Electric Field Dependence of Elastic Properties of TaS_3

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We have measured the electric field dependence of the Young's modulus and internal friction of orthorhombic TaS_3. The modulus is observed to decrease by a few percent when the field exceeds ϵ_T, the threshold for non-Ohmic conduction. At the same field, the internal friction increases rapidly, saturating at $\sim 1\%$ for fields $\epsilon > 2\epsilon_T$.

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Orthorhombic TaS_3 (o-TaS_3) undergoes a charge density wave (CDW) transition at 220 K. In common with a few CDW materials, it exhibits a number of unusual electron transport properties,[1] most prominently non-Ohmic conductivity,[2] when the electric field in the sample exceeds a threshold field, ϵ_T, which is sample and temperature dependent. A variety of physical models have had success in describing these phenemena in terms of CDW motion; i.e., the CDW is pinned to the lattice by defects for fields $\epsilon < \epsilon_T$ and becomes depinned for $\epsilon > \epsilon_T$.[1] It has recently been shown that at low temperatures (~ 100 K) there is a nonzero Peltier coefficient associated with the CDW current; apparently the depinned CDW is accompanied by appreciable thermal phonon drag.[3] However, there has previously been no direct evidence that lattice properties are influenced by the CDW motion, and none of the models of CDW depinning have addressed the question of its effect on the lattice. In this paper, we report on the first measurements of the electric field dependence of elastic properties in o-TaS_3, by use of a modified vibrating-reed apparatus. We find that the Young's modulus rapidly decreases by a few percent as the field exceeds threshold and the CDW becomes depinned, while the internal friction increases by an order of magnitude and saturates at $\epsilon \sim 2\epsilon_T$ to show that the lattice is strongly affected by the CDW depinning.

The vibrating-reed method[4] of measuring the Young's modulus and internal friction of a small crystalline fiber is described in detail elsewhere[5] and the temperature dependence of these properties for o-TaS_3 (along the highly conducting, fiber axis) are also given. In order to apply also a voltage to the sample, both ends are rigidly clamped; the clamps act as current and voltage leads. The resonant frequency of the fundamental flexural resonance of a reed of length l, thickness t, Young's modulus E, and density ρ which is rigidly clamped at both ends is given by[6]

$$f_0 = (a_0 t/l^2)(E/\rho)^{1/2}, \tag{1}$$

where $a_0 = 1.03$.

Rigidly clamping an o-TaS_3 single crystal (typical dimensions: $5 \times 0.02 \times 0.005$ mm) is complicated by the fact that negligible uniaxial stress should be applied, even at low temperatures (i.e., thermal stresses must be minimized). Under the application of uniaxial stress, σ, Eq. (1) is modified to[6,7]

$$f_0 = (\gamma a_0 t/l^2)\{[E + (0.54/\gamma)(l/t)^2\sigma]/\rho\}^{1/2}, \tag{2}$$

where γ depends on σ/E but is of order unity. [$\gamma = 0.45$ for $\sigma(l/t)^2 >> E$.] Because the factor $(l/t)^2 \sim 10^6$, small stresses will greatly increase the resonant frequency. We found that for samples mounted by gluing (with silver paint) each end to a rigid support, differential thermal contraction led to large stresses at low temperatures. We therefore glued one end to a 0.05-mm Constantan wire, which was in turn glued to a stationary rod. The new resonant frequency was generally found to be somewhat ($\sim 30\%$) less than expected from Eq. (1), indicating that the Constantan wire was also vibrating slightly; i.e., the effective modulus determined from Eq. (1) included a contribution from the wire. Consequently the temperature dependence of the effective modulus differed from that reported in Ref. 5, although it was qualitatively similar, having a $\sim 2\%$ dip at 220 K. Unfortunately, more rigid mounts invariably introduced excessive stress at low temperature.

Data were taken by stabilizing temperature and varying the dc current through the sample while monitoring the dc voltage (i.e., a two-probe resistance measurement was done), the amplitude of the resonant signal A, and the resonant frequency f_0. The latter two were done by incorporating the sample in a phase-lock loop[5]; therefore, frequency shifts much less than the bandwidth could be resolved. (Typical resolutions were $\Delta E/E_0 = 2\Delta f/f_0 = 6 \times 10^{-5}$.) Changes in the internal friction, δ, are given by changes in the quality factor and, hence, the amplitude: $\Delta\delta = \Delta(1/Q) = (A_0/Q_0) \times \Delta(1/A)$; Q_0 was determined by measuring the bandwidth at zero current. Because the resonant

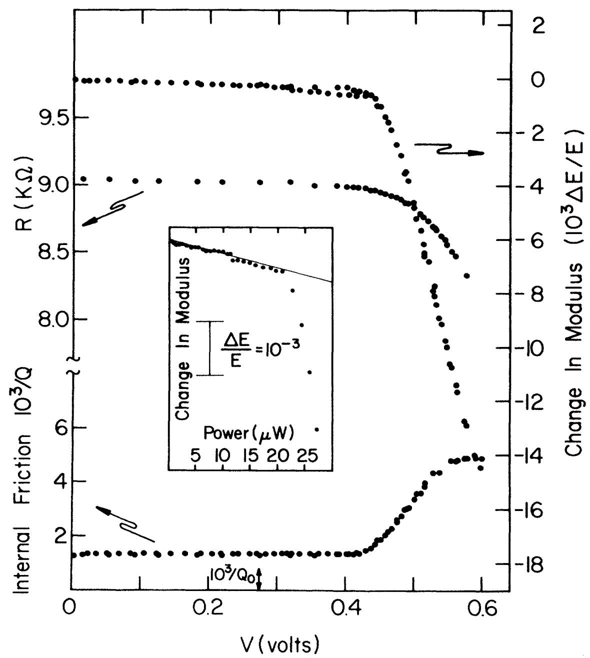

FIG. 1. Resistance, 1/(quality factor), and relative change in modulus $\Delta E/E_0 = 2\Delta f/f_0$ vs voltage at 149 K. Q_0 is the quality factor in vacuum at zero voltage. Inset: $\Delta E/E_0$ vs power dissipated in sample at 149 K. The line shows the proportionality below threshold.

frequency is extremely temperature dependent, excellent temperature equilibration ($\Delta T < 30$ mK) was required; this was facilitated by keeping 0.2 Torr of He gas in the sample chamber, which also greatly reduced the Joule heating of the sample, discussed below. This atmosphere added to the damping of the resonance; we verified that these losses simply added to the current dependent losses, as expected. Typically, our quality factors were 700 in 0.2 Torr and 1200 in vacuum, the latter probably chiefly due to friction at the clamps.

Typical results are shown in Fig. 1. It is seen that near the threshold voltage V_T where the resistance starts decreasing, the internal friction begins increasing and the frequency decreasing sharply. There is a smaller decrease in frequency for $V < V_T$; as shown in the inset, this decrease is proportional to the Joule heat dissipated in the sample and, therefore, the temperature change in the sample. We therefore have subtracted out that part of the modulus change due to heating in our subsequent plots:

$$\Delta E(V)/E_0|_T = 2\Delta f/f - mP(V), \tag{3}$$

where $P(V)$ is the power dissipated and m is the slope experimentally determined from the $V < V_T$ data at each temperature. Typically, $m \sim -3 \times 10^{-5}/\mu$W.

We observed that the resonant frequency at zero voltage changed by $\sim 0.1\%$ after a voltage greater than threshold was applied; an analogous effect occurs in the resistance.[8] Therefore, data were taken by first applying a large "conditioning" current before taking measurements at currents all less than the conditioning current. Results similar to those shown in Fig. 1 were observed for all six samples measured, including samples prepared at the University of California at Los Angeles (UCLA) and the University of Kentucky. All data reported on here are on a single (UCLA) sample of length 7.5 mm and resonant frequency 1300 Hz. Results from several runs at different temperatures are shown in Fig. 2. The resonant frequency and quality factor clearly depend on the CDW depinning; i.e., they change for $V > V_T$. [At each temperature, the threshold voltage for the elastic changes agreed with that for resistance change within the precision (20 mV) of the latter.]

After completing these measurements, the sample was remounted with both ends glued to rigid supports and pulled slightly so that the resonant frequency increased by a factor of 17; i.e., the stress term in Eq. (2) exceeded the modulus term by ~ 1400. (Therefore, $\sigma \sim 0.4$ GPa.[5]) As expected, no anomaly was observed in the resonant frequency at the transition temperature. When current was applied, the frequency decreased in strict proportionality to the Joule power ($|\Delta f/f_0 - mP/2| < 10^{-5}$); i.e., the length and tension change with heating but not with CDW depinning. The internal friction also changed negligibly for the sample under stress [$\Delta(1/Q) < 2\times 10^{-4}$]. These results indicate that the observed changes for the unstressed sample are due to changes in the dynamic modulus and not, e.g., due to changes in the dimensions of the sample or the quality of clamping.

Most surprising is the large magnitude of the field dependence of modulus and internal friction; the modulus change of a few percent is comparable to the (zero voltage) anomaly at T_c, while the internal friction change ($\sim 10^{-2}$ at 100 K) is two orders of magnitude greater than that observed at the transition temperature ($\sim 6\times 10^{-5}$, comparable to those previously reported[5]). However, it is seen that the internal friction change decreases greatly as the temperature approaches T_c, and it is possible that the field-dependent anomaly and small thermal anomaly at T_c are related; e.g., they may both be due to CDW domain wall relaxation under the oscillating strain. When the field exceeds threshold,

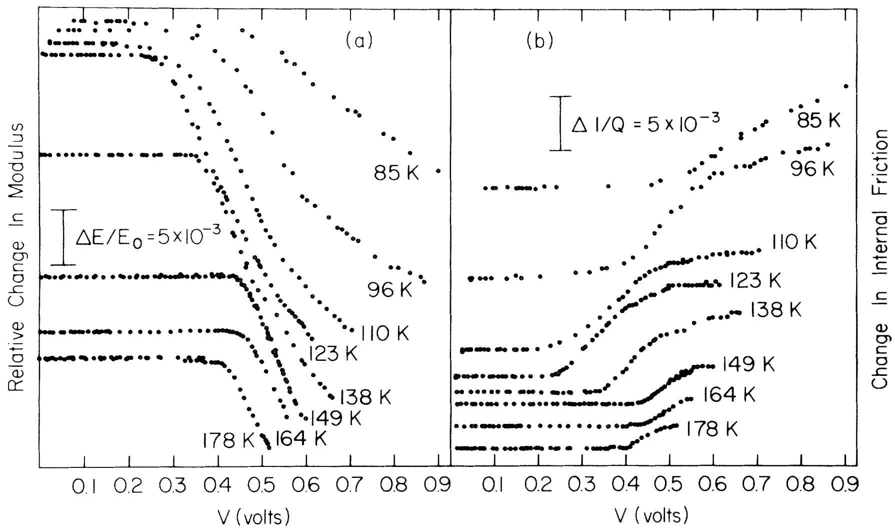

FIG. 2. (a) Relative change in modulus and (b) change in $1/Q$ vs voltage at several temperatures. The modulus is corrected for heating. The vertical offsets of both graphs are arbitrary. $Q^{-1}(V=0) \sim 1.4\times10^{-3}$ at all temperatures.

the domains are believed to realign.[8] When the CDW is pinned, the domain wall relaxation time, τ, may be much greater than when it is depinned, so that the decrease in modulus with increasing field may reflect the change from an unrelaxed ($\omega\tau >> 1$) to more relaxed ($\omega\tau \sim 1$) measurement[4]:

$$[E(\tau)-E(\infty)]/E(\infty) = -F/(1+\omega^2\tau^2),$$
$$\delta(\tau)-\delta(\infty) = F\omega\tau/(1+\omega^2\tau^2), \tag{4}$$

where F is the (temperature dependent) relaxation strength. For example, τ is expected to diverge as ϵ approaches ϵ_T from above. Although the curves shown in Fig. 2 can be qualitatively described by Eq. (4), they are poor quantitative fits, especially at the higher temperatures where the change in friction is much less than that in modulus, whereas comparable changes are predicted by Eq. (4). In fact, we have not been able to extend the field to values at which the modulus is saturated without greatly overheating the sample, whereas the internal friction saturates for $\epsilon \sim \epsilon_T$ to indicate that there is a static contribution to the modulus shift in addition to the dynamic relaxation shift.

While the domain wall relaxation strength will be defect and temperature[8] dependent, a rough upper limit can be estimated by the assumption that response of the total CDW to stress varies from unrelaxed ($\epsilon < \epsilon_T$) to relaxed ($\epsilon > \epsilon_T$). In the former case, the CDW will distort with respect to the lattice and increase the elastic modulus by the effective modulus of the CDW, as derived by Fukuyama and Lee[9]:

$$E_{\rm CDW} = h v_{\rm F}/\lambda^2\Omega, \tag{5}$$

where λ is the CDW wavelength, Ω the area per conducting chain, and $v_{\rm F}$ the Fermi velocity. Taking $v_{\rm F} = 5.4\times10^8$ cm/s[10] and $E \sim 400$ GPa,[5] we find $F_{\max} \sim E_{\rm CDW}/E \sim 2\times10^{-2}$, comparable to the observed shifts.

Alternatively, the increase in the internal friction anomaly with decreasing temperature may be related to the increase in the CDW Peltier current with decreasing temperature,[3] and the field-dependent elastic properties may be intrinsic to the depinned CDW state, depending, e.g., on the CDW drift velocity, $v_{\rm CDW}$. In Fig. 3, we have plotted $\log|\Delta E/E_0|$ vs $\log I_{\rm CDW}$, where the CDW current, $I_{\rm CDW} = I - V/R_0$, is proportional to $v_{\rm CDW}^1$. (R_0 is the normal, $\epsilon < \epsilon_T$, resistance.) It is seen that $\Delta E/E_0$ varies as $I_{\rm CDW}^p$ where $\frac{1}{3} < p < \frac{1}{2}$, which suggests that the changes in the phonon propagation with CDW depinning cannot be treated by a model which treats $v_{\rm CDW}$ as a perturbation parameter.

In summary, we have observed that the Young's modulus and internal friction of o-TaS_3 are strongly electric field dependent for fields greater than the threshold of non-Ohmic conduction; this provides a new test for models of CDW depinning. The modulus decrease is proportional to $I_{\rm CDW}^p$, where $p \sim 0.4$. The internal friction increase can be modeled as a relaxation effect, where the relaxation strength $F \rightarrow 0$ as $T \rightarrow T_c$ and the relaxation time $\tau \rightarrow \infty$ as $\epsilon \rightarrow \epsilon_T$ from above; however, the modulus increase is too large to be described by a

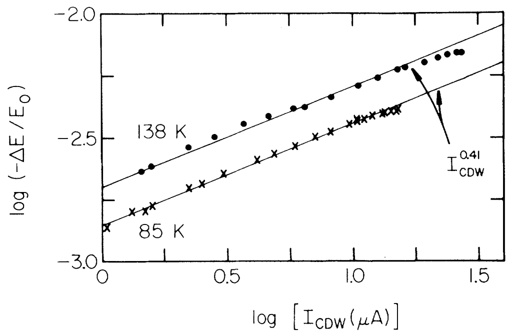

FIG. 3. Relative change in modulus, corrected for heating, vs CDW current at two temperatures. The lines have slopes = 0.41.

simple relaxation process.

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Note added.—Effects qualitatively similar to those reported here have recently been observed in TaS_3, $NbSe_3$, and $(TaSe_4)_2I$ by G. Mozurkewich, P. M. Chaikin, W. G. Clark, and G. Grüner.[11]

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