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# Low-temperature shear compliance of $TaS_3$

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We have measured the low-frequency (13 Hz) and quasistatic shear compliance $(J)$ of orthorhombic $TaS_3$ as a function of applied electric field $(E)$ at 4.2 K. In contrast to liquid-nitrogen temperatures, where $J$ increases by 25–30 % when the electric field depins the charge-density wave, at 4.2 K the compliance is field independent for $E$ up to 270 $E_{NL}$, the field where nonlinear current is first observed, and well into the regime where the conductance is proportional to $E^2$. A few possible reasons for the lack of an elastic anomaly at low temperature are suggested. [S0163-1829(96)02643-4]

$TaS_3$ is one of a handful of quasi-one-dimensional, charge-density-wave (CDW) materials in which the CDW can be depinned by a small electric field, resulting in a number of unusual properties.[1] Among the most poorly understood properties of depinned CDW's are (i) the changes in elastic properties that accompany depinning[2] and (ii) the properties at low temperature, where quasiparticles freeze out and CDW deformations cannot be screened.[3–9] In this paper, we report on measurements of the shear compliance $(J)$ of orthorhombic $TaS_3$ at 4.2 K. We find that, unlike at higher temperature ($\approx$100 K) where $J$ increases by $\approx$25% with depinning,[10,11] $J$ is independent of electric field ($|\Delta J/J| < 2\%$) at low temperature. However, it is not clear if we have bulk depinning of the CDW at the highest fields we reach.

Orthorhombic $TaS_3$ ($o$-$TaS_3$) has a metal-semiconductor transition due to the condensation of a CDW at $T_c = 220$ K.[12] Below $T_c$, application of an electric field greater than a small threshold $E_T$ leads to striking non-Ohmic conductance associated with the depinning and motion of the CDW.[1] At high temperatures, the onset of non-Ohmic current is ''gradual''; e.g., at 100 K, the conductance $(1/R)$ typically doubles at $E = 2E_T$.[13] Since at low temperatures CDW deformations cannot be screened by thermally excited quasiparticles, the field dependence of the conductance becomes extremely abrupt [$1/R \propto (E-E_T)^p$, with $p \approx 15$, above $E_T$],[4] reflecting the lack of quasiparticle damping, and $E_T$ grows exponentially with $1/T$. However, smaller, ''gradual'' nonlinear current is also observed at low temperatures above a much smaller threshold, $E_{NL}$; the nature of this conduction is not well understood,[4–6] as discussed below.

Because of the fibrous morphology of $o$-$TaS_3$ samples (crystals are typically 1 cm$\times$10 $\mu$m$\times$10 $\mu$m in size, with highest conductivity in the long direction[14]), most elastic measurements[2,10,15] have been done by measuring flexural or torsional resonances of the sample. The large increases in elastic compliances [25% for $J$ and 2% for $s_{33}$ (the inverse Young's modulus)][10,15] near 100 K have been modeled in terms of relaxational dynamics; e.g., $J(E,\omega)/J_0 - 1 = \int A(\tau,E)\,d\tau/(1+\omega^2\tau^2)$, where $J_0$ is the unrelaxed compliance and the average relaxation time is assumed to decrease with increasing field above threshold.[15,16] Mozurkewich has shown that the relaxation may be associated with changes in the configuration of phase coherent CDW domains driven by a strain-dependent CDW wave vector $Q$. In this case, the total relaxation strength of the shear compliance is given by $\int A(\tau,E)\,d\tau = J_0 M_{CDW}(d\ln Q/d\epsilon)^2$, where $\epsilon$ is the torsional strain and $M_{CDW}$ the (longitudinal) modulus of the CDW (essentially the Bohm-Staver modulus of the condensed electrons).[16]

Because the frequencies of the mechanical resonances are usually low (<10 kHz) and easily damped by a surrounding medium, it has been difficult to investigate elastic changes at low temperatures, where the conductance is generally studied using short pulses and/or immersion in liquid helium to minimize Joule heating. For example, small changes in the flexural-resonant frequency of the related compound $K_{0.3}MoO_3$ were observed with CDW depinning at 4.2 K, but the authors were not certain that these changes were not due to sample heating.[17]

We recently reported[11] direct measurements of torsional strain (i.e., twist angle $\theta$) as a function of stress (i.e., torque) in $o$-$TaS_3$ at 100 K. A magnetic wire was glued to the center of the sample to make a torsional pendulum, with the sample as elastic element, which was placed inside an rf ($\approx$400 MHz) cavity. Applied torque was proportional to a magnetic field provided by Helmholtz coils surrounding the cavity, and $\theta$ was found by measuring the modulated output of the cavity. The responses to both oscillating (<20 Hz) and ''static'' torques were measured. For the former, the magnitude of the oscillating twist angle $\theta_{ac}$ is proportional to the shear compliance. For constant torques, it was found that strains could be frozen into the sample when the CDW was pinned, but for a fixed applied electric field, the ''quasistatic'' compliance is proportional to the slope of $\theta$ vs magnet current.

Because these measurements are subresonance and damping by the environment inconsequential, it is possible to do them with the sample immersed in a liquid-helium bath, and we report on the results of this investigation here. Two crystals were investigated. Sample 1 was $\approx$4 mm long and $\approx$150 $\mu m^2$ in cross-sectional area. Sample 2 was $\approx$2 mm$\times$12 $\mu m^2$. Silver paint was applied to copper films evaporated on the ends for electrical (and mechanical) contacts. dc currents were applied to the sample and voltages (at the current contacts) measured with an electrometer.

In Fig. 1, we show the current and resistance of the sample vs applied electric field for sample 1. The inset shows the current at low fields with the onset of non-Ohmicity at

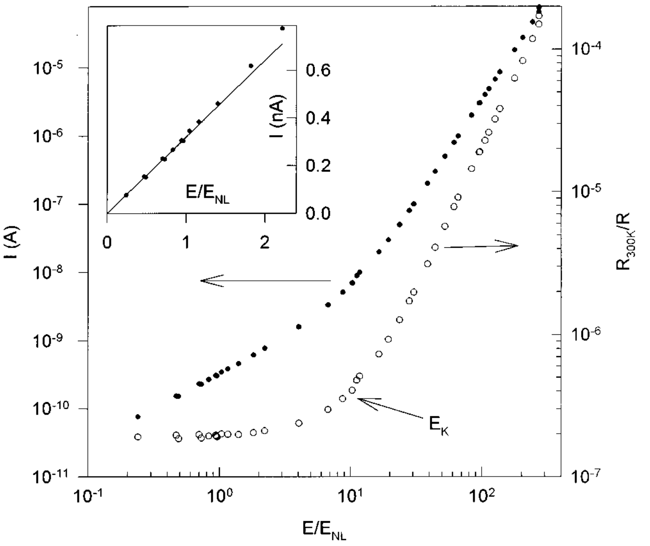

FIG. 1. Current and resistance, normalized to its room-temperature value ($R_{300\ K}=60\ \Omega$), vs electric field, normalized to the onset field for nonlinear response $E_{NL}\approx 270$ mV/cm, for sample 1 at 4.2 K. Above $E_K$, $1/R \propto E^2$. The inset shows the field dependence of the current at low fields in a linear plot.

$E_{NL}\approx 0.27$ V/cm. For currents above 70 $\mu$A (where $E/E_{NL}=270$), the voltage becomes noisy, indicating that significant Joule heating ($>1$ K) of the sample is occurring. Very similar results (with noticeable Joule heating at $E/E_{NL}>125$) were obtained for sample 2.

The field dependence of the resistance of our samples differs somewhat from that reported by Itkis, Nad', and Monceau for a sample with a similar value of $R_{300\ K}/R$. (Note that smaller values of low-temperature conductivity are associated with less pure[18] and/or much smaller[6] samples.) The onset of non-Ohmicity at $E_{NL}$ is much more gradual for our samples, but the conductance starts increasing rapidly ($1/R \propto E^2$) at the knee field $E_K \approx 10 E_{NL}$, whereas $E_K \approx 100 E_{NL}$ in Ref. 4. Despite the immersion in liquid helium, we cannot reach $E_T$ ($=3000\ E_{NL}$ in Ref. 4), where the conductance grows even more rapidly. However, at our highest field, the conductance is $>1000$ times its low-field value and 50% greater than the ratio reported at $E_T$ in Ref. 4. Therefore, while we do not achieve the "rigid" CDW motion ($-d\ \ln R/d\ \ln E \gg 1$) associated[4,5] with $E>E_T$, we do have a huge non-Ohmic current. (In fact, our maximum current is an order of magnitude larger than the CDW current at $3E_T$ at 80 K.) A power-law variation of conductance with $E$, such as we observe above $E_K$, has been associated with bulk CDW motion in other materials at higher temperatures,[7,8] and we feel that it is likely that there is significant, although incoherent, CDW sliding for $E>E_K$.

The field dependence of $\theta_{ac}$ (at 13.3 Hz) for sample 1 at 4.2 and 80 K is shown in Fig. 2. For these measurements, $\theta_{ac}\approx 0.002°$, corresponding to a strain $\approx 10^{-7}$. At 80 K, $J \propto \theta_{ac}$ increases by $\approx 30\%$ with CDW depinning, similar to the behavior at 100 K previously reported.[11] However, at 4.2 K, no change in $\theta_{ac}$ ($\pm 1\%$) with field up to the highest field possible was observed; similarly, no change in $\theta_{ac}$ was observed for sample 2 at 4.2 K.

Results of measurements of twist angle vs time at 80 and 4.2 K at a few electric fields for sample 1 are shown in Fig. 3. For these measurements, the magnet current was linearly ramped with time, as shown; the total change in $\Delta\theta \approx 0.04°$ ($\Delta\epsilon \approx 2\times 10^{-6}$). At 80 K, $J$ (which is proportional to the slope) is $\approx 25\%$ greater at $E=4.6\ E_T$ than at $E=0$, consistent with the ac results of Fig. 2 and previous results at 100 K.[11] At 4.2 K, the same slope ($\pm 2\%$) was observed at all fields checked. Similarly, no electric-field dependence of slope was observed for sample 2.

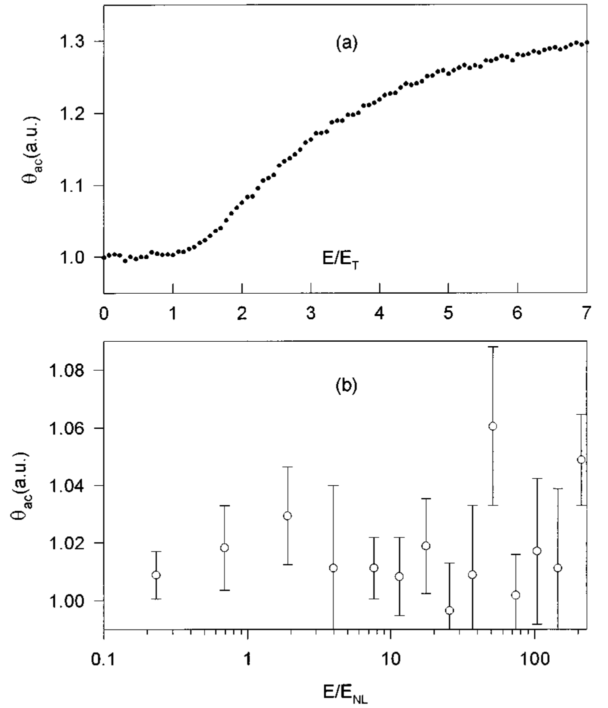

FIG. 2. The electric-field dependence of the magnitude of the oscillating-twist angle, normalized to its low-field value at 13.3 Hz at (a) 80 K and (b) 4.2 K, for sample 1. The threshold field at 80 K is $E_T \approx 350$ mV/cm; at 4.2 K, $E_{NL}\approx 270$ mV/cm.

We therefore conclude that at 4.2 K the shear compliance is independent of field for fields up to $270 E_{NL}$ (and total current $=900$ times the Ohmic current); the compliance certainly does not have the large field dependence observed near 100 K. If the domain relaxation model is correct, this implies that either the relaxation strength has vanished or that the average relaxation time has become field independent. A few possibilities will be discussed.

If the average relaxation time is field independent in this regime, it presumably means that it remains long even for $E>E_K$ (i.e., $\tau_{ave}>$period$/2\pi=70$ s). If the CDW is moving throughout the sample at our largest voltages, then our results imply that, unlike at higher temperature, CDW motion does not allow the phase domains to readjust to the strain in a time $<70$ s, presumably because the lack of quasiparticle screening prevents the local deformations needed to readjust.

On the other hand, the *average* relaxation time may be large at all fields we investigated because the non-Ohmic conductivity below $E_T$ may involve inhomogenous CDW motion in a small fraction of the sample. The anomaly in shear compliance is $<2\%$, as compared to $\Delta J/J_0 \approx 30\%$ at 80 K, suggesting that the CDW is moving in $<7\%$ of the sample at any instant. Such might occur if the sliding CDW percolates between small, weakly pinned regions. A similar

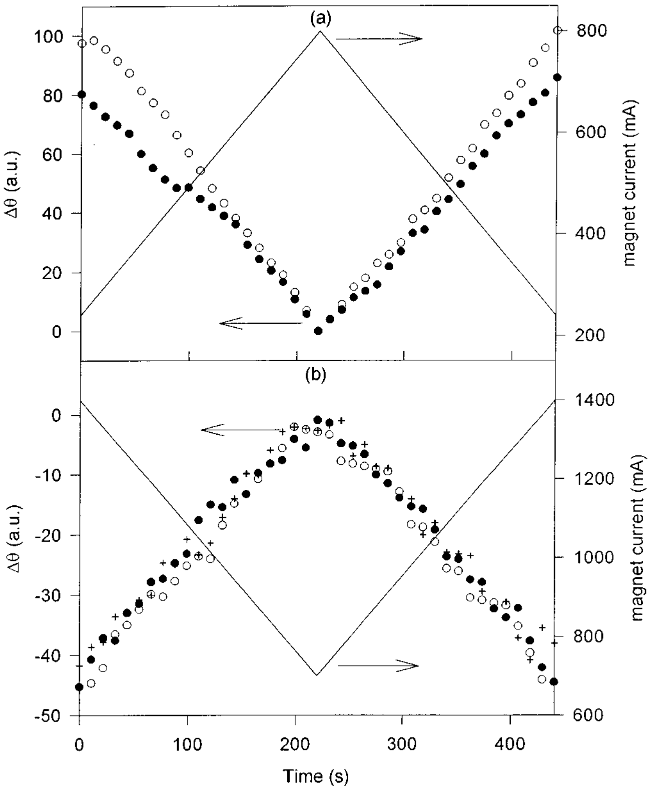

FIG. 3. Changes in the twist angles vs time for sample 1. (a) 80 K (solid circles, $E=0$; open circles, $E=4.6\ E_T$); (b) 4.2 K (solid circles, $E=0$; open circles, $E=95\ E_{\mathrm{NL}}$; pluses, $E=270\ E_{\mathrm{NL}}$). The solid lines show the corresponding currents in the magnet (3.3 mT/A) providing the torque on the sample.

CDW ''creep''[6] mechanism, yielding the power-law dependence of the conductivity on field mentioned above, was suggested for monoclinic $TaS_3$ at $E>E_T$.[8] Alternatively, the current may be limited to a thin-surface layer between the current contacts. Such is often the case for very anisotropic materials,[9,19] although Nad' and Monceau,[8] on the basis of an analysis of the narrow-band noise amplitude, suggest that such does not occur for closely related monoclinic $TaS_3$.

Itkis, Nad', and Monceau[4,5] have suggested that fields $E<E_T$ are inadequate to drive the phase-slip processes[20] needed for bulk motion of the CDW. Instead they suggest that the non-Ohmic conduction at these fields may be due to hopping of carriers (possibly charged dislocation lines[5]) between localized CDW defects, such as soliton states. Such hopping is not expected to have a significant effect in allowing CDW domains to relax.

The most interesting possible reason for the lack of a compliance anomaly is that the relaxation strength vanishes because $d\ \ln Q/d\epsilon$ vanishes at low temperature. Such would be consistent with the lack of thermal hysteresis in resistance at low temperature,[21] since metastability is associated with a CDW wave vector which changes with external parameters. For example, $Q$ may become locked into its close commensurate value ($Q=c^*/4$) (Ref. 22) at low temperature. There has been much speculation but little firm evidence[22] about such a low-temperature lock-in in $o$-$TaS_3$.

In conclusion, we have shown that any changes in the shear compliance of $o$-$TaS_3$ at 4.2 K at fields almost 300 times the non-Ohmic threshold (and ≈30 times the ''knee'' field above which $1/R\propto E^2$) are at least an order of magnitude smaller than the changes observed at $5E_T$ near 100 K. Possible reasons are that (i) the domain relaxation remains very slow due to the lack of quasiparticle screening even though there is CDW motion thoughout the bulk of the sample; (ii) the CDW is only moving in a small fraction of the sample; (iii) the non-Ohmic conduction in this field range is associated with carrier hopping between defects rather than CDW motion; and (iv) the CDW wave vector is independent of strain, e.g., due to a commensurate lock-in.

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