RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 52, NUMBER 12 15 SEPTEMBER 1995-II

Static and low-frequency shear compliance of TaS_3

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(Received 23 March 1995; revised manuscript received 5 July 1995)

We have measured torsional strains in the charge-density-wave (CDW) conductor TaS_3 as functions of stress (i.e., torque), stress frequency (2 mHz to 20 Hz), and sample voltage. At all frequencies, the compliance is observed to increase by $\approx 25\%$ when the voltage is increased above the threshold for CDW depinning, implying that the time constant for CDW relaxation in the pinned state is >84 s. Above 30 mHz, the depinned compliance has a slow frequency dependence, implying a large distribution of relaxation times extending to at least 5 s. We also observe a current-reversible strain-memory effect, which may account for the failure of previous investigators to observe static elastic softening when the CDW is depinned.

There are many surprising anomalous properties of quasi-one-dimensional conductors associated with depinning and motion of density waves when voltages greater than the depinning threshold (V_T) are applied.[1] Among the most poorly understood are large changes in dynamic elastic moduli.[2] In orthorhombic TaS_3,[3] the best-studied material, measurements on mechanical resonances between 10 Hz and 1 kHz have shown that the Young's modulus (Y) decreases by 2% and the shear modulus (G) by 20% when the charge-density wave (CDW) is depinned.[4] While these anomalies are roughly independent of frequency in this range, the Young's modulus anomaly decreases roughly as $f^{-3/4}$ above 1 kHz.[4,5] On the other hand, in static measurements of the Young's modulus, no softening ($|\Delta Y/Y|<10^{-5}$) has been observed.[6]

The most successful model of these elastic effects is that of phase-coherent (Lee-Rice)[1,7] domains relaxing in the changing strain, with an average relaxation time that decreases as the voltage increases beyond V_T.[4,8,9] While Mozurkewich showed that the dynamic Young's modulus anomaly could be quantitatively understood in terms of longitudinal changes in Lee-Rice domains,[8] the large shear modulus anomalies suggest that the most important changes are in the transverse domain dimensions.[10] To better study the dynamics of the shear modulus changes, we developed a system to directly measure torsional strain (twist angle) as a function of voltage and stress (torque) at low frequency (<20 Hz). The shear compliance $J=\Delta\epsilon/\Delta\sigma=1/G$. Unlike the (more precise) resonance techniques, these measurements have the advantage that a continuous range of frequencies of *identical* elastic distortions can be studied. For relaxational dynamics, we expect[4] $J(V,\omega)/J_0-1=\mathrm{Re}\int\{A(\tau,V)/(1+i\omega\tau)\}d\tau\approx\int A(\tau,V)(1-\omega^2\tau^2)d\tau$, where A is the relaxation strength and the last expression holds for $\omega\tau\ll 1$.

TaS_3 crystals grow as thin fibers (typically 1 cm long in the high-conductivity direction, and 10 μm^2 in cross section[3]), ideally suited to be incorporated as elastic elements of torsional pendula.[10] For the present measurements, a thin magnetized steel wire was glued (with silver paint) perpendicular to a TaS_3 crystal at its center. The ends of the sample were clamped with silver paint to current contacts. (Sample resistance was measured with this two-probe configuration.) Torque was applied to the sample with a variable magnetic field created by Helmholtz coils (5 mT/A). (No attempt was made to shield the background magnetic field.) The sample was located in a helical resonator rf cavity[11] ($\nu\approx 450$ MHz, $Q\approx 300$) so that motion of the magnet wire changed the resonant frequency of the cavity. Oscillations in the orientation of the magnet-wire phase modulated the output of the cavity,[11] while static twist of the sample could be detected by driving the cavity with an FM wave.

All results reported below were on a relatively thick (≈ 50 μm^2 cross section), 4.2-mm-long "pure" (threshold electric field at 102 K of 130 mV/cm) crystal of TaS_3 at 102 K, well below the CDW transition (220 K).[3] The magnetic wire was ≈ 60 μm thick and 1 mm long. While similar results were obtained with other (thinner) samples, the relatively large size of this sample and small size of the magnet reduced strains due to the background magnetic field. For this sample, a field of 1 mT (the largest used, in quasistatic measurements) twisted the sample about 1.5°, corresponding to a strain of less than 10^{-4}. Alternating fields were kept an order of magnitude smaller than this, and oscillation frequencies were more than an order of magnitude less than the torsional resonant frequency of the sample. The magnet angle (θ) drifted and occasionally jumped, presumably due to the sample or magnet slipping in the silver paint.

The voltage dependence of compliance measured in alternating magnetic fields (30 mHz–20 Hz) is shown in Fig. 1. Also shown is the resistance measured simultaneously; the decrease in resistance at threshold voltage V_T is generally taken as the sign that the CDW has become depinned and is sliding in the crystal.[1] The inset shows the resistance and compliance at two frequencies for an extended voltage range, while the main figure shows the behavior near threshold for a large range of frequencies. As previously reported, the threshold electric field for elastic changes $V_T(J)$ is less than that for noticeable resistance change $V_T(R)$.[10] However, within its scatter, the elastic threshold is independent of frequency and the compliance below threshold is constant, indicating that the pinned relaxation time is longer than the maximum $1/\omega$ investigated (5 s).

In Fig. 2, we show the frequency dependence of the compliance at several voltages above the elastic threshold. The frequency dependence is logarithmic, much weaker than the quadratic dependence expected from a single (voltage-dependent) relaxation time. The logarithmic dependence ex-

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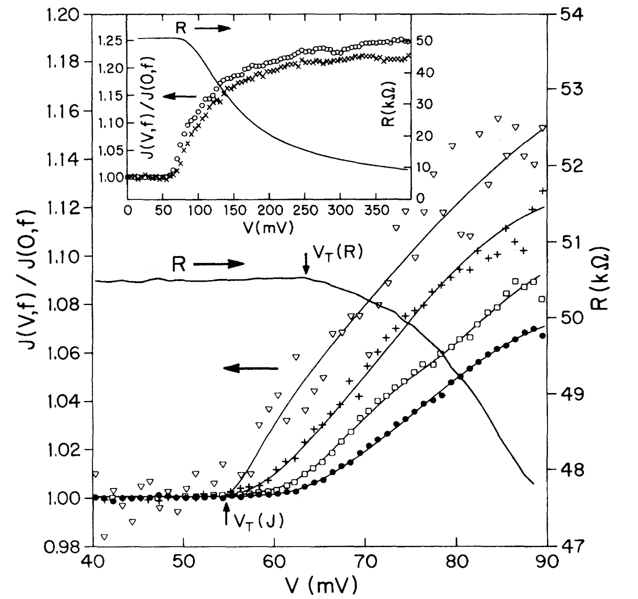

FIG. 1. Voltage dependence of compliance, normalized to its $V=0$ value, at several frequencies: open triangles, 0.030 Hz; plus signs, 0.30 Hz; open squares, 3.2 Hz; closed circles, 19.6 Hz. the curves are guides for the eye. Also shown are the resistance, the elastic threshold voltage $[V_T(J)]$, and the resisitive threshold voltage $[V_T(R)]$. Inset: The normalized compliance and resistance over an extended voltage range: open circles, 1.2 Hz; crosses, 13.2 Hz.

tends, with no sign of saturation, to the lowest frequency investigated, even at voltages much above threshold. This means that (i) for every voltage, the distribution of relaxation times covers several orders of magnitude, with $\tau_{MAX}>5$ s and (ii) it will be necessary to go to even lower frequencies to determine the voltage dependence of the integrated relaxation strength $\int A(\tau,V)d\tau$. If the shear modulus behaves similarly to the Young's modulus, the logarithmic frequency dependence presumably cuts off (for $V>2V_T$) near 1 kHz, where $f^{-3/4}$ behavior sets in for the latter. If so, the distribution of relaxation times also extends to $\tau_{MIN}<0.2$ ms; for small τ, the voltage dependence of A has been shown to be nontrivial.[4,12]

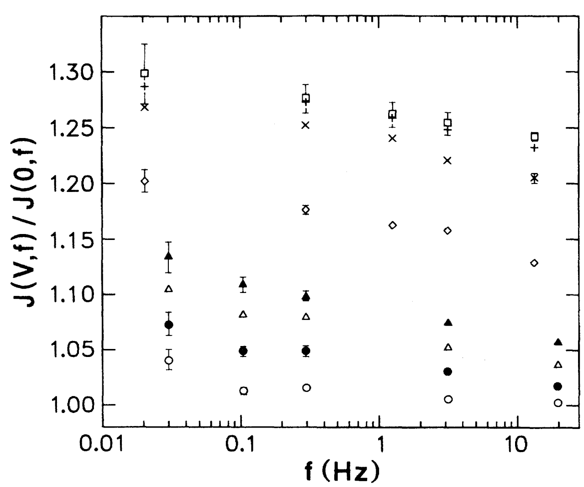

FIG. 2. Frequency dependence of the compliance, normalized to its $V=0$ value, at several voltages: open squares, $5.45V_T(J)$; plus signs, $4.18V_T(J)$; crosses, $3.09V_T(J)$; open diamonds, $2.00V_T(J)$; closed triangles, $1.51V_T(J)$; open triangles, $1.38V_T(J)$; closed circles, $1.25V_T(J)$; open circles, $1.13V_T(J)$.

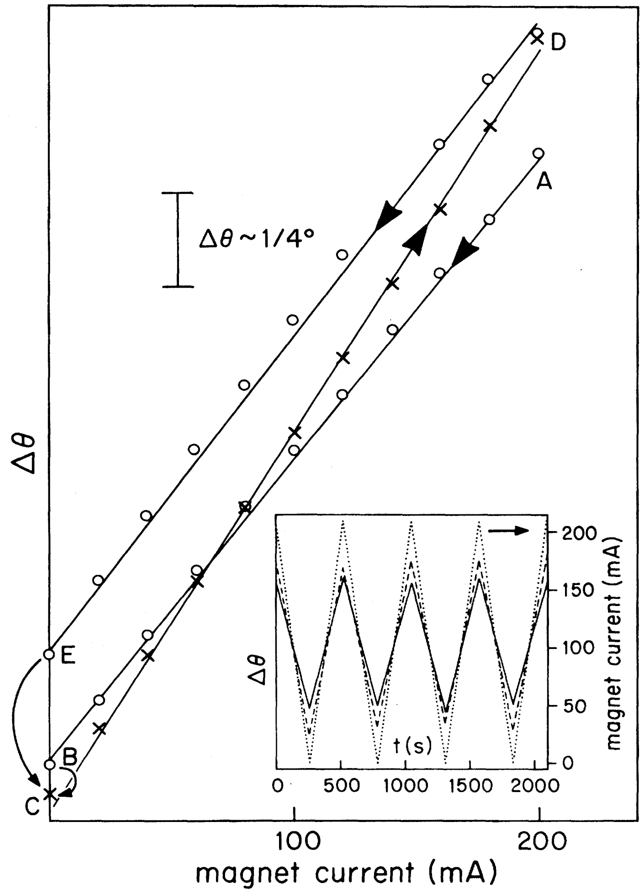

FIG. 3. Change in angle vs magnet current. CD was measured with $V=4V_T(J)$, and AB and DE were measured with $V=0.4V_T(J)$, as described in the text. The curved arrows BC and EC show the changes in angle when the CDW was depinned at zero applied torque (proportional to the magnet current). Inset: Magnet current (dotted curve) and change in angle [solid curve, $V=0.4V_T(J)$; dashed curve, $V=4V_T(J)$] vs time.

We can go to lower frequency by quasistatic measurements using an FM drive. In the inset to Fig. 3 are shown the changes in angle as the magnetic current (i.e., the torque) is slowly varied (with a period of 525 s). The slope $d\theta/dI_{MAGNET}$ is $\approx 25\%$ larger when the CDW is depinned $[V=4V_T(J)]$ than when the CDW is pinned $[V=0.4V_T(J)]$, implying that the pinned relaxation time is greater than $1/\omega=84$ s.

It is interesting to look at these strain vs stress curves in more detail, as shown in Fig. 3. At point A, the CDW is pinned and torque (0.2 A) is applied. As the applied torque is decreased to zero, the angle changes linearly with torque to point B. At this time, the CDW was depinned and there was a small jump in angle (to C), perhaps due to a background magnetic field. When the torque was reapplied, however, the angle changed (along CD) with $\approx$25% greater slope than for AB. When we reached the original torque, the CDW was repinned, but there was *no significant change in angle*. When the torque was now removed, the angle change (along DE) with a slope equal to that of segment AB, so that when we reached zero applied torque, there was a large residual strain, proportional to the original torque. This strain did not decay in 1 h if the voltage was kept at $0.4V_T(J)$, but it disappeared (i.e., the angle returned to point C) when the CDW was depinned [at $4V_T(J)$]. The angle stayed at C if the CDW was then repinned. The sample therefore exhibits "current-reversible strain memory."

One can go directly from point A to point D by depinning the CDW at fixed torque (0.2 A). However, the angle remained at D (for at least 1 h) when the CDW was repinned. That is, if the stress is not changed, one does not observe a significant change in strain with voltage once the CDW is depinned. If there is a similar history dependence of longitudinal strains, this might explain why no depinning anomalies were observed in the static Young's modulus experiments, because these were done by *fixing the stress* and varying the sample voltage.[6,13]

For voltages much above or much below threshold, these quasistatic results can be described by $\epsilon = J_0\sigma + \delta J \sigma_{\text{HIST}}$, where the history-dependent stress offset σ_{HIST} equals the stress when the sample was *last above threshold* (i.e., the present stress when $V \gg V_T$); that is, when above threshold the elastic response is as if from two springs *in series*. When the CDW becomes pinned, the spring governing the anomalous response (δJ) becomes latched, with its strain frozen at $\delta J \sigma_{\text{HIST}}$, and the dynamic response of the sample determined only by the J_0 spring. In fact, the strain hysteresis we observe shows the utility of viewing the anomalies in terms of compliances (series springs) rather than moduli (parallel springs), as was usually done previously.[2] Relaxation dynamics for $V > V_T$ (with a single relaxation time and relaxation strength $\delta J/J_0$) can be recovered by letting $d\sigma_{\text{HIST}}/dt = (\sigma - \sigma_{\text{HIST}})/\tau$.

Hysteresis of conductivity[14] and superlattice spot width[15] (i.e., domain size) with temperature and electric-field cycling are commonly observed in sliding CDW materials, and are associated with the many metastable configurations of phase-coherent domains.[1] History-dependent *longitudinal* strains were also observed in TaS_3 by Hoen *et al.*[16] In their work, the applied stress was small and constant, and strain hysteresis was observed when the direction of CDW sliding was reversed. In contrast, the residual strain we observe is approximately independent of the sign of the CDW current, and because it is proportional to σ_0 it can be much larger than that observed by Hoen *et al.* For example, the residual strain shown in Fig. 3 is $\approx 2\times10^{-5}$, about an order of magnitude greater than that observed by Hoen *et al.*, which we would have difficulty resolving. The explanation of the hysteretic strains they propose, involving expansion and compression of the CDW,[16] differs from the relaxational model of Mozurkewich,[8] which is probably more relevant to the effects we are observing.

In the Mozurkewich model,[8] the Lee-Rice domains are disequilibrated by changes in strain, due to the strain dependence of the Fermi wave vector. For torsional strain, the resulting (low-frequency) $\Delta G = [d\ \ln(k_F)/d\epsilon]^2 M_{\text{CDW}}$, where $M_{\text{CDW}} \approx 2$ GPa (Ref. 8) is the (longitudinal) modulus of the CDW (essentially the Bohm-Staver elastic constant of the condensed electrons). Since $G \approx 5$ GPa,[10] a 20% change in shear modulus implies $d\ \ln(k_F)/d\epsilon \approx 0.7$; e.g., a 1° change in interchain angle gives $\approx 1\%$ change in k_F. This is consistent with the fact[1,10] that, in TaS_3, k_F must depend on interchain coupling to have its observed value ($\approx c^*/8$).[3] However, in the Lee-Rice model,[1,7] the phase varies smoothly (i.e., there are no domain walls), so the history-dependent strain would similarily be distributed throughout the sample. In contrast, small, discrete, strandlike domains have been observed by electron microscopy.[17] Relaxation of these may result in large strains localized at their boundaries.

In conclusion, we have studied the dependence of torsional strains in TaS_3 as functions of stress, stress frequency, and voltage. The pinned relaxation time is >84 s, so that for any frequency we measure, the depinned compliance is $\approx$25% larger than the pinned. We have shown that the compliance has a logarithmic frequency dependence (for $0.03 < f < 20$ Hz) at all voltages above threshold, implying that the distribution of relaxation times is always wide, with $\tau_{\text{MAX}} > 5$ s. If the CDW is repinned when the crystal is strained, that strain will be locked into the crystal until the CDW is again depinned; there is current-reversible strain memory.

We appreciate very helpful conversations with M. E. Itkis, M. J. Skove, and T. M. Tritt. Samples were provided by R. E. Thorne. This work was supported in part by NSF Grant No. DMR-9300507.

[1] G. Grüner, Rev. Mod. Phys. **60**, 1129 (1989); **66**, 1 (1994).
[2] For a review, see George Mozurkewich, in *Perspectives in Physical Acoustics*, edited by F. Yu, R. K. Sundfors, and P. Suntharothok (World Scientific, Singapore, 1992), pp. 237–264.
[3] T. Sambongi, K. Tsutsumi, Y. Shiozaki, Y. Yamamoto, K. Yamaya, and Y. Abe, Solid State Commun. **22**, 729 (1977).
[4] Z. G. Xu and J. W. Brill, Phys. Rev. B **45**, 3953 (1992).
[5] X.-D. Xiang and J. W. Brill, Phys. Rev. Lett. **63**, 1853 (1989).
[6] (a) T. M. Tritt, M. J. Skove, and A. C. Ehrlich, Phys. Rev. B **43**, 9972 (1991); (b) D. Maclean, A. Simpson, and M. H. Jericho, *ibid.* **46**, 12 117 (1992).
[7] P. A. Lee and T. M. Rice, Phys. Rev. B **19**, 3970 (1979).
[8] George Mozurkewich, Phys. Rev. B **42**, 1183 (1990).
[9] Ronald L. Jacobsen, M. B. Weissman, and George Mozurkewich, Phys. Rev. B **43**, 13 198 (1991).
[10] X.-D. Xiang and J. W. Brill, Phys. Rev. B **36**, 2969 (1987).
[11] X.-D. Xiang, J. W. Brill, and W. L. Fuqua, Rev. Sci. Instrum. **60**, 3635 (1989).
[12] M. H. Jericho and A. M. Simpson, Phys. Rev. B **34**, 1116 (1986).
[13] However, no hysteresis was observed by the authors of Ref. 6(a): M. J. Skove and T. M. Tritt (private communications).
[14] A. W. Higgs and J. C. Gill, Solid State Commun. **47**, 737 (1983); G. Mihaly, G. Kriza, and A. Janossy, Phys. Rev. B **30**, 3578 (1984).

[15]T. Tamegai, K. Tsutsumi, S. Kagoshima, M. Sato, K. Tsuji, J. Harada, M. Sakata, and T. Nakajima, Solid State Commun. **51**, 585 (1984); Laszlo Mihaly, Ki-Bong Lee, and Peter W. Stephens, Phys. Rev. B **36**, 1793 (1987); J. Zhang, J. F. Ma, S. E. Nagler, and S. E. Brown, *ibid.* **47**, 1655 (1993).

[16]S. Hoen, B. Burk, A. Zettl, and M. Inui, Phys. Rev. B **46**, 1874 (1992).

[17]C. H. Chen and R. M. Fleming, Solid State Commun. **48,** 777 (1983).