega_d^*/(2\pi)$ values of 2.3×10^{15} and 3.3×10^{15} Hz at $T = 35$ and 28 K, respectively. If the Ohmic conductance due to thermally excited quasiparticles σ_n^{qp} is assumed to determine ω_d^*, then estimates obtained by scaling $\sigma_n(T_{P_2})$ with $\sigma_n(T)$ for the semiconducting CDW materials yield drift frequencies of 6.8×10^{13} and 2.3×10^{13} Hz, respectively. In contrast, the $\omega_d/(2\pi)$ values measured at $E \approx 0.95E_T^*$ are 2.9×10^7 and 3.5×10^6 Hz, respectively, more than six orders of magnitude smaller. Quantitative agreement is only slightly better for $K_{0.3}MoO_3$. Using the data of Mihaly *et al.* and Itkis *et al.*,[16] the predicted drift frequencies near $E = E_T^*$ are $\omega_d^*/2\pi \approx 1.3 \times 10^{10}$ Hz and 2.3×10^8 Hz at $T = 40$ and 30 K, respectively. The measured values are only 1.3×10^6 Hz and 1.4×10^5 Hz, nearly four orders of magnitude smaller.

Using Littlewood's model, Baier and Wonneberger[19] have calculated that the effective phason damping is proportional to the quasiparticle resistance at low frequencies, and that it crosses over to a much smaller value not at ω_r but a much lower frequency $\omega_{peak} \propto \sigma_n$, corresponding to the frequency of the peak in the imaginary part of the CDW dielectric constant.[20] Wonneberger has thus suggested[21] that $\omega_d^* = \omega_{peak}$ may be the relevant criterion for determining E_T^*. Although ω_{peak} is difficult to measure in $NbSe_3$, from the $K_{0.3}MoO_3$ data of Mihaly, Kim, and Gruner and Cava *et al.*,[20] $\omega_{peak}/2\pi \approx 10^2$ Hz at $T = 40$ K, eight orders of magnitude smaller than ω_r and four orders of magnitude smaller than the measured ω_d^*

value.

Do these quantitative discrepancies rule out the CDW–normal-carrier interaction as the origin of switching and related low-temperature effects in $NbSe_3$ and $K_{0.3}MoO_3$? First, we note that Littlewood's model has also successfully accounted for the qualitative features of the temperature- and frequency-dependent CDW dielectric constant in the semiconducting materials.[9] However, Littlewood's prediction for ω_{peak} is four orders of magnitude larger than the measured value in $K_{0.3}MoO_3$;[20] calculations by Baier and Wonneberger[19] yield better agreement. Second, the qualitative success of Littlewood's model in accounting for dc properties follows immediately from the assumptions (1) that the CDW damping is proportional to σ_n at low frequencies; and (2) that the damping decreases above a frequency $\omega_d^* \propto \sigma_n$. Any model with a CDW damping of this form should yield the same qualitative agreement. In the absence of plausible alternative mechanisms for such a damping, we believe that the qualitative successes of Littlewood's model justify further efforts toward obtaining quantitative agreement.

Finally, we note that behavior very similar to that discussed here has been observed in the sliding spin-density-wave (SDW) compound $(TMTSF)_2PF_6$ by Mihaly, Kim, and Gruner.[22] In particular, the SDW conductance roughly scales with the normal conductance at low fields, and exhibits a large increase at high fields. Mihaly *et al.* proposed that the latter increase might arise from SDW tunneling. We suggest that tunneling is not involved, and that the corresponding SDW and CDW phenomena have a common explanation.

In conclusion, we have shown that switching in $NbSe_3$ is a bulk phenomenon and is intrinsic to CDW dynamics. We have emphasized the strong analogy between the general features of low-temperature CDW transport in $NbSe_3$ and those observed in the semiconducting CDW materials. These features are qualitatively consistent with treatments of the Fukuyama-Lee-Rice model which include CDW–normal-carrier interactions, but a quantitative theory of low-temperature dc transport is still needed.

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