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Metastable length states of a random system: TaS_3

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The sample length of the charge-density-wave (CDW) conductor TaS_3 is found to be a sensitive function of the applied dc bias electric field E and time t. The length reflects transitions through metastable CDW configurations for both the pinned and sliding CDW in the presence of random impurities. Although a simple theoretical model can account for some qualitative features of the experimental findings, our experimental results are inconsistent with present CDW elasticity theory.

The physics of random systems is currently of great theoretical and experimental interest. Solid-state random systems such as spin glasses and disordered conductors display unusual effects ranging from slow relaxation and metastability to localization and frequency-dependent conductivity.[1] Charge-density-wave (CDW) conductors are relevant to the study of random systems in that the collective-mode CDW condensate can be pinned by a random distribution of impurities, leading to metastable electronic configurations.[2] Metastable states are believed to be the origin of numerous anomalous CDW transport effects such as polarization memory,[3] low-field resistivity hysteresis[4,5] and relaxation,[6] and stiffness (Young's modulus) hysteresis.[7,8] An intriguing but poorly understood problem is how the metastable electronic CDW configuration is reflected back (via the CDW lattice interaction) onto the underlying lattice structure containing the random impurities.

We have investigated the host crystal lattice structure for different CDW electronic configurations in the CDW conductor TaS_3 through measurements of the total sample length L as a function of applied dc bias field E and time t. We find that L is extremely sensitive to E, in many cases even for $E < E_T$, where E_T is the threshold field for CDW depinning. $L(E)$ is hysteretic but can be carefully and repeatably controlled by E, similar to the situation for piezoelectric insulators. The magnitude of the length change effect with bias field is orders of magnitude larger than that for a comparably sized commercial piezoelectric or than what would be expected exclusively from an E-induced softening of the Young's modulus of TaS_3. We interpret the length states as directly reflecting metastable CDW configurations. Using the length change ΔL as a probe, we follow the metastable state evolution for various E. The qualitative properties of our results are well explained by a simple model of a deformable CDW interacting with fixed impurities, where randomness plays the central role. However, the size of our effect may be difficult to interpret within existing theories of CDW elasticity.[9,10]

The experimental setup consisted of a single high-purity TaS_3 fiber clamped between a piezoelectric bimorph and a soft aluminum foil leaf spring with an adjustable spring preload. Bias current was passed through the ends of the sample using silver paint contacts in a two-probe configuration. In early experiments, the sample length was monitored with a cryogenic tunneling microscope. A more reliable method proved to be a modified version of a helical resonator detector.[11] One end of a copper helix is grounded to the outer wall of an electromagnetic cavity. The cavity resonant frequency f_0 is extremely sensitive to the termination conditions at the free end of the helix. In this experiment, we separate the grounded Al leaf spring and the helix tip by ~ 100 μm. Changes in sample length alter the helix-spring separation and f_0. Extraneous vibrations were filtered out with a time constant of 3 sec, yielding a sample length change resolution better than 0.2 Å. The bimorph was calibrated optically at 300 and 77 K and is only used to map the measured changes in resonant frequency to changes in sample length. Ten TaS_3 crystals were examined, whose lengths and room-temperature resistances varied from 0.5 to 1.0 cm and from 30 Ω to 1 kΩ, respectively. Sample threshold fields were of order 200 mV/cm at 77 K.

Samples were cooled under a small but finite stress ($\sim$MPa) with no applied current in ~ 1 torr of He. After stabilizing the temperature at 77 K with drifts of < 5 mK/min, a continuous triangle-wave bias current at ~ 0.01 Hz was applied to the sample and significant length changes occurred. After several cycles in which $|E|$ exceeded E_T, $L(E)$ would stabilize to a closed hysteresis curve. Figure 1(a) depicts this steady state $L(E)$ for one sample under an applied stress of 0.36 MPa. For this sample, ΔL, the difference of the two lengths at $E = 0$, is 40 Å, corresponding to a difference in strain of $\sim 5 \times 10^{-7}$. Even for $E \sim 3E_T$, the lengths for increasing and decreasing fields are not identical, suggesting some "memory" even in the sliding state. Heating effects are visible as an upward curvature at large E. From the form of $L(E)$ at higher temperatures, we estimate that sample heating accounts for ~ 2 Å length increase at E_T.

If the CDW affected the Young's modulus Y but not the equilibrium interionic spacing, then one would expect $L(E)$ to appear as shown in the lower part of Fig. 1(a). For this calculation we have assumed that Y softens by 2% just above E_T, consistent with low stress vibrating-reed results.[12] (Recent more direct measurements of Y with

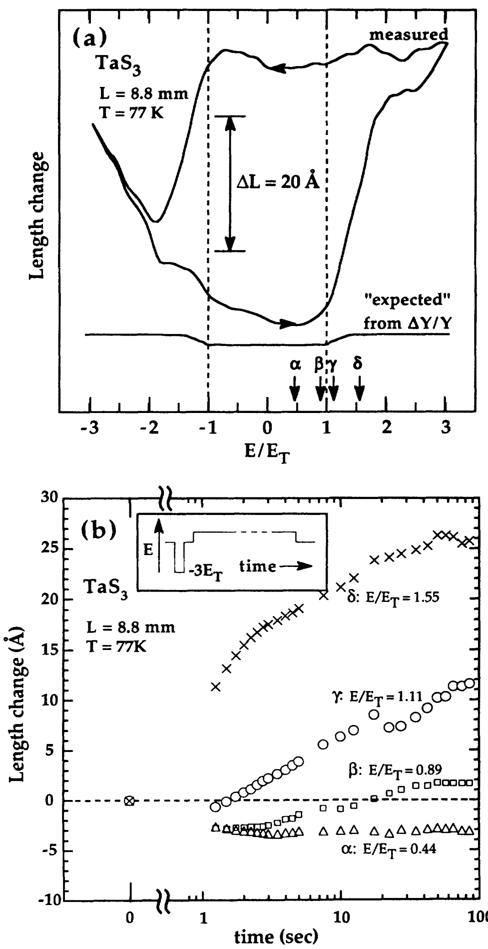

FIG. 1. (a) Change in sample length L vs bias field E. The applied triangular wave form for E has a frequency of 0.01 Hz. The "expected" length change is calculated assuming a softening at E_T of Young's modulus ($\Delta Y/Y$) $\approx$ 2% (see text). (b) Length change plotted as a function of time t for the fixed bias fields E denoted by α, β, γ, and δ in (a). The sample is first "initialized" with a bias pulse of $-3E_T$ as described in the inset. Length changes are measured relative to the length at $t=0$, the rising edge of the positive bias step E.

large applied stresses suggest no softening within $\pm 0.2\%$.[13,14]) Any of the reported hysteretic effects[7,8] in Y would be too small to see on this scale. Clearly the changes we see are too large to be explained by published changes in Y.

Increasing the range over which E is swept changes the hysteresis only slightly. The shape and size of the steady state curve for a given sample are unaffected by repeated warming and cooling cycles and by the direction in which E_T is first exceeded. Typically, the initial sample length does not lie on the steady-state hysteresis curve, a fact we attribute to nonrecoverable thermal stresses being frozen into the sample during cooling. However, no anomalously large length changes are in general detected in the first bias field cycles. The size of the hysteresis curve decreases with increasing temperature and disappears completely above the Peierl's temperature. A surprising characteristic of the length hysteresis is its dependence on the stress distribution. The effects vary between samples, but applying a bending moment to the sample can alter the sense in which the hysteresis loop is traversed and increase its size by an order of magnitude. In some samples, mounted in a different "clamped-free" configuration, significant E-induced bending effects are also detected, the size of which (lateral displacements $\Delta L_\perp$ of $\sim$0.1 μm at the free end of a 3-mm sample) may be large enough to affect the interpretation of previous Young's and shear modulus experiments. The "apparent" length change caused by these bending moments scales as $\Delta L_\perp^2/L$ and is too small to explain our observed length changes.

The metastable length states at $E=0$ appear to be extremely stable with no decay evident during 24 h. Effects suggestive of glassy decays are seen in the time dependence of L for $E\neq 0$. For the four bias field values denoted by α, β, γ, and δ in Fig. 1(a), the sample length L is plotted as a function of time t in Fig. 1(b). As shown in the inset of Fig. 1(b), a bias field pulse of $-3E_T$ is first used to "initialize" the CDW system; at $t=0$ we then apply a positive bias step E after which $L(t)$ is monitored. Even for curves α and β, where $E<E_T$, significant evolution in ΔL occurs—a decrease in L for this sample. As E approaches E_T, two processes occur, a fast decrease in length followed by a slow increase. At intermediate times, we see a roughly logarithmic dependence, a signature of glassy dynamics, which appears to saturate for $t>60$ sec.

Changes in the sample length before depinning are more clearly seen for the sample depicted in Fig. 2(a). From experiments on the resistive hysteresis in TaS_3 and $NbSe_3$, it has been suggested that strains in the CDW alter the number of free carriers and R_0, the resistance at $E=0$.[4,5] The differential resistance in Fig. 2(a) surprisingly shows no dependence on E for $E<E_T$ even for E values for which the length has changed substantially. There must therefore be some CDW strains which do not couple to R_0. In all samples studied, there occurs a small hysteresis in dV/dI; the size of the hysteresis is found to scale roughly with the size of the ΔL hysteresis, the smaller L state having greater differential resistance, suggesting that some CDW strains do couple to R_0.

Many qualitative features of our experimental results can be explained by a simple model of a deformable CDW interacting with impurities fixed in a lattice. We adopt a model similar to the conventional one where the CDW is represented by a number of CDW domains (i.e., phase coherent volumes) interacting with each other via nearest-neighbor coupling.[2,15] The difference is that we have introduced the freedom for the host lattice to deform under the strain generated by the interaction between the CDW's and the host, mediated by the impurity interaction. The Hamiltonian we use for N domains is

$$H=\sum_i \tfrac{1}{2} a_{i,i-1}(\phi_{i,i-1}-\theta_{i,i-1})^2+\sum_i \tfrac{1}{2} b_{i,i-1}(x_{i,i-1}-l_{i,i-1})^2-\sum_i V_i\cos(\phi_i)-E\sum_i \rho\left(\frac{l_{i+1,i}+l_{i,i-1}}{2}\right)\phi_i+g(x_1-x_N)\,, \tag{1}$$

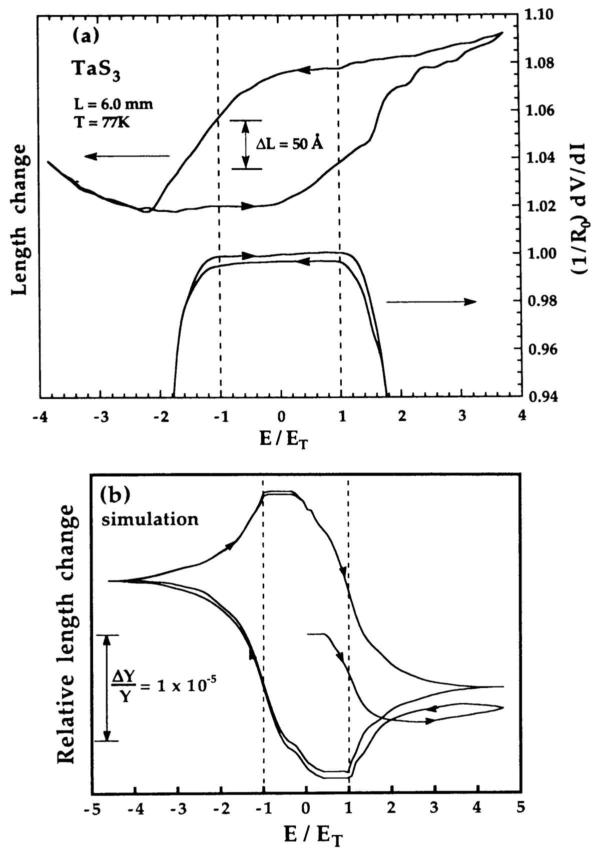

FIG. 2. (a) Change in sample length L and normalized differential resistance $(1/R_0)dV/dI$ plotted as functions of the bias field E. R_0, the low-field resistance, $\approx 77\ \mathrm{k\Omega}$. (b) Relative length change of our simulated system of CDW domains [Eq. (1)] as a function of bias field E (see text).

where $\{\phi_i\}$ are the CDW phases of the different domains, $\{x_i\}$ $(x_i > x_{i-1})$ are the locations of the impurity sites, $\{l_i\}$ are their positions at equilibrium (i.e., in the absence of CDW's), λ is the CDW wavelength, $x_{i,i-1} = x_i - x_{i-1}$, $l_{i,i-1} = l_i - l_{i-1}$, $\theta_{i,i-1} = 2\pi x_{i,i-1}/\lambda$, $\phi_{i,i-1} = \phi_i - \phi_{i-1}$, V_i is the strength of the impurity potential at x_i, E is the external electric field, ρ is the CDW charge density, and g is the external tension. The spring constants for the CDW's and the lattice are dependent on $\{l_i\}$ so that $a_{i,i-1} = A/l_{i,i-1}$ and $b_{i,i-1} = B/l_{i,i-1}$, where A and B are the Young's moduli of the CDW and lattice, respectively. The randomness, which is essential in reproducing the experiment, is introduced in $\{l_i\}$ and $\{V_i\}$. The equations of motion are obtained by assuming overdamped dynamics typical of CDW's:

$$\dot{\phi}_i = -\gamma_{\phi,i}\frac{\partial H}{\partial \phi_i}, \quad \dot{x}_i = -\gamma_{x,i}\frac{\partial H}{\partial x_i}. \tag{2}$$

We assume that the domains of smaller size have higher relaxation rates and use the form $\gamma_{\phi,i} = 2\Gamma_\phi l_0/(l_{i+1,i} + l_{i,i-1})$ and $\gamma_{x,i} = 2\Gamma_x l_0/(l_{i+1,i} + l_{i,i-1})$ where l_0 is the average of $l_{i,i-1}$, and Γ_ϕ and Γ_x are the average relaxation rates for the CDW and the lattice, respectively.

We have performed our simulation with 32 CDW domains and parameters[16] which are roughly consistent with experimental results ($Y_{\mathrm{CDW}}/Y_{\mathrm{lattice}} \sim 0.02$ and $l_0 \sim 0.7\ \mu$m). Both the lattice and the CDW are given free boundary conditions. If fixed boundary conditions are used for the CDW, no length changes occur. For the simulation we use a slow triangle wave signal for E and calculate the total sample length L for different E, similar to the experimental determination of $L(E)$ at long time t. [Because of computational difficulties associated with bias field singularities, we have not studied the detailed time dependence of $L(E,t)$ at fixed E.] The results of two cycles of E is shown in Fig. 2(b). Many of the features seen in the experiment are reproduced: (a) we observe only two length states as the bias is swept toward zero; (b) changes in the internal state of the CDW below threshold $(|E| < E_T)$ are clearly observed in the length; (c) the length for sliding states $(|E| > E_T)$ depends on sample history; (d) the final hysteresis curve is insensitive to the initial sweep direction (positive or negative); and (e) our CDW state at $t = 0$, which we initially generated by relaxing the CDW from "equilibrium," is never recovered once a bias field is applied. With these agreements, we believe that the model Hamiltonian, Eq. (1), captures much of the essential physics involved in the experiment.

Based on these results, we suggest that the measured length hysteresis in TaS_3 is the result of changing $\Delta\Phi$, the total CDW phase change between the two ends of the sample ($\phi_N - \phi_1$ in our simulation). Decreasing $\Delta\Phi$, by removing CDW wavelengths from the interior of the sample, expands the CDW which then exerts a compressive stress on the host lattice. Length changes can only occur if phase slip occurs either at the contacts or within the sample. For the observed length hysteresis to occur, the inversion symmetry of a perfect TaS_3 crystal must be broken. In our model, this occurs through randomly distributed impurities. In actual samples, it must arise from the randomly distributed impurities together with any significant applied stress distributions or contact perturbations.

In the simulation, length changes arise from fluctuations in the pinning potentials and the relative length changes, $\Delta L/L$, vary as $D^{-1/2}$ for fixed domain size where D is the number of domains. Scaling the results of the simulation ($\Delta L/L \sim 2\times10^{-5}$) to an actual sample of 10^8 domains yields an expected $\Delta L/L \sim 10^{-9}$, much smaller than our measured $\Delta L/L \sim 10^{-6}$. Our sample volume is $0.88\ \mathrm{cm}\times 25\ \mu\mathrm{m}\times 25\ \mu\mathrm{m}$ and we use a domain volume of $0.7\times0.2\times0.2\ \mu\mathrm{m}^3$. (There are larger published values for l_0, the coherence length along the conduction axis;[5] however, if we increase l_0 at fixed sample length in our model, then $\Delta L/L$ scales as $l_0^{-1/2}$.) Though we expect the model to yield only approximate results for ΔL, this large discrepancy is surprising. It suggests that there are coherent effects between various domains; these effects are not accounted for by the existing theories of CDW elasticity.[9,10]

To further investigate the remarkable sensitivity of L to different CDW states, several experiments suggest themselves. It may be possible to correlate the length changes to the amount of disorder by introducing imperfections or altering the sample length, and it may also be possible to observe the pulse duration memory effect as discussed by Coppersmith and Littlewood[17] in a length measurement. It would be interesting to examine other CDW systems, such as $NbSe_3$, $(TaSe_4)_2I$, or $K_{0.3}MoO_3$ (in $K_{0.3}MoO_3$, a material which exhibits no ΔY at E_T, no changes in L to ± 1 ppm have been detected[18]). The effect of bending should also be investigated to determine whether it affected previous Y measurements.

In summary, we have found the first results that the CDW metastable state directly affects the macroscopic length of samples of TaS_3. The following information has been shown about the CDW metastable state: Significant changes in the metastable state occur for $E < E_T$ which are not detected in the low-field resistance; the dynamics of the random CDW system are such that only two possible CDW states occur as $E \rightarrow 0$ from either above or below; and relative to the CDW elastic state no definite threshold field is evident. Many qualitative features of our experiment can be explained by a surprisingly simple model of a deformable CDW interacting with randomly located impurities; however, the size of the length changes is difficult to explain within existing theories of CDW elasticity. This effect is an important tool for investigating the evolution of metastable states in CDW systems and in the study of random systems in general.

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