Contribution of spontaneous phase slippage to linear and nonlinear conduction near the Peierls transition in thin samples of TaS_3

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In the Peierls state very thin samples of TaS_3 (cross-section area $\sim 10^{-3}$ μm^2) are found to demonstrate smearing of the *I-V* curves near the threshold field. With approaching the Peierls transition temperature, T_P, the smearing evolves into smooth growth of conductance from zero voltage interpreted by us as the contribution of fluctuations to the nonlinear conductance. We identify independently the fluctuation contribution to the linear conductance near T_P. Both linear and nonlinear contributions depend on temperature with close activation energies $\sim(2-4)\times 10^3$ K and apparently reveal the same process. We reject creep of the *continuous* charge-density waves (CDW's) as the origin of this effect and show that it is spontaneous phase slippage that results in creep of the CDW. A model is proposed, accounting for both the linear and nonlinear parts of the fluctuation conduction up to T_P.

I. INTRODUCTION

Though the Peierls transition in quasi-one-dimensional conductors was discovered more than 30 years ago, its mechanism and the role of fluctuations still remain an unsettled problem.[1,2] The fluctuations in quasi-one-dimensional compounds below T_P are seen from various studies including transport properties, such as the large width of the Peierls transition[1,2] in comparison with that expected from the BCS-type onset of the gap, the smeared edge of the Peierls gap revealed through the optical investigations,[3] and the appearance of spontaneous current noise, which is associated with thermally initiated phase slip (PS) developing several kelvins below T_P.[4]

A decrease of the cross-section area s of the samples results in growth of the fluctuations. For example, in the samples of o-TaS_3 with $s\sim 10^{-2}$ μm^2 and below, the Peierls transition is smeared out and substantially shifted down to lower temperatures.[5] Conductance hysteresis in such thin samples is absent within decades of kelvins below T_P; in this temperature range spontaneous PS is observed, and the conductivity strongly deviates from the Arrhenius law.[4,6] Another fluctuation effect known as threshold rounding consists in smearing of the onset of the nonlinear current at the threshold field E_T.[7–9] This effect is found in $NbSe_3$. The rounding increases both with increasing T and decreasing thickness t of the crystals; in the thinnest crystals the growth of conductivity starts from zero field. In Refs. 7–9 phase slippage has been discussed as a possible basis of the rounding, but the authors did not find enough arguments in favor of this explanation. Another interpretation was found to be more reasonable:[7–9] the rounding was attributed to the thermally assisted creep of charge-density waves (CDW's) in the framework of the weak-pinning model.[1,2] This approach implies that in very thin samples the pinning energy of the phase-correlation volume becomes comparable with kT, and activated creep of the continuous CDW within the correlation lengths $L_{2\pi}$ is possible. Estimates for $NbSe_3$ based on the mean-field BCS dependence for $\Delta(T)$, showed that thermal depinning of the CDW is probable. This interpretation, however, is rather dubious for TaS_3, where the mean-field approach fails near T_P: in highly anisotropic compounds such as TaS_3 and $K_{0.3}MoO_3$, the onset of the gap near T_P does not follow the BCS dependence,[10,11] and the pseudogap does not vanish tens of kelvins above T_P.[3,12,13]

In the present paper the threshold rounding in thin samples of TaS_3 is reported. Independently we observe a fluctuation contribution to the linear conductivity. It is shown that creep of the *continuous* CDW cannot account for the threshold rounding in TaS_3. Alternatively, we show that spontaneous PS observed near T_P results in local creep of the CDW and contributes to the linear and nonlinear conductivity, in agreement with our experiment. The result is generalized for large samples. We discuss the mechanism of the Peierls transition in the light of the PS-induced creep.

II. EXPERIMENTAL TECHNIQUE AND RESULTS

Thin samples of TaS_3 were placed on sapphire substrates. We used vacuum-deposited indium contacts.[14] The cross-section area of the samples was estimated from the values of the room-temperature resistance (3×10^{-4} Ω cm) and the visible contact separation.[14] Similar results are observed on five samples from high-quality batches. Most of the data reported here are obtained on the representative sample with the dimensions $L=4.5$ μm, $s=0.3\times 10^{-3}$ μm^2.

The dependencies of conduction σ on temperature and voltage V are presented in Figs. 1 and 2, respectively. One can see (Fig. 1) that the Peierls transition is smeared out in comparison with the transitions in usual-sized samples (shown with a dotted line), in agreement with Ref. 5. Deviation from the Arrhenius law is observed tens of kelvins below T_P (indicated by an arrow), the latter being considerably shifted downwards[5] in comparison with $T_P=220$ K observed in thick samples. The activation dependence $\sigma\propto\exp(-\Delta/T)$ with $\Delta=800$ K extrapolated from the low temperatures is shown by the broken line; we denote the resulting conductivity as σ_Δ. We shall consider the temperature and sample-

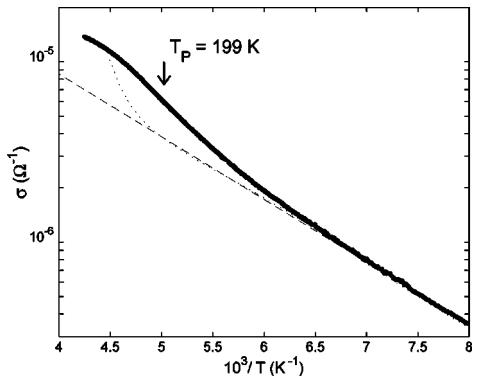

FIG. 1. Temperature dependence of the low-field resistance for the representative sample measured at $V=20$ mV ($T<178$ K) and at $V=10$ mV ($T>178$ K). The broken line, σ_Δ, shows the extrapolation of the low-temperature conductivity $\sigma_\Delta \propto \exp(\Delta/T)$, $\Delta=800$ K. The dotted line is normalized to a low-temperature $\sigma(T)$ curve for a typical thick pure sample.

size dependence of the Peierls gap, 2Δ, to be insignificant, which is supported by the results of Refs. 3, 9, and 13. Then, the difference $\delta\sigma \equiv \sigma - \sigma_\Delta$ can be considered as the fluctuation contribution to the conductivity. The Arrhenius plot $\delta\sigma$ vs $1/T$ is shown in Fig. 3 with dots. The dependence is close to a straight line up to $T \approx T_P$; the activation energy W, being about 2400 K, is well above Δ. For samples with higher cross-section areas we obtained somewhat larger activation energies, up to $W \approx (5-7)\times 10^3$ K for the normal-sized samples, as it was reported earlier.[15]

Turning to the σ vs V dependences (Fig. 2) we note that the onset of the nonlinear conduction is smeared, the threshold rounding being clearly seen above $T=120$ K. The scale of the voltages applied corresponds to large fields, above 1 kV/cm. At lower temperatures (about 120 K) the onset of the nonlinear conduction is relatively sharp, and we can estimate the threshold for collective conduction as $V_T \approx 0.2$ V ($E_T \approx 400$ V/cm), in accordance with the size effect.[5] We shall

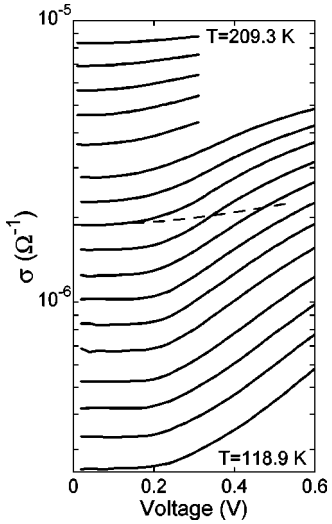

FIG. 2. Voltage dependencies of the conductivity (I/V) at $T=209.3$, 203.4, 197.4, 191.4, 184.5, 176.8, 171.2, 165.9, 160.1, 154.6, 149.3, 144.3, 139.1, 133.6, 128.8, 123.9, and 118.9 K. The broken line shows an example of the fit of $\sigma_{nl}(E)$ by Eq. (5), where $E_T=480$ V/cm, $\sigma_l=\sigma-\sigma_\Delta$ (see text).

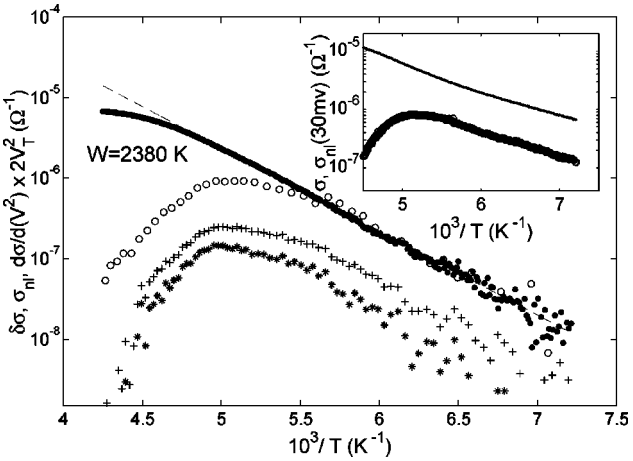

FIG. 3. Temperature dependencies of the excess linear conductivity (dots), nonlinear conductivity for two values of V below V_T: *, $V=120$ mV; +, $V=160$ mV. The open circles show the fit of the nonlinear conductivity in accordance with Eq. (5) with $V_T=220$ mV ($E_T=480$ V/cm) (see text). The inset shows the total conductivity (dots) together with nonlinear conductivity (circles) at fixed $V=300$ mV ($V>V_T$).

assume that at higher temperatures the value of E_T is approximately the same. Thus, the nonlinear conduction for $V<0.2$ V will be referred to as subthreshold nonlinear conduction, while at V well above 0.2 V collective conduction is expected. Below we shall see that this division is not unphysical.

One can see (Fig. 2) that the rounding progresses with approaching T_P. At high temperatures ($T \gtrsim 170$ K) it is impossible to define a voltage range of linear conduction: the nonlinearity starts from zero voltage. Figure 3 shows the nonlinear conductance, $\sigma_{nl} \equiv \sigma(V) - \sigma(0)$, at fixed values $V<V_T$ as a function of T, together with $\delta\sigma(T)$. One can see that $\delta\sigma(T)$ and $\sigma_{nl}(T)$ behave in similar ways up to $T \approx 175$ K, while at higher temperatures σ_{nl} deviates downwards. Evidently the excess conductivity $\delta\sigma$ and the threshold rounding have a common underlying mechanism at least at the lower temperatures. The possibility of coupling of nonlinear conduction below E_T with an enhancement of low-field conductivity was also noticed in Ref. 7 for thin samples of $NbSe_3$.

The scaling observed resembles that between the linear and nonlinear conduction *above* V_T (Ref. 16) known for different CDW conductors, including TaS_3.[17] Our samples also demonstrate such a scaling: The inset to Fig. 3 shows the dependence $\sigma(T)$ together with $\sigma_{nl}(T)$ at fixed $V>V_T$. In agreement with earlier observations, both values depend on temperature in a similar way, with the activation energy $\sim\Delta$. It is clear that the scaling between $\sigma_{nl}(T)$ ($V<V_T$) and $\delta\sigma(T)$ is quite different, as the slopes of the curves correspond to much higher activation energies, $W \gg \Delta$.

III. DISCUSSION

The large values of E_T result from the small transversal dimensions of the samples, in accordance with the size effect observed in TaS_3.[5] As both transversal dimensions of our TaS_3 samples are of the same order of magnitude,[5] we

expect the pinning to be one-dimensional, rather than two-dimensional, as in the thin samples of $NbSe_3$.[7,9,18] Following the explanations of rounding in $NbSe_3$,[7–9] we could assume that the pinning energies of phase-correlation volumes in TaS_3 nanosamples are small enough to enable thermal depinning of phase-coherent volumes.

The lowest-energy local depinning of the CDW results in a phase gain by the value $\sim 2\pi$ over the phase-correlation length.[20] Such a deformation will cause a CDW stress, resulting in a local variation of resistance by percents for our samples.[21] Meanwhile, metastable states cannot exist in such thin samples in the vicinity of T_P: the hysteresis loop develops only below 140 K in the representative sample [in usual-sized pure samples the hysteresis develops 5–7 K below T_P (Refs. 4 and 22)]. So, any deformation immediately relaxes via a PS event, i.e., a plastic deformation of the CDW. The PS event is followed, sequentially, by local creep of the CDW.[21] This results in a phase perturbation of the same order of magnitude as the initial elementary act of creep,[21] and so PS is to be taken into account. Below we discuss in detail the conditions for the spontaneous PS (Refs. 4, 6, and 23) and its effects on the conductivity.

It is remarkable that the activation energy for the fluctuations is nearly independent of the field applied while it is below E_T: the slopes of the excess linear conductivity and of the nonlinear conductivity at $V=160$ mV (which is quite close to $V_T \sim 200$ mV) are close, while the activation energy for the creep should run to zero at $E \rightarrow E_T$.[24] So the process initiating the fluctuation conduction is other than creep of the CDW. At the same time, at low temperatures ($\lesssim 130$ K) the nonlinear conductivity becomes distinguishable only close to E_T, i.e., reveals itself as the threshold rounding. So E_T is a characteristic field for the fluctuation conductivity, and the latter is in a way coupled to the CDW creep. This apparent contradiction is removed by the the following consideration.

Evidently, the mechanism initiating the conductivity is the PS: the high activation energy is typical for PS in TaS_3,[4,5] and its independence of E at $T>120$ K was reported in Ref. 25. At the same time, according to Ref. 21 each PS event is followed by temporary creep (rearrangement) of the CDW in the vicinity of the point where the PS occurred. In the presence of an external electric field the creep prevails in the direction defined by the field and provides a mechanism of the CDW conduction below E_T. At $E \rightarrow E_T$ the CDW phase-correlation length diverges,[26] so E_T is expected to be the critical point for the conduction. A hypothesis that phase slippage (in particular, edge dislocations) could facilitate CDW creep was also mentioned in Ref. 7.

In the case of one-dimentional pinning (which could be applied to our samples) and PS involving the whole cross-section area, we can estimate the current induced by the PS. For simplicity let us consider the initial state to be uniform, i.e., the shift of the chemical potential $\zeta = \text{const}$. Entering of a new period in the absence of external field is followed by CDW creep under the internal electric fields $E_{int} = d\zeta/dx$. The creep proceeds while the effect of E_{int} exceeds the effect of impurities, which we for simplicity describe by the average value, E_T. The resulting phase perturbation (Fig. 4) covers the length[21]

$$L_{2\pi} \approx 2\sqrt{\pi (d\zeta/dq)/E_T}, \tag{1}$$

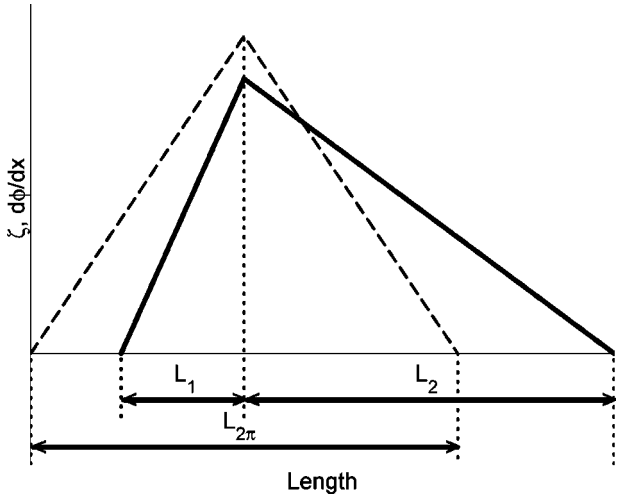

FIG. 4. Phase (Φ) redistribution following a PS event at zero (broken line) and nonzero field $E=\frac{1}{2}E_T$ (solid line) according to the critical-state model. The PS-induced creep results in an average shift of the charge density to the right.

where $d\zeta/dq$ characterizes the CDW elastic modulus, q being the in-chain component of the CDW wave vector. Note that $L_{2\pi}$ appears to be of the order of the phase-correlation length.[1,21] Under an external electric field $E<E_T$, the creep proceeds asymmetrically with respect to the point of the PS nucleation, giving the divergence of $L_{2\pi}$ at $E \rightarrow E_T$.[21] The new period is distributed so that the $d\zeta/dx = E_T + E$ from one side of the maximum remnant deformation and $-(E_T - E)$ from the other side (Fig. 4). The resulting progress of the CDW (and of the coupled charge $2e$ per chain) in the direction defined by E could be estimated as $\delta L = \frac{1}{3}(L_2 - L_1)$, where L_1 and L_2 are the lengths of the phase perturbations in the two directions (Fig. 4).[27] With the condition that the areas under the triangles (Fig. 4) should be equal and correspond with the phase gain 2π, we obtain from simple calculations

$$\delta L(E) = \frac{1}{3} L_{2\pi} \frac{E}{E_T} \frac{1}{\sqrt{1-(E/E_T)^2}}. \tag{2}$$

If the PS nucleation rate per unit length is $\nu(T,E)$, then the resulting mean current is

$$I_{PS} = 2e\,\nu\,\delta L \tag{3}$$

per chain. As each PS event (fluctuator) affects the length $\sim L_{2\pi}$, $L_{2\pi}\nu \equiv f$ may be considered as a typical frequency of switching of independent fluctuators. The temperature dependence of the PS rate could be empirically presented as $\exp(-W/T)$,[28] where $W \sim (5-7)\times 10^3$ K.[4,5,30] So, Eqs. (2) and (3) give the dependence of the excess current both on T and E. As $E \rightarrow E_T$ an unphysical divergence of I_{PS} occurs, because in the model we have neglected the time of creep, τ_{cr}, following each PS event. With approaching E_T τ_{cr} grows together with L_2 (Fig. 4), and the PS frequency becomes dominated by $1/\tau_{cr}$. At low temperatures when f is relatively small, I_{PS} becomes noticeable only for E close to E_T: Equations (2) and (3) thus feature the threshold rounding. At higher T (and f) the area of validity of Eq. (3) shrinks

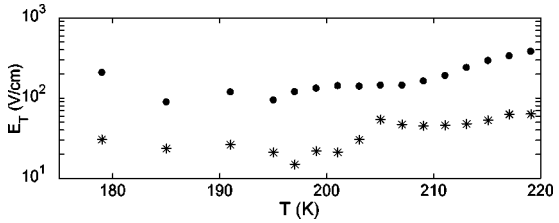

FIG. 5. The T dependence of E_T for the sample with $L=4$ μm, $s=1.5\times10^{-3}$ μm^2. *, direct measurements; dots, from the values of $d\sigma/d(E^2)$ and $\delta\sigma$ (see text).

down to smaller $|E|$. In the limit of small $|E|$, neglecting the dependence of ν on E,[25] we obtain:

$$I_{PS}=\frac{2}{3}ef\left[\frac{E}{E_T}+\frac{1}{2}\left(\frac{E}{E_T}\right)^3\right]\equiv I_l+I_{nl}. \tag{4}$$

Thus, spontaneous PS gives contributions both to linear (I_l) and nonlinear (I_{nl}) currents. Note that extrapolation of E to E_T gives a relation between I_{PS} and f that is very similar to that between the CDW current and the fundamental frequency. This is natural, because for $E\rightarrow E_T$ each pair of electrons entering the CDW via a PS event creeps along the whole sample.

From Eq. (4) we obtain

$$\sigma_{nl}=\frac{1}{2}\sigma_l\frac{E^2}{E_T^2}, \tag{5}$$

where σ_l is the PS-induced part of the linear conductivity. At fixed E/E_T ($E<E_T$) the fluctuation nonlinear conductivity should have the same temperature dependence as the linear conductivity: both are dominated by $f\propto\exp(-W/T)$ [Eq. (4)]. Neglecting the dependence of E_T on T we see that the scaling between $\delta\sigma$ and σ_{nl} is the same as that observed in Fig. 3. For a quantitative comparison of $\delta\sigma$ and σ_{nl} note that $[d\sigma/d(E^2)]2E_T^2$ is simply $\sigma_l(\equiv\delta\sigma)$ [Eq. (5)]. To check this, we show the value $[d\sigma/d(E^2)]2E_T^2$ in Fig. 3, where $d\sigma/d(E^2)$ is determined from the best fit of $\sigma(V)$ ($V\ll V_T$) with parabolic dependencies and E_T is a fitting parameter. An example of the fit of $\sigma(V)$ by Eq. (5) is shown in Fig. 2 with a broken line. For the representative sample we get $E_T=480$ V/cm. For different samples, the values obtained from the fit by Eq. (5) agree with the results of direct measurements, though they are somewhat larger.[31]

Additional evidence of the correlation between the threshold field and nonlinear fluctuation conduction is provided by the measurements of another sample with approximately the same length but larger cross-section area, $s=1.5\times10^{-3}$ μm^2, and somewhat larger activation energy for $\delta\sigma(T)$, $W=3400$ K. A similar treatment of σ_{nl} with the help of Eq. (5) gave us the dependence $E_T(T)$. In addition, we were able to measure $E_T(T)$ up to $T=T_P$ and higher[13] directly, as the onset of sharp nonlinear conduction. This was possible after subtracting the part of conductivity $\propto E^2$ from each $\sigma(V)$ curve.[13] The values of E_T determined both ways are presented in Fig. 5 as a function of temperature. Both dependences show mesoscopic-type irregular variations of E_T with temperature,[32] though E_T obtained from Eq. (5) is about 5 times larger (see Ref. 31). One can see correlation between the two dependences.

From Eq. (4) we obtain reasonable estimates of the frequencies of switching for the fluctuators. To observe distinct excess conductivity (at 140 K for the representative sample, Fig. 3), we should take $f\sim10^7$ Hz. This is only two to three orders of magnitude higher than we were able to see directly from the time domains of the fluctuations,[6,23] the latter being restricted by the electric scheme. Thus, both the values and the activation energy for the linear and nonlinear fluctuation currents are fairly described by the model proposed.

It is worth mentioning that the dependence of $|d^3V/dI^3|$ vs T below T_P presented in Ref. 7 fits the Arrhenius law fairly well with $W\approx4500$ K, in agreement with the PS measurements at the contacts.[30]

The lowering of W with the decrease of the sample's cross-section area also finds natural explanation within the model proposed. In fact, a large threshold field corresponds to high inhomogeneous stress of the CDW in the thin samples:[21]

$$\langle\zeta^2\rangle^{1/2}\sim2\sqrt{\pi E_T(d\zeta/dq)}. \tag{6}$$

The stress lowers the barrier for the PS in certain points.[23] The decrease of the sample cross-section area reveals itself in the growth of E_T, and thus results in the lowering of the activation energies for $\delta\sigma(T)$ and $\sigma_{nl}(T)$. Earlier we have explained in a similar way the broadening of the range of the fluctuations and of the Peierls transition along the temperature scale in the thin samples.[23] Note that the model of thermal depinning of the phase-coherent volumes[7,9] gives a much stronger size dependence of the excess conductivity: the depinning energy is proportional to $s^{2/3}$.[9] So, for the sample with $s=1.5\times10^{-3}$ μm^2 we should expect W to be about 7000 K (instead of 3400 K), and the excess conductivity should become negligible in the thick samples. Actually, we found no qualitative difference between the excess conductivity of the thick and thin samples. The activation energy for $\delta\sigma(T)$ in thick samples is $(5-7)\times10^3$ K,[4,15] in agreement with the activation energy found from the noise probing of the spontaneous PS.[4] The threshold rounding is also noticeable in thick samples,[2,13] though the study of the nonlinear fluctuation conductivity is complicated because of its narrow temperature range and small E_T.

Note that the dependence $\delta\sigma(T)$ follows the activation law up to T_P, and even a little bit above it (Fig. 3), no feature being observed at T_P. So, the state a little bit above T_P could be considered as a CDW saturated with climbing dislocations rather than a single-electron state. The conduction of such a mixture is supplied by the processes of nucleation and motion of the domain boundaries, which exert high internal electric fields to the domains. The fact that the dependencies characterizing the nonlinear conductivity deviate from the Arrhenius law at lower temperatures than $\delta\sigma(T)$ could be ascribed to the growth of E_T near T_P (Fig. 5); note also that with increasing T the model fails first at finite E, and then at $E\rightarrow0$.

In conclusion, we have observed the fluctuation contribution to the conductivity of thin samples of o-TaS_3, which comprises linear and nonlinear parts. We have shown that the spontaneous phase slippage observed in the CDW in the

vicinity of T_P results to the excess conductivity whose temperature and electric-field dependences match our experimental observations.

The simple model proposed requires further development. In particular, the mechanism of PS nucleation and evolution should be considered in terms of nucleation and propagation of dislocation loops in the CDW.[29] A possible contribution of glide of the dislocation lines to the current[33,34] also requires analysis. In the case of bulk [three-dimensional (3D)] samples the loops cover only part of the cross-section areas, so transversal interaction of the chains while the local creep proceeds should be considered. A special case is the ribbon-like (2D) samples treated in Refs. 7–9. A dislocation loop degenerates into a pair of points interacting logarithmically. Then the system acquires the features of a 2D crystal exhibiting the Kosterlitz-Thouless transition.[35] This approach can explain the lowering of T_P in thin crystals and gradual power-law dependences of σ_{nl} on (T).

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[28] We do not reject the model (Ref. 29), according to which the PS results from the nucleation of a dislocation loop of critical radius. However, this model does not give spontaneous PS in large pure samples and needs additional specifications to describe the loops in real samples. The simple one-barrier model looks good enough to describe empirically the PS dependence on T without deepening into its microscopics.

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[31] In the model above we approximated the interaction of the CDW with the impurities by an average value E_T. Actually, E_T forms over the phase-correlation length scale, which could exceed $L_{2\pi}$ [Eq. (1)] by a unity factor, while we apply the approximation over the length $L_{2\pi}/2$. On this length scale a higher value of E_T is expected, in accordance with the size effect in 0D pinning (Ref. 32).

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