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Thermal fluctuations and charge-density-wave depinning in $NbSe_3$: Evidence for phase creep

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We have studied the temperature dependence of the threshold electric field E_T for charge-density-wave (CDW) depinning in $NbSe_3$. At temperatures well below the Peierls transition T_P, the fractional increase in E_T with decreasing temperature is independent of crystal thickness. In very thin crystals near T_P, the sharp threshold vanishes and nonlinear conduction occurs at arbitrarily small fields. A weak-pinning analysis suggests (1) that E_T's low-temperature increase may arise from a temperature dependence of the effective condensate density, and (2) that E_T's vanishing in thin crystals is due to thermally assisted depinning of the CDW.

Above a threshold electric field E_T charge-density waves (CDW's) are observed to depin from impurities, resulting in a variety of unusual transport properties.[1] Many of these properties show strong variations with temperature. In $NbSe_3$ and several related materials, E_T decreases gradually with increasing temperature up to $T_{\min} \approx 0.85\ T_P$, and then increases rapidly as $T \rightarrow T_P$.[1,2] The divergence near the Peierls transition at T_P is accepted to arise from differences in how the CDW elasticity, impurity coupling, and electric-field coupling vary with the CDW order parameter $\Delta(T)$.[3,4] Less certain has been the origin of the E_T increase below $T_{\min}$, where $\Delta(T) \approx \Delta(0)$. Suggested possibilities include a temperature variation of the CDW electric-field coupling[3] and thermal fluctuations of the CDW phase.[4,5] Since most calculations of CDW transport properties have neglected thermal fluctuations, establishing their role in the depinning process is of considerable importance.

Recently, we have shown[6] that charge-density waves in undoped and Ta-doped $NbSe_3$ are weakly pinned by impurities, and that the dimensionality of the pinning is an important experimental parameter. In particular, we found that E_T varies with Ta concentration n_i and crystal thickness t as $E_T \propto n_i^2$ in thick crystals and as $E_T \propto n_i/t$ in thin crystals, consistent with weak pinning in three and two dimensions, respectively. The dimensionality crossover occurs when the transverse CDW phase-phase correlation length $l_\perp$ is comparable to the crystal thickness, i.e., $t_c \approx l_\perp$. For the CDW which forms below $T_{P_1} \approx 145$ K, E_T vs t measurements and high-resolution x-ray-scattering measurements[7] indicate

$$l_\perp(T=77\ \text{K}) \approx 0.01 r_R\ \mu\text{m}\,, \qquad (1)$$

where $r_R = R(300\ \text{K})/R(4.2\ \text{K}) \propto 1/n_i$ is a measure of crystal purity.[8] For undoped $NbSe_3$ crystals, $l_\perp$ is a few micrometers.

Here we use crystal thickness and pinning dimensionality to investigate the role of thermal phase fluctuations in CDW depinning. We show that such fluctuations are not responsible for the $T \rightarrow 0$ increase in E_T. However, in very thin crystals near T_P, we observe evidence for thermally-assisted depinning. Experiments of this type may facilitate understanding of flux creep and flow in superconductors.

$NbSe_3$ single crystals prepared by vapor transport have the form of long ribbonlike whiskers. Crystal lengths can be several centimeters, thicknesses vary from ~ 0.1 to $\sim 10\ \mu$m, and widths are typically ten times larger than the thickness. Threshold electric fields in both undoped and Ta-doped $NbSe_3$ crystals were determined from four-probe measurements of the differential resistance dV/dI. Ohmic heating was minimized through the use of a He exchange gas, and characterized by checking for V^2 corrections in dV/dI. Heating effects were negligible in all data presented here. Crystal dimensions were determined as described in Ref. 6.

Figure 1 shows E_T versus temperature for crystals with thicknesses between 0.05 and 20 μm, taken from an undoped growth having $r_R \approx 220$ and t_c $(T=77\ \text{K})$ ≈ 2 μm. Two features are of note: (1) The temperature at which E_T is a minimum, $T_{\min}$, increases with decreasing thickness; and (2) the fractional changes in $E_T(T)$ for

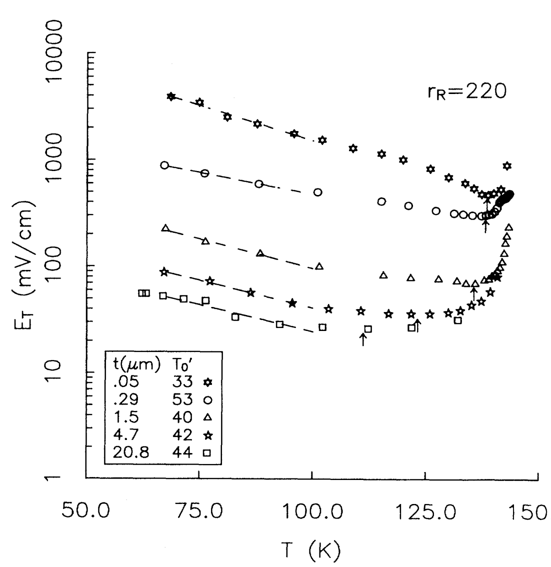

FIG. 1. Threshold electric field E_T vs temperature for the T_{P_1} CDW in undoped $NbSe_3$. For $T < 100$ K, $E_T \propto \exp(-T/T_0')$ (dashed lines) and T_0' is independent of crystal thickness. The arrows indicate where E_T is a minimum.

$T < T_{\min}$ are approximately independent of crystal thickness.

Figure 2 compares $E_T(T)$ for three crystals containing different Ta concentrations. The percent increase in E_T as $T \rightarrow 0$ is greatly suppressed with increasing doping. Previous experiments indicated both a similar suppression[9] and no suppression.[10] Some were performed on the T_{P_2} CDW, where the onset of switching[1,11] below $T \approx 35$ K complicates interpretation of the $E_T(T)$ data. Finite-size effects and the presence of impurities other than Ta may also have contributed to this discrepancy.

To discuss the origin of E_T's increase with decreasing temperature for $T < T_{\min}$, let $v\rho_1(T)$ denote the CDW-impurity coupling strength (where v is the impurity potential and ρ_1 is the CDW charge modulation amplitude), $\rho_{\text{eff}}(T)$ the effective condensate density (which determines the CDW coupling to the electric field), and let $f(T)$ denote a quantity which contains the temperature dependence of the CDW elasticity. Then in three-dimensional (3D) weak pinning the temperature dependence of E_T is given by

$$E_T^{(3\text{D})} \sim \frac{[v\rho_1(T)]^4}{f(T)^3} \frac{1}{\rho_{\text{eff}}(T)}, \tag{2}$$

while in 2D weak pinning,

$$E_T^{(2\text{D})} \sim \frac{[v\rho_1(T)]^2}{f(T)} \frac{1}{\rho_{\text{eff}}(T)}. \tag{3}$$

Maki and Virosztek[4] have associated E_T's temperature dependence with thermal fluctuations of the CDW phase ϕ. These fluctuations renormalize the CDW-impurity interaction strength $v\rho_1$ by a factor e^{-T/T_0}, where $T/T_0 = \langle \phi^2 \rangle /2$. In this interpretation, when $\Delta(T) \approx \Delta(0)$, f and ρ_{eff} are assumed to be independent of T and

$$E_T(T) \propto e^{-T/T_0'}, \tag{4}$$

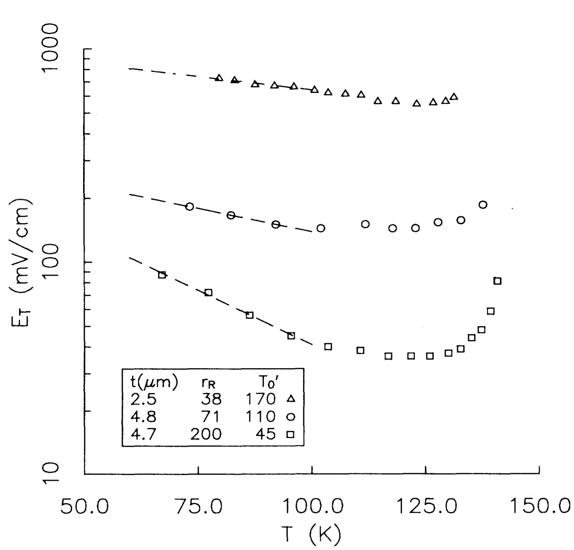

FIG. 2. Threshold electric field E_T vs temperature for crystals containing different Ta concentrations. Dashed lines indicate the fit $E_T \propto \exp(-T/T_0')$.

where $T_0' = nT_0$ and $n = 4$, 2, and 1 for 3D weak pinning, 2D weak pinning, and strong pinning, respectively. This functional form has been shown to describe $E_T(T)$ in most CDW materials, with the notable exception of $K_{0.3}MoO_3$.[12] As indicated by the dashed lines in Figs. 1 and 2, it provides a reasonable description of our $E_T(T)$ data as well. However, this interpretation is inconsistent with experiments in several ways:

(1) Experimental T_0' values for the undoped crystals in Fig. 1 do not depend on crystal thickness. Since t_c (77 K) ≈ 2 μm, the largest crystals are in the 3D weak pinning regime, while the smallest are in the 2D regime, so a factor of 2 difference in T_0' is predicted.

(2) Experimental T_0' values vary strongly with Ta impurity concentration, as indicated in Fig. 2. T_0 is predicted to be independent of impurity concentration and type.

(3) Using parameters appropriate for $NbSe_3$, calculated T_0 values are at least an order of magnitude larger than those obtained from fits to experiment, as noted by Maki.[4] The rms CDW phase displacements implied by the experimental T_0 values are on the order of π, or half a CDW wavelength. Such large displacements seem inconsistent with the observed sharp onset of CDW conduction at threshold.

The first discrepancy is the most revealing. From Eqs. (2) and (3), the temperature dependence of E_T for $T \ll T_P$ will be independent of pinning dimensionality only if the temperature dependencies of $\rho_1(T)$ and $f(T)$ are the same. Since $\rho_1(T)$ should not change with T when $\Delta(T) \approx \Delta(0)$, E_T's temperature variation is thus predicted to result from that of the effective CDW condensate density ρ_{eff}, according to $E_T(T) \propto 1/\rho_{\text{eff}}(T)$, as first suggested by Lee and Rice.[3]

The effective charge ρ_{eff}, which determines both the coupling of the CDW phase to the electric field and the current associated with a given CDW velocity, differs from the "bare" CDW condensate density ρ_c because of CDW interaction with normal carriers[3,13] (either thermally excited quasiparticles or carriers from ungapped parts of the Fermi surface). In a simple model,[3] ρ_{eff} depends upon the normal carrier density ρ_n and the CDW-normal carrier and normal carrier-lattice momentum relaxation times. Although detailed estimates of the relaxation times are difficult, they will depend both upon temperature and impurity concentration. Temperature dependence of ρ_{eff} is thus expected even when $\Delta(T) \approx \Delta(0)$ and ρ_c is independent of T.

Richard and Monceau[14] have recently deduced $\rho_{\text{eff}}(T)$ from measurements of the coherent oscillations using the relation $\rho_{\text{eff}} = J_{\text{CDW}}/ev_n\lambda$, where v_n is the oscillation frequency associated with a given CDW current density J_{CDW} and λ is the CDW wavelength. They find a strong decrease in ρ_{eff} at low temperatures, whose magnitude is comparable to but slightly smaller than the increase in E_T observed in thick crystals. Although the present analysis neglects many effects (e.g., those arising from complicated band structures) which may be important in real CDW materials, a temperature dependence of ρ_{eff} appears to provide at least a partial explanation for the origin of the temperature dependence of E_T.

Other possible explanations for $E_T(T)$ include temper-

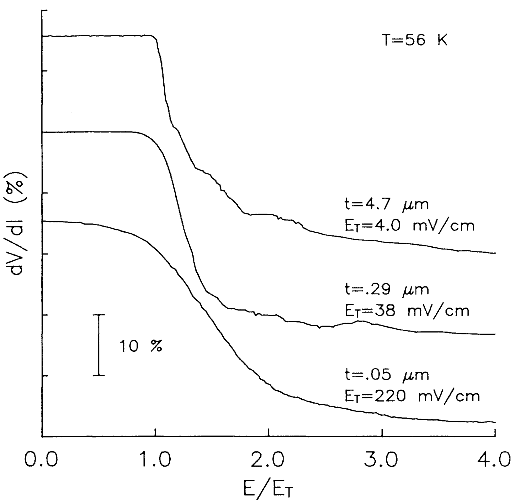

FIG. 3. Differential resistance dV/dI vs E/E_T at $T=56$ K for undoped $NbSe_3$ crystals with different thicknesses. E_T for the $t=0.05$-μm crystal is ill defined, and was estimated by scaling its dV/dI curve with those of the thicker crystals.

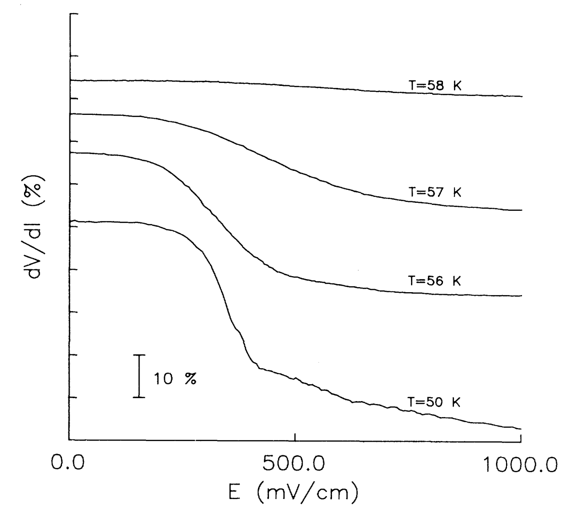

FIG. 4. Differential resistance dV/dI vs E for the $t=0.5$-μm crystal of Fig. 3 at several temperatures near T_{P_2}.

ature dependence of the phase slip which occurs at electrical contacts or of velocity shear which might occur if the CDW is more strongly pinned near the surface than in the bulk.[15] However, E_T's temperature variation for $T < T_{min}$ shows no crystal length dependence in long crystals ($L > 200$ μm) and no thickness dependence, so both of these explanations can be ruled out.

In light of recent measurements of the CDW correlation lengths in $NbSe_3$,[7] our conclusion that thermal phase fluctuations are not the cause of E_T's temperature variation is not surprising. From these lengths and measured E_T values, the pinning energy per phase-coherent CDW domain in undoped $NbSe_3$ crystals of ordinary thickness is 3–5 orders of magnitude greater than $k_B T_{min}$. $T=0$ treatments of CDW dynamics thus appear justified.

However, thermal fluctuations appear to be important in very thin crystals near T_P. Figure 3 shows the differential resistance dV/dI vs dc voltage V at $T=56$ K for three undoped $NbSe_3$ crystals with thicknesses of 4.7, 0.29, and 0.05 μm. The sharp conduction threshold observed in larger crystals is absent in the thinnest crystal, and nonlinear conduction appears to occur at arbitrarily small voltages. As shown in Fig. 4, this rounding is observed from T_P to temperatures where $\Delta(T)$ is near its $T \rightarrow 0$ value.[16] Similar behavior is observed in all crystals of comparable thickness and purity studied to date. More pronounced rounding in even thinner crystals has been independently observed by Gill and Zaitsev-Zotov.[17]

Thermal CDW depinning provides a natural explanation for the data of Figs. 3 and 4. In the 2D weak pinning limit, which should apply for thin crystals, E_T and the pinning energy per unit volume increase with decreasing thickness as $1/t$. However, the phase-coherent domain volume scales as t^2, so that the pinning energy per domain ε_{pin} is reduced from its bulk value by a factor $\Delta(T)^2(t/l_\perp)$, where $l_\perp$ is the bulk correlation length. The time-averaged CDW drift frequency due to thermal phase "hopping" may be estimated as $\omega_n \sim \omega_p e^{-\varepsilon_{pin}/k_B T} \times \sinh(\alpha E/k_B T)$, where ω_p is the pinning frequency and $\alpha \sim \varepsilon_{pin}/E_T$. For $NbSe_3$ crystals with $r_R \approx 200$ at $T=56$ K, $\varepsilon_{pin}{}^{(3D)} \approx 1\times 10^4$ $k_B T$,[18] Δ $(T=56$ K$) \approx 0.35\Delta(0)$,[1] $l_\perp \approx 5$ μm, and $\omega_p \approx 10^9$ s^{-1}, so that for $t=0.05$ μm, $\varepsilon_{pin}{}^{(2D)} \approx 12$ $k_B T$. This ε_{pin} is small enough to account for the observed CDW conduction for $E < E_T$. Near the Peierls transition ε_{pin} is even smaller. The CDW is then nearly always depinned and contributes to the $E \rightarrow 0$ conductance, reducing the rounding in dV/dI.

Rounding of dV/dI might also be expected if phase slip were to occur throughout the crystal volume, allowing independent depinning of regions with different pinning strengths. However, rounding is observed for crystal thicknesses 2 orders of magnitude larger than the amplitude coherence length ($\xi_{a^*} \approx 10$ Å) and for temperatures where $\Delta(T)$ is a substantial fraction of its $T \rightarrow 0$ value. There is no obvious reason for phase slip to be more prevalent under these conditions.

In conclusion, we have shown that thermal fluctuations of the CDW phase are not important to CDW depinning in typical $NbSe_3$ crystals, but that such fluctuations can be significant in very thin crystals near the Peierls transition. Thermal CDW depinning should be strongly analogous to flux creep and thermally assisted flux flow in superconducting vortex lattices, the subject of substantial current interest in the context of the oxide superconductors. Since the mechanism of CDW pinning is relatively well understood, and since the CDW pinning strength ε_{pin} can be continuously varied by varying the crystal thickness, $NbSe_3$ may provide a much simpler model system for study of these phenomena.

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[1]For comprehensive reviews of CDWs, see P. Monceau, in *Electronic Properties of Quasi-One-Dimensional Materials* (Reidel, Dordrecht, 1985), Part II, p. 139; G. Grüner, Rev. Mod. Phys. **60**, 1129 (1988).

[2]R. M. Fleming, Phys. Rev. B **22**, 5606 (1980).

[3]P. A. Lee and T. M. Rice, Phys. Rev. B **19**, 3970 (1979).

[4]K. Maki, Phys. Rev. B **33**, 2852 (1986); K. Maki and A. Virosztek, **42**, 655 (1990).

[5]S. Abe, J. Phys. Soc. Jpn. **56**, 1532 (1987).

[6]J. McCarten, M. Maher, T. L. Adelman, and R. E. Thorne, Phys. Rev. Lett. **63**, 2841 (1989); D. A. DiCarlo, J. McCarten, T. L. Adelman, M. Maher, and R. E. Thorne, Phys. Rev. B **42**, 7643 (1990).

[7]E. Sweetland, C-Y. Tsai, B. A. Wintner, J. D. Brock, and R. E. Thorne, Phys. Rev. Lett. **65**, 3165 (1990); also see A. H. Moudden *et al.*, Phys. Rev. Lett. **65**, 223 (1990).

[8]The r_R is also crystal-size dependent (Ref. 6). All quoted r_R values are averages for large crystals from the same growth.

[9]J. W. Brill, N. P. Ong, J. C. Eckert, J. W. Savage, S. K. Khanna, and R. B. Somoano, Phys. Rev. B **23**, 1517 (1981).

[10]K. Tsutsumi and T. Temagai, J. Phys. C **3**, 1677 (1983); A. Maeda, K. Uchinokura, and S. Tanaka, Synth. Met. **19**, 831 (1987).

[11]R. P. Hall, M. F. Hundley, and A. Zettl, Phys. Rev. B **38**, 13002 (1988).

[12]C. Schlenker, J. Dumas, C. Escribe-Filippini, and M. Boujida, Phys. Scr. **T29**, 55 (1989).

[13]T. M. Rice, P. A. Lee, and M. C. Cross, Phys. Rev. B **20**, 1345 (1979).

[14]J. Richard and P. Monceau (private communication).

[15]P. J. Yetman and J. C. Gill, Solid State Commun. **62**, 201 (1987).

[16]Rounding of dV/dI near T_P can often be observed in thick crystals, but is due to Ohmic heating and the strong decrease of the low-field ($E < E_T$) resistance with increasing temperature near T_P.

[17]J. C. Gill and S. V. Zaitsev-Zotov (private communication).

[18]$\varepsilon_{\rm pin} \approx e\rho_c E_T(\lambda/2) l_\perp l_{b^*} l_{c^*}$ with $\rho_c \approx 1.9\times10^{21}$ cm^{-3}, $E_T \approx 2$ mV/cm, $l_\perp \approx 5$ μm, $l_{b^*} \approx 50$ μm, and $l_{c^*} \approx 0.7$ μm. The correlation lengths are estimated from Ref. 7 with the assumption that their ratio for the T_{P_1} and T_{P_2} CDWs in $NbSe_3$ scale with the square root of the E_T ratio.