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Macroscopic coherence length of charge-density waves in orthorhombic TaS_3

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Four-probe continuous and pulsed dc current-voltage measurements on orthorhombic TaS_3 showed that the distance between potential contacts influences (i) the sharpness of metal-semiconductor transition and (ii) the threshold potential at which, in the low-temperature phase, nonlinearity sets in. The closeness of voltage contacts on the the 100-μm scale smears out the phase transition and increases the threshold field, indicating that the phase coherence of charge-density-wave states is destroyed by the perturbation of contacts.

A new class of quasi-one-dimensional materials, including TaS_3, $NbSe_3$, Ta_2Se_8I, has attracted general interest in the last few years. A common feature of these materials is the presence of pinned charge-density waves (CDW) at low temperature.[1] Their unusual electronic properties (field dependency, metastable conductivity, narrow-band noise, etc.) due to possible depinning of CDW's by electric field are well known,[2-6] even though certain ones are not yet fully understood. Various theoretical works assign the phase coherence of CDW's a central role. In Bardeen's model[7] of CDW tunneling, either the coherence length ξ (for $NbSe_3$) or the correlation length L (for TaS_3) is in the 10–100-μm range. In the Lee-Rice model[8] of weakly pinned deformable CDW's the threshold field E_T of the nonlinear conductivity is determined by the collective effect of impurities within a coherent domain. Fisher[9] attempted to associate the narrow-band noise above E_T to finite-size effects caused by a divergent coherence length $\xi \sim (E-E_T)^{-\nu}$, where ν is a critical exponent.

Some of the experimental findings such as the small threshold field and the high value of static dielectric constant[3,10] really suggest long-range correlation for CDW's. The combined ac-dc measurements supported speculations about the analogy between the pinned CDW system and the Josephson junction,[11] where the presence of macroscopic quantum states is crucial.

While many authors predicted a coherence length longer than 10 μm no one has made a systematic search for size effects in a more direct manner. In this Rapid Communication we report continuous and pulsed dc conductivity measurements on orthorhombic TaS_3 demonstrating the length dependence of the intrinsic properties of the material.

In the continuous dc measurement, two current and five potential leads with gradually changing distances (60–1200 μm) were connected to the TaS_3 fiber. The contacts were made of 7-μm annealed gold wire using silver paint. The maximum applied field was always less than one-fifth of the threshold field of the nonlinear conductivity. In the pulsed measurements, standard four contact samples were investigated. After a complete temperature run, new potential contacts were mounted on the same sample at a different distance and the measurement was repeated.

Samples were prepared by the gradient furnace method in the presence of excess sulfur. Crystals referred to as A and B were synthesized at the University of California, Los Angeles, while samples donoted by C and D were produced in our laboratory. The cross sections of crystals A and B investigated in the continuous dc measurements were 45 and 65 μm^2; those of samples C and D studied by pulse measurements were 35 and 67 μm^2, respectively.

Room-temperature measurements were carried out to investigate the influence of the finite contact area on the measured potential drop. The magnitude of effective contact distance l is crucial in calculating the electric field E from the potential drop U, particularly for small l comparable to the length of the contact area. As the contact sizes are always much larger than the sample diameter it is somewhat surprising that the resistivity scales quite well with the distance between the middle of the contacts (Fig. 1) meaning that the current lines remain approximately parallel to the sample axis. For an isotropic material with good contacts the current lines are strongly distorted and the appropriate parameter would be the free, noncovered segment length between contacts. Our finding can be attributed either to a high resistance layer at the TaS_3–silver-paint interface or to the highly anisotropic ($\sigma_\perp << \sigma_\parallel$) conductivity of TaS_3. In both cases, charge accumulates in the boundary region caus-

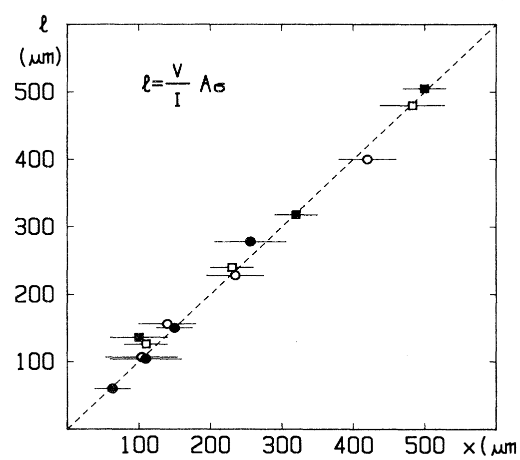

FIG. 1. Effective segment length l vs distance x between the middle of the potential contacts measured by optical microscope. V is the potential difference of the contacts, I is the current. The cross section A is calculated from the resistivity of the longest (x = 700–1200 μm) segment using room-temperature conductivity $\sigma = 2000\ \Omega^{-1}\,cm^{-1}$. Here and in the following figures samples A, B, C, and D are denoted by □, ■, ●, and ○, respectively. The horizontal bars mark the range covered by the two contacts, i.e., the left end of the bars corresponds to the length of the noncovered crystal segment. The slope of the dashed line is 45°.

ing the current to flow approximately parallel to the sample axis.

In the following, the segment length l is calculated from the room-temperature resistivity ratio to the longest segment on the given sample (see Fig. 1).

Temperature-dependent conductivity is presented in Fig. 2. Comparing different segments of the same sample one can see a striking difference, correlating with the segment length. The logarithmic derivative of the conductivity shows that transition temperature T_P is unchanged; however, the transition broadens for shorter contact distance. Outside the transition region the conductivity differences diminish.

The continuous measurements were repeated at ten times smaller currents to check the heating effects and the same features were observed. In the pulsed measurements heating was excluded also.

In a recent paper[5] we have reported that, unless high-field conditioning pulses are applied the Ohmic conductivity depends on the temperature prehistory of the sample. A similar phenomenon was observed[6] on $NbSe_3$. For TaS_3 this effect is well pronounced in the temperature range $100 < T < 180$ K. In the present study we found that around T_P the application of conditioning pulses does not modify the $\sigma(T)$ function, nor does it have any effect on the broadening of the phase transition at shorter lengths.

The pulsed method was applied to measure $\sigma(E,l)$. The threshold voltage V_T for several lengths is given in Fig. 3. Had we used the noncovered distance between the contacts instead of l the deviation from simple $V_P \sim l$ proportionality would have been even greater. Note that the cross sections of samples C and D differ by a factor of 2.

The length dependence of the nonlinear conductivity manifests itself also in the full $\sigma(E)$ function. To demonstrate this point, we present the $\sigma(E)$ curves for three segments of sample C (Fig. 3 inset). We found that for small l the detailed behavior of $\sigma(E)$ is not uniform and may depend on the sample and contact shape. To reduce these spurious effects, the samples were painted all around at the voltage contacts.

In Fig. 4 the temperature dependence of E_T is plotted for two segments of sample D. The threshold field seems to be shifted to higher values for shorter segments in the whole temperature range.[12] The inset shows the length dependence of the threshold voltage for three temperatures.

The different phase transition temperatures of samples B and C indicate differences between the two preparation batches (see Fig. 2). Earlier findings showed that E_T and T_P correlates[13] and accordingly the threshold field for samples A and B were found to be a factor of 2 higher than for samples C and D. Apart from this difference, the $V_T(l)$

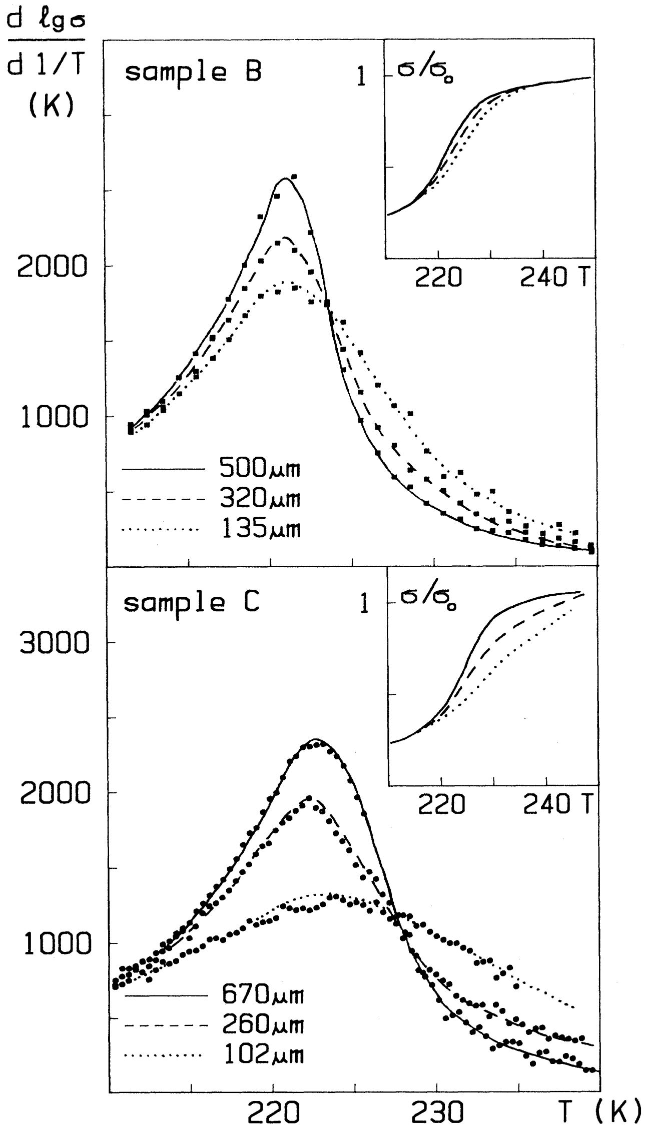

FIG. 2. Logarithmic derivative of the conductivity as a function of temperature. Line types indicate different contact distances. Sample B measured by continuous method and sample C for the pulsed measurements are from different batches. The insets show the conductivities around the phase transition.

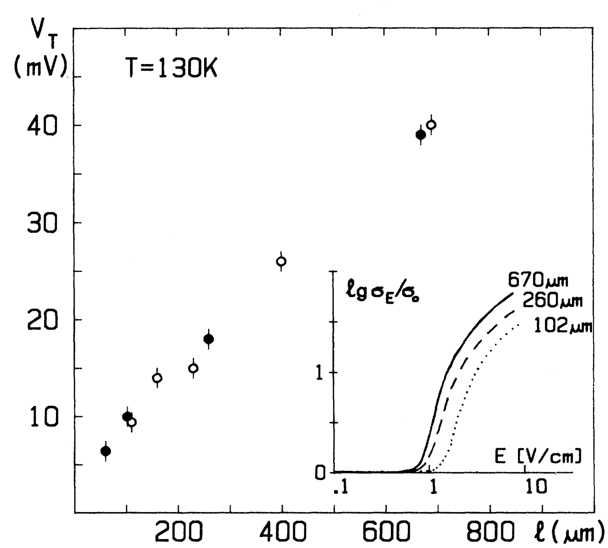

FIG. 3. Threshold voltage vs sample length (●: sample C; ○: sample D). The inset shows the field dependence of the normalized conductivity for three segments of sample C.

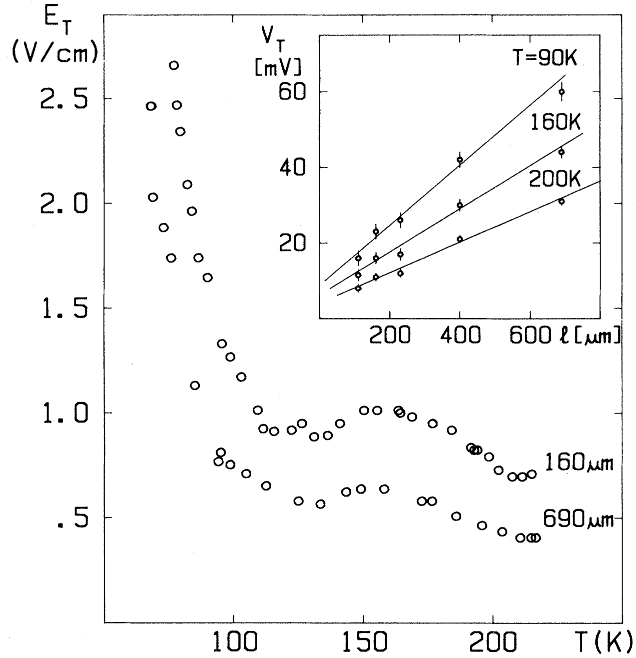

FIG. 4. Threshold field as a function of temperature for two segments of sample D. The inset shows the threshold voltage at different temperatures for five segments of sample D. Solid lines suggest the relationship $V_T(T)=E_0(T)(l+l_0)$.

and $E_T(T)$ curves for samples A and B scale to those of samples C and D (given in Figs. 3 and 4) by the same factor.

In conclusion, we have found size effects in temperature-dependent conductivity around T_P and in the threshold field of the nonlinear conductivity. It is known that although the contact resistance does not play a role in four-probe measurements the potential contacts cannot be considered as weak perturbations.[14,15] We emphasize that our measurements do not support the view that current flows completely in and out at potential contacts.[14] Instead, we believe that there are space-charge distributions within the sample keeping the current lines approximately parallel to the sample axis.

Our conductivity measurements show that contacts closer than 200 μm significantly modify the phase transition, i.e., the contact-induced perturbations spread well over the contact area. The most plausible explanation for this is the presence of macroscopic CDW coherence length in the material.

The critical voltage proved to increase with the contact distance; however, the simple $V_T \sim l$ proportionality does not describe our findings, but rather $V_T=V_0+E_0 l =E_0(l+l_0)$ holds with $l_0 \approx 100$ μm independent of temperature. The complete $\sigma(E)$ curve follows the trend of the length dependence of E_T. The shape of the $\sigma(E)$ function may be influenced by the finite contact sizes, especially for short segments. Nevertheless it is clear that the high-field part of $\sigma(E)$ can neither be scaled by introducing an $l'=l+l_0$ effective length nor by putting an effective voltage of $V'=V-V_0$.

This study showed that those properties connected with ordering and with depinning of CDW's are modified by the short contact distance. However, the conductivity does not depend on the length outside the phase transition region and in the Ohmic regime. Therefore, the observed characteristic distance in the order of hundred microns has to be the correlation or coherence length itself of CDW's.

In a model of deformable CDW's (Ref. 8) Lee and Rice implied that the phase varies on a length scale of $L \approx |\psi|^2/a_x a_y n_i \epsilon^2$, where a_x, a_y are the lattice constants, n_i is the density of pinning centers, and $\epsilon^2/|\psi|^2 < 1$ characterize the strength of the pinnings. For 10-ppm impurities one gets $L > 10$–100 μm domain size. If the potential contacts are closer than L, modification in the depinning process is expected due to the strong perturbation of contacts. As far as the broadening of the phase transition is concerned, the divergence of the Ginzburg-Landau coherence length $\xi=\xi_z(|T-T_P|/T_P)^{-1/2}$ may account for it, but the estimated $\xi_z=100$ lattice spacings[8] is too small to be in accordance with our observations.

In the Bardeen model[7] there are also two characteristic lengths: the correlation length L related to the threshold field by $E_T=\epsilon_g/2e^*L$ and the coherence length $\xi=2\hbar v_F/\pi\epsilon_g$ (here ϵ_g is the pinning gap, e^* is the effective charge of the CDW, v_F is the Fermi velocity, and E^* is a fitting parameter to describe the high-field conductivity). Taking reasonable estimates[3] for TaS_3 ($\epsilon_g=10^{-17}$ erg, $v_F=10^7$ cm/sec, and $E^*=5E_T$) one obtains $\xi=6$ μm and $L=60$ μm, the latter one being close to the range we reported here. It would be interesting to investigate $NbSe_3$ by a similar method because in that case[7] $L \ll \xi \approx 100$ μm and the Bardeen model predicts a different type of size effect.

It has been demonstrated[4,16] that two critical fields exist in TaS_3: E_{MS}, where metastable conductivity change begins to develop, and E_T for nonlinear conductivity. We observed that although E_T increases with decreasing l the other critical quantity E_{MS} is constant.[16] A more detailed study of the connection between these two quantities and the pulse sign memory[4,5] of the pinned CDW's is in progress.

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