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Change in Young's modulus at low frequency upon charge-density-wave depinning in TaS_3

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We have measured statically (at frequency $\omega < 0.1$ Hz) the stress-strain (σ-ε) relationship of the charge-density-wave (CDW) material TaS_3, while simultaneously monitoring the four-probe resistance. We found that an electric field E up to 2–3 times larger than the threshold field E_T for CDW motion had at most a small effect ($0 < \Delta\varepsilon/\varepsilon < 0.2\%$) on the strain at constant stress ($\Delta\varepsilon/\varepsilon \approx -\Delta Y/Y$ at constant σ), in contrast to vibrating-reed determinations of $\Delta Y/Y$. For $E > 3E_T$ we observed a change in the strain, i.e., the sample length, which was linear in the power; this was suggestive of sample heating. We discuss our results in relation to the recently discovered frequency dependence of Y [X. D. Xiang and J. W. Brill, Phys. Rev. Lett. **63**, 1853 (1990)] and the recent phase-relaxation model of elastic softening [G. Mozurkewich, Phys. Rev. B **42**, 11 183 (1990)].

Recently there has been interest, both experimental[1] and theoretical,[2] in the frequency dependence of the elastic properties of charge-density-wave (CDW) conductors. Xiang and Brill[1] reported measurements of the frequency dependence of the change in Young's modulus $\Delta Y/Y$ upon depinning the CDW in $NbSe_3$ and in TaS_3. They modified the vibrating-reed (VR) technique using a helical-resonator detector to provide increased sensitivity.[3] The standard VR method[4–8] uses the sample as part of a capacitor in a phase-locked loop to measure the flexural resonant frequency ($\omega_n \approx A_n\sqrt{Y}$) of a cantilevered beam (the sample). Thus, the resonant frequency yields Y and the sharpness of the resonance yields the internal friction ($\sim 1/Q$), where Q is the quality factor. This method is excellent for measuring changes in Y, since $\Delta Y/Y \approx \Delta\omega_n/\omega_n$ and ω can be measured quite precisely. The modified VR method of Xiang and Brill enables them to observe the higher harmonics associated with Y, thus increasing the frequency range over which $\Delta Y/Y$ can be measured. For TaS_3 they found $|\Delta Y/Y|_{\text{depin}}$ to be proportional to ω^{-p}, with $\frac{1}{2} < p < \frac{3}{4}$ when ω is in the range 300 Hz to 100 kHz;

$$|\Delta Y/Y|_{\text{depin}} = [Y(E=0) - Y(E=4E_T)]/Y(0)$$

and E_T is the threshold electric field for CDW motion. At $\omega = 300$ Hz, they found $|\Delta Y/Y|_{\text{depin}}$ to be approximately 2%. At higher frequencies ($\omega \approx 10$ MHz), Jerico and Simpson[9] found the elasticity changes to be much smaller, by orders of magnitude, compared to the low-kHz frequencies. This all supports a decreasing $\Delta Y/Y$ with increasing frequency.

The electric-field dependence of the above elastic properties of TaS_3 were discovered independently by Brill and co-workers[10,11] and Mozurkewich *et al.*[12,13] They found that at E_T, there is a sharp reduction in Y, and Y continues to decrease as E is increased with an overall decrease in Y of approximately 2% for $E = 4E_T$. They also found that the internal friction increases by a comparable fraction. Later, Brill showed that the shear modulus G was also a function of an electric field for $E > E_T$, with a much larger change than Y ($\Delta G/G \approx 20\%$).[14] Similar, but smaller effects of the elastic properties have been observed in $NbSe_3$.[15] Jacobsen and Mozurkewich[16] have recently shown that Y_{CDW}, the modulus difference between the stationary and rapidly sliding CDW states, increases as the square of the order parameter just below T_p and that the shape of the $\Delta Y/Y$-vs-E curve is controlled predominantly by the CDW current I_{CDW}.

Initially, the elastic softening for $E > E_T$ was ascribed to a decoupling, when the CDW slides, of a modulus associated with the CDW and the modulus of the lattice.[12,17–20] Mozurkewich *et al.* modeled the CDW and the lattice with balls and springs. They suggested that the lattice and the CDW were coupled through impurity pinning of the CDW, and assumed the coupling vanishes when the CDW slides. Maki and Virosztek's microscopic model[21,22] suggests that the coupling between the lattice and the CDW is due to an electron-phonon interaction. They suggest that the elastic softening for $E > E_T$ is due to changes in the ability of the CDW to screen acoustic phonons and could be dependent on the impurity density. Jacobsen and Mozurkewich[23] recently reported an experiment in which they varied the pinning of the CDW by electron irradiation, and found that the reduction in modulus on depinning was not sensitive to the pin density.

Brill and co-workers[11,24] proposed a model in which the softening of the modulus was due to the relaxation of an unspecified defect in the CDW. They proposed that the relaxation times τ_f and τ_s of the fixed and sliding states differ, with $\tau_f \gg \tau_s$. If the modulus is measured in a time t such that $\tau_s < t < \tau_f$, then the modulus will be reduced in the sliding state. Thus, the time it takes to make the measurement compared to the relaxation time of the measured quantity is relevant. Brill's model has recently been extended by Mozurkewich[2] where he ascribes the difference in elastic properties of the stationary (fixed) and sliding states to the difference in the thermodynamic potentials (and their derivatives, such as elastic constants) of equilibrium and metastable phase configurations of the CDW. He associated the "defect" of Brill's model with the Lee-Rice coherence domains. The relaxation times are the times for the phase configuration to relax to equilibrium in the fixed and sliding states. Mozurkewich finds that the difference between the CDW spring constant and amplitude in the metastable states and in the equilibrium state are insufficient to explain the observed softening of Young's modulus, but the effects of wavelength (or phase) differences on Young's modulus are of the right order of magnitude. The softening of the modulus in this theory is independent of the number of pins, which is in agreement with experiment.[23] Brill has expressed concern about relaxation models being appropriate descriptions of CDW depinning, since the shape of the elastic properties as a function of E should vary with frequency and he has shown they do not.[1]

Models which involve decoupling of the lattice and the CDW predict $|\Delta Y/Y|$ to be independent of frequency, in contrast to the observations of Xiang and Brill.[1] Maki's model does not address frequency dependence of the elastic softening.[25] Mozurkewich's phase-relaxation model predicts a frequency dependence, but if a single τ_s is assumed, the frequency dependence of $|\Delta Y/Y|$ should be limited to a small region around $\omega = 1/\tau_s$. If a distribution of τ_s's is assumed, a power law may be obtained. Such "glasslike" distributions of relaxation times have been observed in the resistance and dielectric constants of CDW materials.[26–28] Further, the Mozurkewich model predicts for $\omega \ll 1/\tau_s$ that $|\Delta Y/Y| \rightarrow 0$. Thus, a low-frequency maximum in the frequency dependence of $|\Delta Y/Y|$ is suggested by the Mozurkewich model and also by the experiment of Xiang and Brill, since $|\Delta Y/Y|$ should not be expected to keep growing as ω^{-p}. One of the very important questions then concerns the behavior of $|\Delta Y/Y|$ at the lower frequencies ($\omega \ll 300$ Hz), frequencies which may be inaccessible to the VR technique.

In this paper, we report a direct or static measurement of Young's modulus as a function of electric field using a stress-strain device we developed which operates at essentially zero frequency, and thus enables us to address this central question. It is also possible to operate this device at a few hertz using a modulation coil. We have found, in agreement with the predictions of Mozurkewich's model, that at a very low frequency (in our case essentially zero) there is much less change in Young's modulus above E_T than at a higher frequency: at least an order of magnitude less change in Y than that found at the higher VR frequencies ($\omega > 300$ Hz).

Our stress-strain device consists of an upper (movable) rod which is held by two leaf springs, so that the static friction is unmeasurably small. On this rod, two magnets apply a force to the rod proportional to the current in a surrounding solenoid. The magnets are centered with the ends of the coil. Also on the rod is one plate of a capacitor. The other plate is fixed, and the capacitance is inversely proportional to the distance of the plate on the rod from the fixed plate. One end of the sample is attached to the movable rod and the other is fixed to the body of the device. Thus, the stress on the sample is related to the current in the solenoid and the resulting strain related to the capacitance. With this, we can resolve a minimum motion of ≈ 10 nm. A more complete description of this device and the calibration methods are given elsewhere.[29]

The upper (movable) end of the sample is actually attached to two copper wires $(+I, +V)$ allowing electrical contacts to be made to the sample. These wires in turn are fixed to a flat aluminum mounting plate, extending below and secured to the movable rod. The other end of the sample is attached to stationary wires $(-I, -V)$, mounted on the main body of the device. du Pont silver paint (No. 5007) was used to make contact between the sample and the copper wires. Although the silver paint makes a reasonable mechanical contact as well as electrical contact, epoxy is placed between the current and potential contacts, after the sample is mounted, to inhibit the sample from slipping through the contacts.[30–32] The stress applied to the sample is uniaxial, except for a small region near the contacts that compensates for any possible misalignment of the sample. The length L_0 of the sample was taken to be the distance between the potential contacts, which also formed the mechanical end clamps. The area A of the sample was deduced from the room-temperature resistivity of TaS_3, $\rho(300\text{ K}) = 320\ \mu\Omega\,\text{cm}$,[33] the measured room-temperature resistance, and L_0.

Figure 1(a) shows the total force applied versus the displacement of the plates. When the sample becomes unbowed and gets tight, there is a very sharp change in slope. The initial slope is due to leaf springs and is the spring constant of the apparatus $k = 0.1013$ mN/μm. The force on the sample is given by $F_{\text{sam}} = F_T - k\Delta d$, where F_T is the force applied by the magnets and $k\Delta d$ is the force necessary to move the leaf springs. The elongation of the

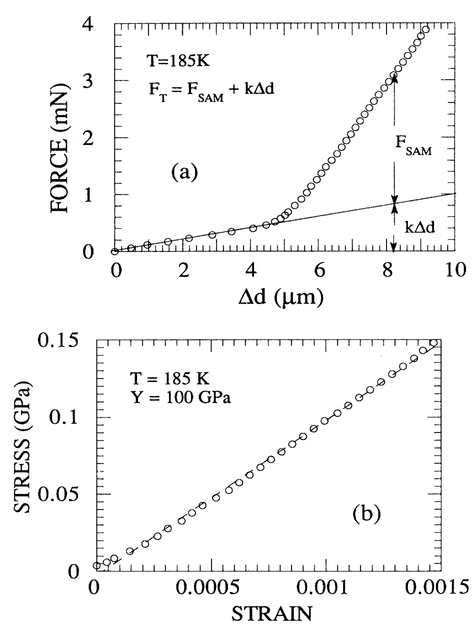

FIG. 1. (a) The total force applied vs the displacement of the rod. When the sample becomes tight there is a sharp change in slope in F_T vs Δd. The smaller slope is due to the springs and is essentially the spring constant of the apparatus ($k = 0.1013$ mN/μm). The force on the sample then is given by $F_{\text{sam}} = F_T - k\Delta d$. (b) The uniaxial stress applied to the sample vs the strain ($\Delta L/L_0$) of the sample at $T = 185$ K. The slope yields a value for Young's modulus of 100 ± 1 GPa. For this sample, $L_0 = 2.90$ mm, $R_S(300\text{ K}) = 463\ \Omega$, and $A = 20\ \mu\text{m}^2$.

sample ΔL is the displacement of the rod from the position at which the sample is just tight. A plot of the stress $\sigma = F_{sam}/A$ versus the strain $\varepsilon = \Delta L/L$, the slope of which is Y, for a TaS_3 sample at $T = 185$ K is shown in Fig. 1(b). The slope in Fig. 1(b) yields $Y = 100 \pm 1$ GPa. An average value for Y for many TaS_3 samples is 100 ± 20 GPa,[34] with the greatest source of uncertainty in the sample cross section. Continuous measurements of Young's modulus as a function of temperature are more difficult with this device than with the VR device due to the sensitivity of our device to differential thermal contractions as the temperature is changed. Our device operates best by stabilizing the temperature and then performing the stress-strain measurement.

We have measured the electric-field dependence of Y in TaS_3 below T_p and found changes much less than expected. Figure 2 is a plot of the normalized change in strain as a function of E at a constant stress of 300 MPa (with $\Delta\varepsilon/\varepsilon_0 \approx -\Delta Y/Y$ at constant stress) where ε_0 is the applied strain for $E \ll E_T$. As seen, there is not a sharp change in the strain at threshold. Only above $E \approx 2\text{–}3E_T$ is there a measurable change. Above $3E_T$, the normalized change in strain ($\approx \Delta L$) grows linearly with E, without indication of saturating, such that $\Delta\varepsilon/\varepsilon_0 \approx 0.7\%$ at $5E_T$. We have also simultaneously measured the resistance of TaS_3. The inset in Fig. 2 shows the resistance as a function of E at a strain $\varepsilon = 0.3\%$. These resistance measurements are in agreement with others.[30,31] A stress-induced incommensurate-commensurate transition around 0.4% strain has been reported in TaS_3.[30,31,35] We have applied stresses on either side of this transition strain and found no measurable effects on the data.

Figure 3 shows the change in length of the sample, at $\varepsilon_0 = 0.3\%$, versus the power into the sample. This shows a linear dependence of the sample length with power, suggestive of thermal expansion due to sample heating. We cannot measure the thermal expansion with our device because we do not know ΔT precisely. If we use the predicted value for the thermal expansion for TaS_3,[30] then an

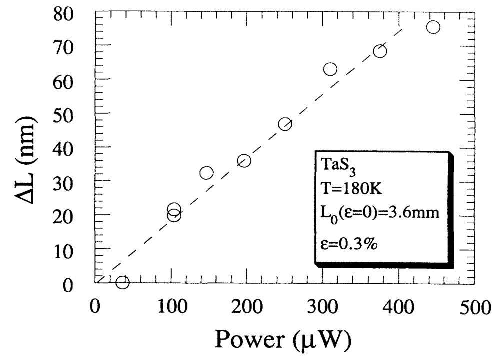

FIG. 3. The elongation of the sample vs the applied power at $T = 180$ K with $\varepsilon_0 = 0.3\%$. The conditions and sample characteristics are the same as for Fig. 2.

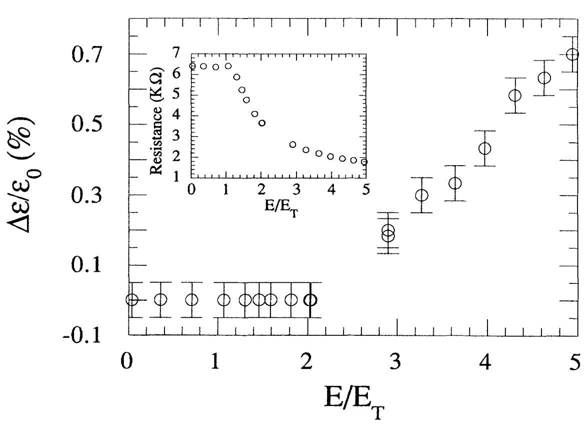

FIG. 2. The change in strain at a constant stress vs E/E_T at $T = 180$ K. The change in strain is normalized to the $E < E_T$ strain ε_0. The applied stress is 300 MPa, which corresponds to a strain $\varepsilon_0 = 0.3\%$. For this sample, $L_0 = 3.6$ mm, $E_T = 500$ mV/cm, and $A \approx 13\ \mu m^2$. The inset shows the corresponding resistance as a function of E/E_T.

overall change in sample length of 75 nm in a sample of length 3.6 mm would correspond to an equivalent temperature change of only 5 K of the sample. Thus, it is not unreasonable that a significant part of the change we observe at high E might be due to thermal expansion from sample heating. Nevertheless, even if none of the change in strain (and thus in Y) is due to heating, $\Delta Y/Y$ is only 0.7% at $5E_T$, which is much less than expected from an extrapolation of the higher-frequency data.

At lower temperatures ($T = 130$ K) and with $\sigma = 75$ MPa (causing ε to be 7×10^{-4}), an electric field $E = 3E_T$ was found to change ε by less than 1×10^{-6}. Thus, at this σ, Y changes less than about 1.5 parts in 10^3 (0.15%) when the CDW is depinned. The lowest limit we achieved at $|\Delta Y/Y| < 0.05\%$ at $E \approx 2E_T$. All these values are smaller than found by Xiang and Brill at frequencies below 3 kHz in either of their reported samples. Thus, we have an upper limit of $|\Delta Y/Y| \leq 0.2\%$ for $E \leq 3E_T$, with no measurable change at E_T, and sample heating very possibly contributing to such changes as we do see at higher E.

The results of our experiments give a direct measure of Y, but do not have the sensitivity of the VR method. The two methods are not strictly comparable in that we apply a large spatially and temporally uniform, uniaxial tension, whereas the VR method applies a small spatially and temporally varying tension. It is not apparent, however, how or why this difference would account for the contrast in the results, especially given the frequency dependence of the VR results. To address this as well as possible, we have measured $|\Delta Y/Y|$ at a small stress of 20 MPa causing ε to be 0.02%. Our resolution for $|\Delta Y/Y|$ at this small stress is only about 1% because the resulting strain in the sample is also small and the uncertainty in determining $\Delta\varepsilon$ is thus more of a factor. We find $0 < |\Delta Y/Y| < 1\%$ at $E = 3E_T$. We have also performed an experiment using an oscillating σ ($\omega \approx 6$ Hz) in addition to the dc stress experiment. This produces an oscillating strain with an amplitude proportional to Y. The change in this amplitude when $E = 3E_T$ was $0 < |\Delta Y/Y| < 1\%$ at $\sigma \approx 20$ MPa. Thus, within experimental error, $\Delta Y/Y \approx 0$ is again consistent with the data. On the other hand, whereas a 1% change in Y would be large, it is still less than that found at $4E_T$ by Xiang and Brill at the fundamental frequencies

(300 Hz and 3 kHz) of their samples.

The phase-relaxation theory of Mozurkewich predicts $|\Delta Y/Y| \rightarrow 0$ as ω becomes much less than the lowest $\omega_s = 1/\tau_s$. Estimates of ω_s are around 1 kHz,[24] which is near the lowest frequency (≈ 300 Hz) attained by Xiang and Brill. Thus, while the CDW is sliding, it may be that the phase relaxes to its equilibrium value in a time $\tau_s \approx 1$ ms, but when the CDW is fixed to the lattice, the phase relaxes to its equilibrium value in a time $\tau_f \gtrsim 1$ s. Our experiments would then be in a domain of frequency in which neither relaxation process affects Y, and the VR experiments are in a region where Y is affected by the relaxation process associated with τ_s. If this were true, then there should be a maximum in $|\Delta Y/Y|$ in the frequency domain $10 < \omega < 300$ Hz. Verification of these ideas requires the VR measurements to be extended to lower frequency ($\omega \leq 300$ Hz), and that we extend our measurements to higher frequency ($\omega \leq 10$–15 Hz), in the hope of finding the maximum $|\Delta Y/Y|$, which both experiments suggest and Mozurkewich's model predicts should be present.

In summary, we have measured the static Young's modulus in TaS_3 directly using a stress-strain method which operates at essentially zero frequency. We applied a constant stress and measured the electric field dependence of $\Delta\varepsilon/\varepsilon_0$ ($\Delta\varepsilon/\varepsilon_0 \approx -\Delta Y/Y$ at constant σ). This method yielded no measurable change in Y with electric field to within a resolution of 0.2% up to electric fields of $3E_T$ and less than 0.7% at $5E_T$. The contributions at high E may be influenced by thermal expansion of the sample. Our results are in contrast with the frequency dependence found by Xiang and Brill, but complement the low-frequency behavior predicted from Mozurkewich's phase-relaxation theory.

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