

Throne and McCarten Reply: Our Letter presents experimental data for the tantalum-impurity-concentration and crystal-size dependence of the threshold electric field E_T for charge-density-wave (CDW) depinning in NbSe₃. We show that finite-size-related increases in E_T and in the residual-resistance ratio (r_R) are substantial in crystals of ordinary dimensions, and must be accounted for in evaluating the effects of impurities. For crystals with large cross-sectional dimensions, $E_T \propto r_R^{-2}$, where r_R is inversely proportional to the Ta concentration n_i . For small crystals, E_T varies approximately as n_i/t , where t is the crystal thickness. These results are consistent with weak CDW pinning in three and two dimensions, respectively, suggesting that the size effects occur when the crystal thickness becomes comparable to the transverse phase-phase correlation length.

The Comments by Tucker¹ and Gill² raise three objections to our interpretation: (1) Our inferred transverse phase-phase correlation length is in obvious conflict with other experimental evidence; (2) the observed size effects can be accounted for by surface pinning; and (3) the evidence for weak pinning by Ta impurities is unconvincing.

Our inferred transverse phase-phase correlation length L_x , on the order of 1 μm , is indeed nearly 2 orders of magnitude larger than the most widely accepted value, and our domain volumes are more than 4 orders of magnitude larger. The large transverse correlation lengths, together with uncertainty as to whether CDWs in NbSe₃ are weakly or strongly pinned, have led many to reject what would otherwise be the most natural explanation for the size effects. The accepted L_x value has been obtained from the TEM experiments of Fung and Steeds,³ who observed strandlike "domains" with typical dimensions of 2 $\mu\text{m} \times 200 \text{ \AA} \times 200 \text{ \AA}$ in dark-field images of the CDW superlattice in NbSe₃. These domains have been accepted by many (although not by Steeds, Fung, and McKernan, or by Chen, Fleming, and Petroff⁴) to be the impurity-pinned phase-coherent CDW domains described by Fukuyama, Lee, and Rice.

It is extremely unlikely that the TEM domains have any relevance to CDW pinning in large Ta-doped NbSe₃ crystals. First, the TEM domains are insensitive to temperature changes, radiation damage (including Se loss), electric fields of up to 0.5 V/cm, and macroscopic crystal imperfections,⁴ so that their connection with CDW pinning is unclear. Second, the TEM domain dimensions must be limited by the small thickness (300–500 \AA) of the crystals studied. Third, the TEM domain shape anisotropy is inconsistent with the measured anisotropy of the room-temperature electrical conductivity σ . The amplitude coherence lengths and weak-pinning phase-phase correlation lengths are expected⁵ to vary as $\sigma^{1/2}$, and should have values along the z , y , and x directions (corresponding to the crystallographic b , c , and a axes) in the ratio 10:2.5:1. The TEM domain anisotropy, 100:1:1, is thus far too large. In large crystals, if the b -

axis phase-phase correlation length is several microns, as is widely accepted, then the transverse correlation lengths should be on the order of 1 μm , as the weak-pinning dimensionality-crossover interpretation of size effects suggests.

Our estimate of the phase-phase correlation length is consistent with the measured low-frequency dielectric constant, within the considerable experimental and theoretical uncertainties. L_x was estimated from the size dependence of E_T in our Fig. 3 by a fit drawn through the data. However, the 2D fit should provide a lower asymptote. Also, the effects of finite crystal widths, which are important in crystals with small cross-sectional aspect ratios, and of the nonuniform cross sections typical of NbSe₃ crystals were ignored. The L_x values obtained in this way are likely too large; they are a factor of 3 larger than values estimated from the bulk E_T . This uncertainty does not affect the essential implication of our analysis: that the phase-correlated volumes are much larger than previously thought.

Strong evidence for the weak-pinning interpretation is provided by the measured crystal-size dependence of the narrow-band noise (voltage oscillation) amplitude δV . δV is expected to be roughly proportional to $V_T/N^{-1/2}$, where N is the number of phase-coherent volumes within the crystal (although there is no reason to expect a proportionality constant of order 1, as is usually assumed in estimating N). In 3D weak pinning, V_T^{3D} and the domain volume V_d^{3D} are independent of crystal size, and both δV and $\delta V/V_T$ vary as $N^{-1/2} \propto V^{-1/2}$, where V is the crystal volume. In 2D weak pinning, $V_T \propto 1/t$, the phase-phase correlation lengths in the unconstrained directions L_y and L_z vary as $t^{1/2}$, and the domain volume V_d^{2D} varies as t^2 . In a crystal of width w , $N \propto w/t$ so that $\delta V \propto (wt)^{-1/2} \propto V^{-1/2}$, as in 3D weak pinning. However, for $w \propto t$, as is approximately true for randomly selected NbSe₃ crystals, $\delta V/V_T$ is independent of V . Mozurkewich and Grüner⁶ measured δV at $T=42$ K in undoped NbSe₃ crystals having cross-sectional areas A ranging from 1 to 1000 μm^2 , and found that $\delta V \propto A^{-1/2}$. For NbSe₃ crystals of comparable purity at comparable temperatures, we find $E_T \propto 1/t$ for $A < 1000 \mu\text{m}^2$ (corresponding to $t \leq 10 \mu\text{m}$). With the assumption $A \propto t^2$, this implies that $\delta V/V_T$ for Mozurkewich and Grüner's crystals was approximately independent of A over 3 orders of magnitude in crystal area. This size independence of the narrow-band noise amplitude $\delta V/V_T$ is consistent with the size dependence of the domain volume expected in 2D weak pinning.

A second criticism of our Letter is that we have not adequately considered alternative origins for the size dependence of E_T . We abandoned surface-pinning models for the weak-pinning dimensionality-crossover model only after several experiments provided no clear evidence for the former. In the simplest surface-pinning model, the CDW is pinned by defects at the crystal surface. We rejected this model because it does not account for the

variation of size effects with bulk impurity concentration, and because we observed no dependence of E_T on surface preparation conditions. In the surface-pinning model proposed by Gill, the CDW near the surface adjusts its wave vector towards the commensurate value and becomes strongly pinned. CDW sliding then occurs when the more weakly pinned bulk CDW shears by glide of edge dislocations from the strongly pinned surface CDW, a process which is expected to be thermally activated.⁷ While this model might plausibly reproduce some of the observed behavior, there is not direct evidence either for surface CDW commensurability⁸ or for the importance of edge dislocations in micron-thick crystals. Further, we find no significant size-related variations in the temperature dependence of the threshold field (except near the Peierls transition). And we find that the maximum width of the Shapiro steps observed on the I_{CDW} - V characteristic when ac voltages are applied, which reflects the maximum time-averaged polarization which can be developed by the CDW in the sliding state, scales with the threshold voltage, i.e., $\Delta V/V_T$ is independent of crystal thickness. Our results suggest that the essential character of CDW pinning is the same in the bulk and size-dependent regimes, making it unlikely that fundamentally different pinning mechanisms are involved.

Finally, both Comments suggest that the evidence that CDWs in Ta-doped NbSe₃ are weakly pinned is not convincing. We summarize below experimental results from our Letter and from work to be described elsewhere which are consistent with weak pinning.

(1) In the bulk or size-independent regime, $E_T \propto r_R^{-2}$ and thus to n_i^2 for both the high-temperature ($T_P = 145$ K) and low-temperature ($T_P = 59$ K) CDWs in Ta-doped NbSe₃, as shown in Fig. 1. In Ti-doped NbSe₃, the measured E_T - n_i relation is also consistent with weak pinning. In strong pinning, $E_T \propto n_i^\alpha$, where $\alpha = \frac{4}{3}$ (dashed lines in Fig. 1) or 1.

(2) For a given Ta concentration, the ratio of the minimum bulk thresholds for the high- and low-temperature CDWs $E_T^{min}/E_T^{min} \approx 9$. In strong pinning, $E_{T1}/E_{T2} \approx \Delta_1/\Delta_2 \approx 2.4$. The threshold ratio in weak pinning is expected to be much larger, but is very sensitive to parameters whose values are imprecisely known.

(3) For NbSe₃ crystals at $T = 77$ K, E_T for a given Ta concentration is roughly 40 times larger than for an equal Ta concentration. In a revised view of strong pinning,⁹ E_T shows no strong dependence on impurity type.

(4) In the size-dependent regime, $E_T \propto n_i/t$, consistent with 2D weak pinning.

(5) The ratio $[E_T^{2D}t]^2/E_T^{3D}$, where E_T^{2D} is the threshold field in the size-dependent regime for a given thickness t and E_T^{3D} is the bulk threshold, is approximately the same for Ti and Ta impurities, as expected in weak pinning.

In the absence of a plausible, quantitative alternative interpretation, we interpret these results as strong evi-

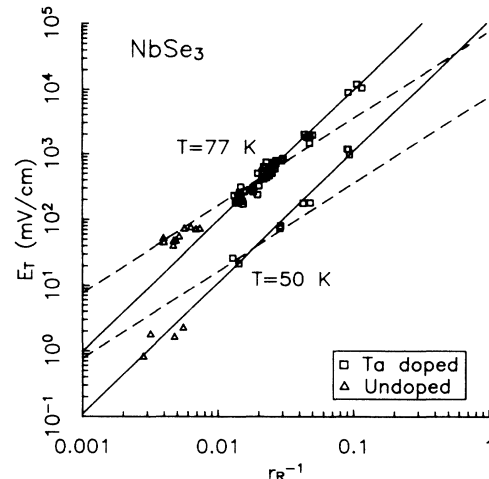


FIG. 1. Threshold electric field E_T vs inverse residual-resistance ratio for the two CDWs in NbSe₃, measured in large crystals. The solid and dashed lines represent least-squares fits of the form $E_T \propto r_R^{-2}$ and $E_T \propto r_R^{-4/3}$, respectively. The undoped crystals, which were prepared by a different method and contain almost no Ta, are excluded from the square-law fits since the defects which pin the CDWs in these crystals are expected to have different E_T - r_R relations.

dence that CDWs in Ta-doped NbSe₃ are weakly pinned.

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¹J. R. Tucker, preceding Comment, Phys. Rev. Lett. **65**, 270 (1990).

²J. C. Gill, preceding Comment, Phys. Rev. Lett. **65**, 271 (1990).

³K. K. Fung and J. W. Steeds, Phys. Rev. Lett. **45**, 1696 (1980).

⁴C. H. Chen, R. M. Fleming, and P. M. Petroff, Phys. Rev. B **27**, 4459 (1983); J. W. Steeds, K. K. Fung, and S. McKernan, J. Phys. C **3**, 1623 (1983); S. McKernan (private communication).

⁵For example, see S. Barisic, in *Electronic Properties of Quasi-One-Dimensional Materials*, edited by P. Monceau (Reidel, Dordrecht, 1985), p. 1.

⁶G. Mozurkewich and G. Grüner, Phys. Rev. Lett. **51**, 2206 (1983).

⁷J. C. Gill, J. Phys. C **19**, 6589 (1986).

⁸X.-M. Zhu, I. Robinson, R. Moret, E. Vlieg, H. Zabel, and R. M. Fleming (to be published).

⁹J. R. Tucker, W. G. Lyons, and G. Gammie, Phys. Rev. B **38**, 1148 (1988).