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Rapid Communications

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Current-induced remanent polarization of charge-density waves

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It is found by measuring the low-field conductivity of segments of TaS_3 single crystals that a current pulse equal to or larger than threshold for sliding charge-density-wave (CDW) conduction permanently destroys the inversion symmetry of the sample. The segments are limited at one end by the usual silver paint contacts which strongly perturb the motion of CDW's while at the other end a special weakly perturbing contact is applied. The low-field resistance of the segments depends on whether the last sliding CDW current pulse enters or exists the strongly perturbing contact. The results are interpreted as a direct confirmation of current-induced remanent CDW polarization recently reported by Janossy, Mihaly, and Kriza.

The presence of metastable states and consequently the appearance of various hysteretic or "memory" effects seems to be a general property of charge-density-wave (CDW) conductors. Metastable CDW states depending on the electric[1-7] and thermal[8-11] history and also a combination[8,12] of these have been reported by a number of authors. The simplest manifestation of metastability is the thermal hysteresis of the low-electric-field conductivity observed up to the present in $NbSe_3$,[13] TaS_3,[8,9] and $K_{0.3}MoO_3$.[11] Another striking phenomenon is pulse sign memory charging,[3,4] a transient observed in pulses with alternating signs, which indicates a remanent CDW displacement. Metastable states may be rearranged by CDW currents as seen by the sudden changes in the thermally induced metastable resistance if high-field pulses are applied.[8] Thermal relaxation of these states in samples left alone is, however, extremely slow,[10] thus the expression "memory effects" is in general use.

Metastable states are assumed to reflect nonequilibrium deformations of the CDW system. Sneddon, Cross, and Fisher[14] pointed out that a quantitative agreement between theory and experiment may be expected by a full analysis of the deformable CDW model of Fukuyama and Lee.[15] The memory effects together with other switching and hysteretic phenomena prompted speculations about charge-density-wave deformations.

Recently, a coherent qualitative description[7,8,12] of memory effects has been proposed. It is suggested[12] that an electric field above threshold for depinning the CDW's induces a long-range deformation which becomes frozen into the system once the field is switched off. A sample so treated is brought into an asymmetric state, in which the CDW's are compressed toward one end of the sample and depressed toward the other. The low-field resistivity is assumed[8] to probe the CDW deformation by measuring the number of normal charge carriers, i.e., the magnitude of the single particle gap ϵ_g which, in turn, depends on the CDW deformation. The current-induced deformation is inhomogeneous; it is antisymmetric around the center of the sample, and $\epsilon_g(x)$ together with the resistivity $r(x)$ depend on position (Fig. 1). The inhomogeneity of ϵ_g induced by a high current pulse has been experimentally verified through an indirect method by Janossy, Mihaly, and Kriza.[12]

In this Rapid Communication we present direct evidence for a CDW current-induced remanent asymmetry in orthorhombic TaS_3 single crystals by measuring the resistance of segments at 77 K. The idea of the experiment is the following. Conventional silver paint electrode contacts act as extremely strong pinning centers as demonstrated by the narrow-band noise spectra of $NbSe_3$ segments[16] and the broadening of the Peierls transition[17] in short segments of TaS_3. The silver paint effectively short circuits the portion of the sample covered by it and significantly decreases the local electric field. A sliding CDW current induces a CDW deformation between silver contacts. However, the deformation is antisymmetric around the center and to a first approximation there is no change in the total resistance R between the silver paint contacts. The increase of $r(x)$ toward one contact is compensated by a decrease toward the other. In order to probe the potential without significantly disturbing the CDW motion we simply touched a 10-μm-diam gold wire to the sample and fixed it with insulating glue. The resulting high (≈ 200 kΩ) contact resistance has a negligible effect on the current flow. It is quite possible that the touching contact is a tunneling probe.

The contact arrangement is shown on Fig. 1(a). Altogether five contacts are applied. Contacts 1,5 and 2,4 are silver painted current leads and voltage electrodes, respectively, while the touching contact 3 serves to probe the potential drops of the two segments limited by contacts 2,3 and 3,4.

Conditioning pulses I_c are applied alternately in both directions between 1 and 5. The current is considered posi-

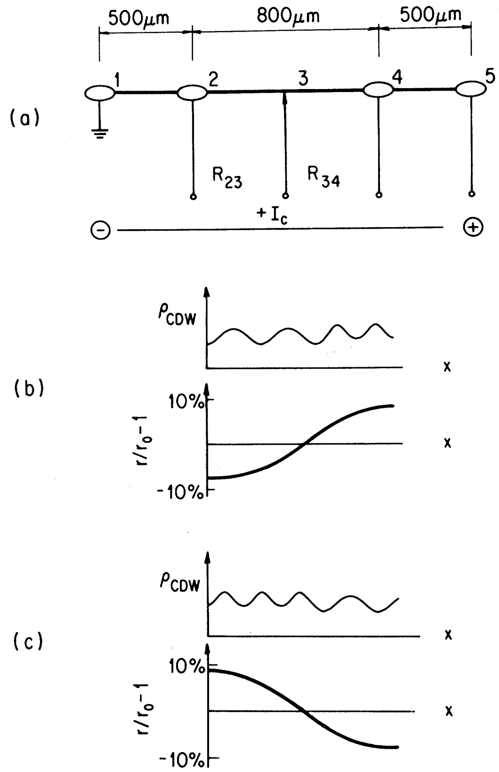

FIG. 1. (a) Five-probe sample arrangement to study the current-induced asymmetry of the resistivity. Contacts 1, 2, 4, and 5 are silver painted, contact 3 is a touching gold wire. The conditioning current I_c is considered positive if contact 5 is at a higher potential than contact 1. (b) and (c) Deformed charge-density wave and inhomogeneous resistivity schematically drawn. (b) and (c) differ in the sign of the pulse I_c above threshold preceding measurement. ρ_{CDW} is the oscillating part of the CDW. r is the local resistivity; r_0 is the resistivity of the undeformed state.

tive if the potential at contact 5 is greater than at contact 1. Following each pulse I_c the low-field resistances between segments 2,3 and 3,4 (and as a check between the silver paint contacts 2,4) was measured by a 100-MΩ input impedance lock-in amplifier using a 500-Hz 0.1-μA current through contacts 1,5. We shall denote the measured low-field resistances of segments 2,3 by $R_{23}(+I_c)$, $R_{23}(-I_c)$, and similarly for segment 3,4 by understanding that these are measured by currents much smaller than I_c and reflect the metastable resistance *after* a pulse I_c or $-I_c$ was applied.

Six samples of the same high purity TaS_3 batch were measured. We obtained similar results in all crystals: A CDW current-direction-history dependent effect was observed with about the same magnitude. In addition to the effect reported here, some of the crystals showed an asymmetry between silver contacts 2,4 which may be associated with small irregularities along the sample or at the contacts.[5]

We discuss in detail results obtained on a sample 800 μm long between silver contacts 2 and 4 with a gold wire touching contact 3 placed halfway [Fig. 1(a)]. The sample was placed in liquid N_2 in order to maintain a homogeneous stable temperature. At 77 K the threshold voltage for nonlinear conduction between contacts 2,4 was $U_T=90$ mV ($E_T=1.1$ V/cm) and the corresponding threshold current was $I_T=25$ μA. The width of conditioning current pulses I_c was typically 100 ms.

Rapid cooling of the sample from room temperature to 77 K gave rise to a nonequilibrium resistance due to a temperature-induced deformation.[8] Application of a current pulse $I_c=4\ I_T$ increases the resistance R_{24} between contacts 2,4 by 7%. After this initial increase, no more change was observed in R_{24} with application of further current pulses of any magnitude or sign. As has been shown,[8] after the first large current pulse R_{24} becomes equal in magnitude to the resistance in the equilibrium state measured by the average of the temperature hysteresis loop. Our experiment shows, however, that current pulses above threshold do not leave the sample in an equilibrium state despite the fact that resistance R_{24} between two strongly perturbing silver paint contacts remains unchanged with time.

This is demonstrated by measuring the resistances of two segments R_{23} and R_{34}, each limited by a strongly perturbing silver paint contact (2 or 4) at only one end, the common contact 3 does not perturb appreciably the CDW current. The low-field resistances are measured after current pulses I_c and $-I_c$ of alternating signs. We find that if I_c is larger than threshold and positive (flowing from 1 to 5) then R_{23} is increased and R_{34} decreased compared to the values they have following a negative current pulse $-I_c$. Figure 2 displays the relative resistance differences $\delta_{23}=\Delta R_{23}/R_{23}$ and $\delta_{34}=\Delta R_{34}/R_{34}$, where $\Delta R_{23}=R_{23}(+I_c)-R_{23}(-I_c)$ and $\Delta R_{34}=R_{34}(+I_c)-R_{34}(-I_c)$, respectively. For conditioning currents I_c below threshold δ_{23} and δ_{34} are zero. For I_c above threshold the resistances depend on the preceding current pulse, we find $\delta_{23}=-\delta_{34}$. δ_{23} increases with I_c until about $I_c=2I_T$, above this it becomes constant. This means that once a current pulse $I_c>2I_T$ is applied, the resistance change with the polarity of I_c is independent of the magnitude of I_c at least up to $6I_T$. The maximum relative resistance changes are $\delta_{23}^{max}=-\delta_{34}^{max}=3\%$.

The above observations can be arranged in a coherent picture,[12] based on the deformability of CDW's, together with

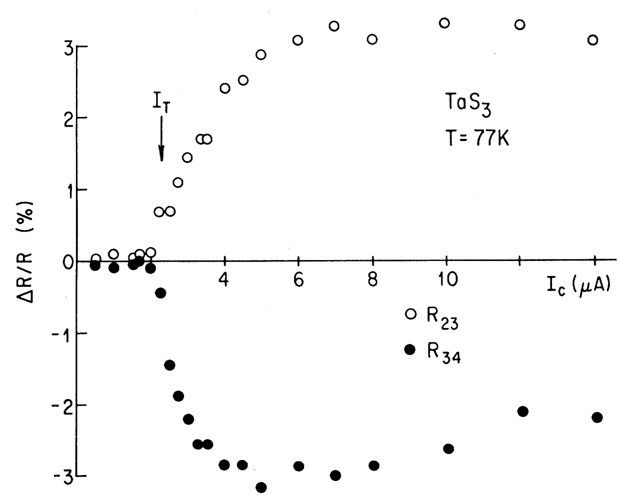

FIG. 2. Resistance changes of segments 2,3 and 3,4 as a function of preceding conditioning currents I_c. Circles correspond to $\Delta R/R=2[R_{23}(+I_c)-R_{23}(-I_c)]/[R_{23}(+I_c)+R_{23}(-I_c)]$, full dots denote the same quantity for R_{34}. Note the opposite signs of the effect for R_{23} and R_{34}: If a given current pulse increases R_{23} then it decreases R_{34} by approximately the same amount.

the pulse sign memory, the temperature and electric-field-dependent hysteresis of the low-field conductivity. Here we briefly discuss the case of an electric-field-induced deformation relevant to the experiment.

Consider a case where an electric field between the strongly perturbing contacts is somewhat above the threshold for depinning the CDW's from impurities. The electric field in the part of the sample under the silver paint contacts is much smaller than within the two contacts and it will be below threshold so here the CDW remains pinned.[16] The charge carried by a CDW current entering the contact must boil into normal carriers in a region near the contact[18] where the order parameter falls to zero. This region creates a barrier to CDW conduction and a long-range deformation of the CDW system appears with opposite signs at the two contacts. Thus the contacts give rise to a giant CDW polarization. If the CDW current is switched off the polarization, i.e., the antisymmetrically deformed state remains frozen by impurity pinning which inhibits continuous changes of the CDW phase in the absence of an electric field. The frozen metastable state relaxes very slowly[7,10] towards the equilibrium homogeneous state. A CDW current of opposite sign reverses the polarity of the deformation.

The experiment gives evidence for the polarized state as the normal resistivity at a given part of the sample measures the local CDW deformation in the following way. A deformation can be viewed as a local change in the charge transferred from the lattice to the metal ion chains. In regions where the electron charge transfer is increased from equilibrium the Fermi momentum k_F also increases giving rise to a change in the gap for normal excitations ϵ_g. In regions of deformations of opposite signs k_F decreases and ϵ_g changes also in the opposite way. The change in resistivity $\exp(\epsilon_g/kT)$ is a result of the local change in ϵ_g.

A measurement of the asymmetric change of ϵ_g by a current pulse was reported by Janossy, Mihaly, and Kriza.[12] By the method presented in this Rapid Communication we measured directly the variation of the resistivity along the sample. The results show an increase of resistivity in the half of the sample where previously a CDW current entered through a strongly perturbing contact and a decrease in the half where the CDW current left through such a contact. The resistivity changes are antisymmetric for the two segments of the sample and occur only when the field is above the threshold for CDW conduction. These results are in agreement with the model: Only CDW currents can change the polarization.

The appearance of a polarized CDW state may have important consequences on the dynamics of the CDW motion. It certainly helps understanding some of the complicated time-dependent phenomena. The pulse sign memory charging,[3,4] a highly conducting transient state observed when a current above threshold is reversed, is due to the displacement CDW current occurring while the giant polarization is reversed. Switching[2] between a normal and a highly conducting state observed for currents near threshold is also a consequence of the giant CDW polarization but in a more subtle way.[7]

In conclusion, we demonstrated that current pulses above the threshold current for sliding CDW conduction induce a remanent polarization in the CDW system of TaS_3. In order to measure the polarization we developed a method to measure the potential along the sample without significantly disturbing the motion of the CDW. The results are interpreted in the framework of a model proposed to describe the metastability in CDW systems.

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