Frequency and voltage dependence of the complex shear compliance of TaS_3: A relaxation analysis

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We have measured the complex shear compliance of the charge-density-wave (CDW) conductor orthorhombic TaS_3 for elastic frequencies between 0.1 and 30 Hz, by directly measuring the sample strain (twist angle) for an applied alternating stress (torque). Both the real and imaginary parts of the compliance increase when the CDW is depinned. If the CDW is depinned with an ac current, the elastic anomalies depend on the frequency of the current, as previously observed for the Young's modulus at higher frequencies. For dc depinning, we have fit the complex compliance to a generalized Debye relaxation expression [S. Havriliak and S. Negami, J. Polym. Sci. C **14**, 99 (1966)]. This fit allows us to determine the voltage dependence of the relaxation strength and average relaxation time: $\tau \sim (V-V_T)^{-3}$, where V_T is the threshold for CDW depinning. [S0163-1829(97)07928-9]

INTRODUCTION

Quasi-one-dimensional conductors exhibiting sliding charge-density-waves (CDW's) are among the most unusual materials known.[1,2] For zero applied electric field, the CDW is pinned by crystal impurities and does not contribute to the (dc) conduction. Above a small threshold electric field (as low as 1 mV/cm), however, the CDW becomes depinned and carries current. Among the anomalous properties of CDW conductors, associated with depinning and deforming the CDW, are large non-Ohmic conductivities, huge ($\epsilon \approx 10^9$) dielectric constants, a variety of memory effects associated with the many CDW degrees of freedom and resulting metastability, and electric-field-dependent elastic moduli.[1–3]

Most measurements of the elastic moduli have been performed by measuring the resonant frequencies and bandwidths of mechanical oscillators incorporating the samples as the elastic elements: i.e., vibrating reed measurements of Young's modulus (Y) and torsional pendulum measurements of shear modulus (G).[3] For orthorhombic TaS_3 (o-TaS_3), the best studied material, the real parts of Y and G decrease by $\approx 2\%$ and 20%, respectively, with CDW depinning for low frequency (<1 kHz) oscillations. The internal friction (proportional to the imaginary parts of the moduli) increases rapidly (by $\approx 10^{-3}$ and 10^{-2}, respectively) as the electric field is increased above threshold.[3,4]

Interpretation of these elastic changes has proven elusive. Models[5] that treated the anomalies as intrinsic to a uniformly depinned and sliding CDW have met both theoretical objections[3,6,7] and been inconsistent with several experimental observations.[3,8] Alternatively, it has been suggested that the elastic anomalies are associated with the relaxation of CDW domains in the strained crystal.[9,10] In this case, the average relaxation time is assumed to diverge at the depinning threshold. However, quantitatively determining the relaxation parameters with resonance experiments has proven difficult,[4] because one is thereby forced to work at a few widely seperated frequencies and/or resonance modes involving different strains in the crystal.

Recently, we developed a technique to directly measure torsional strain as a function of torque, and therefore the shear compliance ($J \equiv 1/G$), in thin samples of o-TaS_3 for frequencies between 0.03 and 30 Hz, and showed that the increase in the real part of J with CDW depinning grows (logarithmically) with decreasing frequency in this range.[11] We have now extended these measurements to include the imaginary part of the compliance. These measurements show that for a relaxation model to work, the relaxation strength as well as the relaxation time must be field dependent. We show that the complex compliance can be described by a generalized Debye-like relaxation equation, and thereby determine the field dependence of the relaxation strength and average relaxation time for one sample at two temperatures. (Note, however, that the parameters describing the elastic response are sample dependent, and are also greatly affected by sample aging.) These and related measurements and analysis are discussed in this paper.

BACKGROUND

o-TaS_3 is a quasi-one-dimensional metal that undergoes a Peierls transition at $T_c = 220$ K into a semiconducting state due to the [three-dimensional] condensation of a periodic lattice distortion and associated charge-density wave.[12] Crystals are filamentary in shape, with typical dimensions 1 cm in the high conductivity direction and 10 μm^2 in cross section. In the CDW state, the electronic density is given by $\rho = \rho_0 + \rho_1 \cos(qz+\phi)$, where z is the high-conductivity direction, $q/2 \approx k_F$, the Fermi wave vector, ρ_0 is the density of condensed electrons (essentially the entire conduction band for $T < T_c/2$ in this semiconducting state), and ρ_1 is the CDW amplitude. For small applied electric fields, the local phase $\phi(\mathbf{r},t)$ is collectively pinned by crystal impurities. The length scale for ϕ changes is given by ξ_{LR}, the Lee-Rice length; $\xi_{\mathrm{LR}} \propto k_F/n_I$, where n_I is the impurity concentration, and is of the order 1 μm in typical crystals.[13] Because of the complexity of the dependence of the free energy on the phase configuration, the CDW is generally trapped in a metastable configuration when a parameter such as temperature

changes.[1] When the CDW is pinned, dc current is carried by quasiparticles thermally excited across the Peierls gap (≈ 150 meV).[12]

For applied voltages greater than a threshold $V_T \propto n_I^2$, the CDW is depinned and carries current.[1,2] At 100 K, the collective CDW current for $V \gg V_T$ is two orders of magnitude larger than the quasiparticle current.[1] Nonetheless, the CDW transport does not proceed by the smooth variation of ϕ with time; rather, in a manner similar to slip-stick friction, the phase undergoes rapid 2π jumps.[14] Thus, the CDW maintains local phase memory even when ''sliding.''

Using vibrating-reed measurements on flexural resonances (≈ 1 kHz), it was found that the Young's modulus of o-TaS_3 reversibly decreased with CDW depinning.[9,15] Near threshold, $\Delta Y \propto (V - V_T)^2$, and the modulus decrease saturated between 1 and 2% for $V \approx 10 V_T$.[9] The internal friction, given by the reciprocal of the quality factor of the resonance, increased abruptly as the voltage was increased above threshold and then either saturated or slowly decreased for larger voltages. The magnitude of the internal friction change was sample dependent, but was usually $\approx 2 \times 10^{-3}$.[9] Using the sample as a torsional pendulum (near 100 Hz), it was later found that the shear modulus had a similar voltage dependence, but that its anomalies were an order of magnitude larger.[6]

These voltage dependences suggested that the elastic anomalies were relaxational in nature. For a single relaxation process with relative strength A and relaxation time τ, a complex stiffness modulus M is given by the Debye expression[4,9]

$$[M(\omega,V)/M_U]^{-1} = 1 + A/[1 + i\omega\tau(V)], \qquad (1)$$

where M_U is the unrelaxed (i.e., $\omega\tau = \infty$) stiffness modulus, presumed to apply at $V = 0$. (For simplicity in comparison later, the relaxation is expressed in terms of the compliance, $1/M$, instead of the stiffness modulus, as done in Refs. 4 and 9. For $A \ll 1$, the distinction is trivial.) The internal friction $= M\ \mathrm{Im}(1/M)$ and peaks at $A/2$ for $\omega\tau = 1$. If the CDW structure needs to adjust to oscillating strain, it is expected that the relaxation time for these adjustments would be long for the pinned CDW and would decrease above threshold. The elastic anomalies therefore have a natural explanation in terms of relaxation. Consistent with a relaxation model was the observation that the static Young's modulus, measured with uniaxial stress, did not change with CDW depinning.[16]

It was initially suggested that the relaxing quantities were velocity coherent domains,[9] since the velocity coherence length was thought to diverge at threshold.[17] However, this idea was not developed, in part because most o-TaS_3 crystals do not exhibit the expected[9,17] dependence of CDW current on voltage [$I_{\mathrm{CDW}} \sim (V - V_T)^{3/2}$]; furthermore, it was found that the shear modulus anomalies commenced at a voltage somewhat below that for which nonzero CDW velocity (i.e., nonlinear current) could be observed.[6,11]

On the other hand, the fact that the CDW maintains phase memory when sliding makes it possible that the phase configuration is the relaxing quantity, and this was proposed by Mozurkewich in 1990.[10] He showed that if the phase domains were driven out of equilibrium by the strain dependence of k_F, then one would expect a decrease in Young's modulus on the order of 1%, independent of sample purity, as observed. The expected change in shear modulus in this model is[10,11]

$$\Delta G/G_U = -(M_{\mathrm{CDW}}/G)(\partial \ln k_F/d\epsilon)^2, \qquad (2)$$

where M_{CDW} is the CDW's (longitudinal) rigidity and ϵ is torsional strain. Taking[6,10] $G_U \approx 5$ GPa and $M_{\mathrm{CDW}} \approx 2$ GPa therefore implies $\partial \ln k_F/d\epsilon \approx 1$ for a 20% decrease in shear modulus. Of course, any nontrivial dependence of k_F on long-wavelength strains implies a non-rigid band structure; in the present case, it means that the Fermi surface and/or Brillouin zone is extremely sensitive to changes in bond angles, reflecting cation/anion covalency and/or interchain charge transfer.

Subsequently, it was found that if instead of depinning the CDW with a dc voltage, a symmetric square wave of frequency f_I was applied to the sample, so that the direction of CDW motion was periodically switched, the Young's modulus anomalies varied with switching frequency: the decrease in the real part of Y with depinning was greatest for switching frequencies a few times the flexural resonance frequency, while the depinned internal friction had a pronounced maximum when the two frequencies were equal.[18,4] Such a dependence on switching was strong evidence that the elastic anomalies could not be intrinsic (i.e., ''equilibrium'' effects) to the depinned CDW. Jacobsen *et al.*[18] suggested that switching affected both the relaxation time and the relaxation strength by opening new channels for relaxation (stirring) for low f_I, but preventing relaxation by the continual change of the relaxed ''ground state'' for large f_I.

Despite the qualitative successes of the phase relaxation model, it has proven difficult to determine how the relaxation time varies with voltage, even in the dc situation. The magnitudes and voltage dependences of the real and imaginary parts of the moduli implied that a distribution of relaxation times was needed at every voltage, i.e., Eq. (1) needed to be replaced with[4]

$$[M(\omega,V)/M_U]^{-1} = 1 + \int d\tau\, \hat{A}(\tau,V)/(1 + i\omega\tau), \qquad (3)$$

but again, $\hat{A}(\tau,V)$ could not be determined. The hope, of course, was that the total relaxation strength, $\int d\tau\, \hat{A}(\tau,V)$, would be independent of voltage.[3,10] Complicating the analysis was the finding, using higher flexural overtones, that the anomalies in the real part of the Young's modulus decreased with frequency above 1 kHz as $\omega^{-3/4}$, but that the shape [i.e., normalized voltage dependence, $F(V)$] of the anomalies was independent of frequency.[8] This would imply that $\hat{A} \propto \tau^{-1/4} F(V)$, but this is inconsistent with the observed magnitude, frequency dependence, and voltage dependence of the internal friction.[4]

Jacobsen *et al.*[18] took the alternative track of plotting, for a single flexural resonance, the imaginary part of $1/Y$ versus its real part, with voltage as an implicit parameter. They showed that the data could be well represented by the generalized Debye expression

$$[M(\omega,V)/M_U]^{-1} = 1 + A/\{1 + [i\omega\tau(V)]^{1-\alpha}\}^{\beta}, \qquad (4)$$

where α, β, and the relaxation strength A were taken as constants. In this expression, α controls the size of the anomalies in the imaginary part and β controls the skewedness of the plot. (For simple Debye relaxation, i.e., $\alpha=0$ and $\beta=1$, a plot of the imaginary versus real parts of $1/M$ is a semicircle.) This expression had originally been used by Havriliak and Negami[19] (HN) to describe the dielectric relaxation of polymers, and was subsequently used to fit the $V\approx 0$ (i.e., pinned CDW) dielectric properties of CDW materials.[20] [Since in principle the modulus described by Eq. (4) can be expressed in terms of an integral of Debye relaxation modes, i.e., Eq. (3), we will refer to Eq. (4) as an HN distribution of relaxation times.] Surprisingly, Jacobsen *et al.*[18] found that $\alpha\approx 0.6$ and $\beta\approx 1.35$, close to the values found for the pinned dielectric constant of o-TaS_3.[20] The dielectric constant had previously been shown to be correlated with broad band noise (BBN) produced by the sliding CDW;[21] e.g., both vary as $\omega^{-3/4}$, like the Young's modulus anomaly. Jacobsen *et al.* therefore suggested that the BBN might be related to the elastic anomalies, and pointed out that the internal friction had a qualitatively similar voltage dependence to the BBN.[18]

To best examine the onset of relaxation at threshold, what was required was to examine a single sample at a range of low frequencies. We originally attempted this, for both the shear and Young's moduli, by studying mechanical resonances with different weights placed on the samples.[4] While qualitative information was thus obtained, it was impossible to make quantitative comparisons, because when changing weights one also changed the detailed nature of the resonant mode. However, we did fit a number of resonances to Eq. (4) and found that different values of α and β were needed for different modes (or that A was voltage dependent) and that the similarity to dielectric properties surmised by Jacobsen *et al.*[18] was coincidental.[4]

We therefore developed a subresonance technique for examining the shear compliance ($J\equiv 1/G$) of $o-TaS_3$ samples directly by measuring the twist of the sample resulting from an applied torque, and reported on changes of the magnitude of the compliance with voltage.[11] We found that the increase in compliance with CDW depinning grows logarithmically with decreasing frequency between 20 Hz and 30 mHz. We also found that the static, as well as dynamic, shear compliance increases by 25% (i.e., G decreases by 20%) with CDW depinning. This is in striking contrast to the Young's modulus, for which no static depinning anomaly was found,[16] and implies that while the pinned torsional time constant is much greater than the experiment time (84 s),[11] the pinned longitudinal time constant is smaller.

We have now extended these low-frequency measurements to include the phase shift between the applied torque and resulting twist, and hence both the real and imaginary parts of the shear compliance. The experimental results, and an analysis in terms of the HN expression [Eq. (4)], are discussed below.

EXPERIMENTAL TECHNIQUES

Relatively thick ($>25\ \mu m^2$ cross section) o-TaS_3 crystals were used for these experiments. The ends of the sample, cut to a length of a few millimeters, were glued with silver paint to current contacts, and a thin, magnetized steel wire ($\approx 60\ \mu$m diameter, 1 mm long) was glued with silver paint to the center of the sample. The sample was located in the center of a helical resonator rf cavity[22] ($\nu\approx 450$ MHz, $Q\approx 300$), with the wire near (≈ 0.1 mm) the tip of the helix, so that as the sample twisted, it modulated the resonant frequency of the cavity. Measurements were made by driving the rf cavity at resonance, and measuring the phase modulation[22] of the transmitted signal with a lock-in amplifier, operating in a magnitude/phase mode.

Oscillating torque was applied by a magnetic field provided by Helmholtz coils (27 G/A). Typically, a dc field of a few Gauss was applied to the sample during an experiment to maintain the magnetization of the wire. The ac field was kept slightly smaller than this so that the twist angle oscillated by $<0.5°$, corresponding to a maximum strain (i.e., at the surface) $<10^{-5}$; we checked that the response was linear for these measurements.

If the magnitude of the oscillating magnetic field was kept constant, the magnitude of the resulting rf modulation is proportional to the magnitude of the compliance and the change in the internal friction equals the change in $\tan(\delta)$, where δ is the phase shift. In most of our experiments, J was measured for a fixed twist frequency f_θ while varying the dc voltage across the sample and simultaneously measuring the (two-probe) resistance. The magnitude and phase of J were normalized to their values at low voltage, which were found to be constant (see Fig. 3). To compare with the Young's modulus results,[4,18] we also measured J and $\tan(\delta)$ applying a square wave voltage of fixed amplitude and variable frequency.

The minimum twist frequency of these measurements (0.1 Hz) was limited by the $1/f$ noise of the demodulating circuit and stability of the helical resonator. The maximum f_θ, ≈ 30 Hz, was an order of magnitude below the torsional resonant frequency of the sample, and was limited by the effective impedance of the magnet circuit. The total frequency response of the system was measured at a few temperatures by using a $NbSe_3$ crystal, which has a similar morphology to TaS_3 but much smaller elastic anomalies,[23] as the torsional element. We found that the compliance of o-TaS_3 at low voltage, normalized to $NbSe_3$, was independent of frequency (magnitude $\pm 1\%$, phase $\pm 0.5°$).

As mentioned above, Jacobsen *et al.*[18] had pointed out that the internal friction had a similar voltage dependence to the BBN, but both of these quantities are sample dependent, and they had never been measured for the same sample. Therefore, after completing elastic measurements on one sample, we used a lock-in amplifier to measure the noise generated in the sample (with no applied torque) driven by a dc current. An attractive feature of this comparison is that the noise could be measured at the same frequency as the internal friction. Probably due to the large necessary mechanical contacts on the sample, the noise below threshold is larger than generally observed in noise measurements.[21]

RESULTS AND DISCUSSION

In Fig. 1(a) are shown the magnitude of the shear compliance and the internal friction for an o-TaS_3 crystal as a function of (dc) voltage measured with $f_\theta=3.3$ Hz at 102 K. The

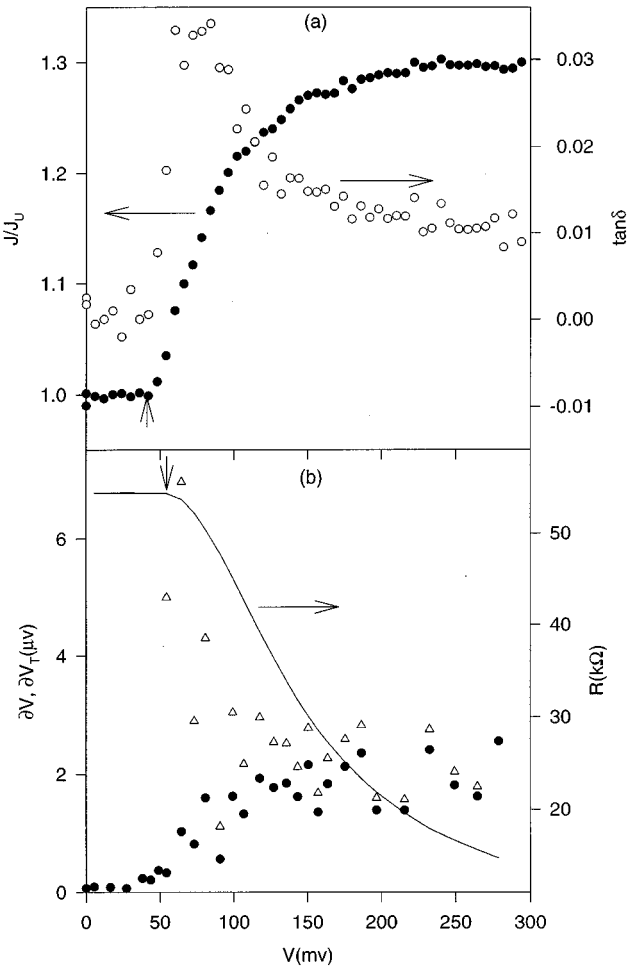

FIG. 1. (a) Compliance and internal friction vs dc voltage for sample No. 1 at $T=102$ K and $f_\theta=3.3$ Hz. (b) dc voltage dependence of resistance and noise ∂V (solid circles), measured at 3.3 Hz. Also shown is the calculated [Eq. (5)] noise in the threshold voltage, ∂V_T (open triangles). The vertical arrows show the elastic and resistance threshold voltages.

results are similar to those we previously reported, although for this sample the magnitude of the change in compliance was slightly larger (30%) than usually observed (25%). In Fig. 1(b) are shown the voltage dependence of the sample's resistance and voltage noise when driven with a dc current; the noise was also measured at 3.3 Hz with a bandwidth of 0.03 Hz.

As previously noted, the threshold for the elastic changes lies below that for changes in resistance.[6,11] In the CDW conductor $K_{0.3}MoO_3$, the threshold for resistance changes has been shown to be greater than the voltage needed to depin and polarize the CDW in the bulk of the crystal. The difference has been associated with the voltage needed to drive the phase slip processes at the current contacts needed to convert quasiparticle current into CDW current.[24] We suggest that the different thresholds observed for nonlinear current and elastic changes in o-TaS_3 has the same origin, and that the onset of the elastic changes at the lower threshold reflects the fact that the elastic changes are sensitive to depinning in the bulk rather than the contacts, as expected.

Interestingly, the enhanced broadband noise has the same threshold as the compliance, suggesting that it too is driven by bulk rather than contact depinning. We are not aware of a previous report of this lower threshold for the BBN in o-TaS_3, but the phase slip voltage may be especially large in our samples, for which the current contacts also serve as mechanical contacts. However, the voltage dependence of the BBN differs significantly from that of the internal friction, in contrast to the discussion of Jacobsen *et al.*[18] More recently, it was suggested[25] that, instead of considering the noise in the voltage, ∂V, one should consider the corresponding noise in the threshold voltage,

$$\partial V_T \equiv (\partial V - \partial V_0)/[1-(R_0 dI/dV)^{-1}], \tag{5}$$

where R_0 and ∂V_0 are the resistance and noise at $V=0$. This expression cannot be applied below the resistance threshold, which is unfortunately where the internal friction is varying most rapidly. However, the calculated threshold noise voltage above the resistance threshold is shown in Fig. 1(b) and it does resemble that of the internal friction.

For the remainder of this paper, V_T will be used to denote the threshold voltage for the elastic anomalies.

In Fig. 2 are shown the effects of reversing the current direction on the shear compliance anomalies; as discussed above, the fact that the Young's modulus anomalies depended on the frequency of current reversals (f_I) was taken as strong evidence that the elastic anomalies could not be understood in terms of ''intrinsic,'' single domain models.[18] The f_I dependence of the compliance and internal friction, measured at $f_\theta=3.3$ and 23.7 Hz, for a square wave voltage of magnitude $5V_T$ are shown. It is seen that, for both torque frequencies, the compliance anomaly doubles when f_I is a few times f_θ; we have previously also observed doubling of the Young's modulus anomaly with changing f_I.[4] More dramatic is the dependence of the internal friction on f_I: when $f_I=f_\theta$, the internal friction anomaly increases by almost an order of magnitude from its dc value, a few times greater than the largest change observed for a flexural oscillation.[18] We emphasize that in addition to doing measurements on a different elastic modulus than before, we are working at frequencies two to four orders of magnitude smaller. These results are a striking confirmation that arguments made in favor of a relaxational mechanism for the elastic anomalies still apply at these low frequencies.

In Figs. 3 and 4 are shown the dc voltage dependences of the compliance and internal friction of another sample at 85 and 102 K. Data are shown for a few values of f_θ; similar data at other intermediate frequencies are omitted for clarity. The J dependence is similar to that shown in Ref. 11; in particular, at a given voltage above threshold the compliance increases logarithmically with decreasing frequency. The frequency dependence is seen to be slightly greater at 85 K than at 102 K.

As previously noted at higher frequencies, the internal friction increases much more rapidly at threshold than the compliance. In a relaxation model, e.g., Eqs. (1) or (3), this reflects the fact that $\tan(\delta)\approx\omega\tau(J/J_U-1)$ as τ diverges at threshold, where J_U is the unrelaxed ($V=0$) compliance. As for the compliance, the frequency dependence of $\tan(\delta)$ is greater at $T=85$ K than at $T=102$ K; this has the results that the sharp cusp observed at the lower frequencies disappears at the higher frequencies at 85 K, but not 102 K.

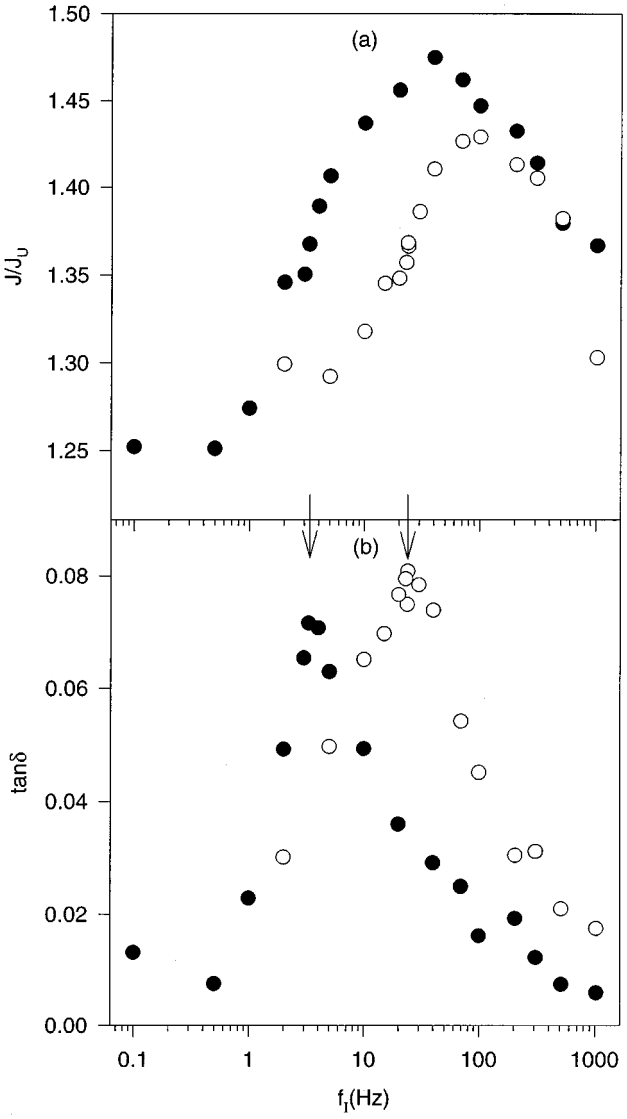

FIG. 2. Compliance and internal friction measured with square wave voltages $V=5V_T$ at frequency f_I, at $T=102$ K for sample No. 2. The elastic frequencies $f_\theta=3.3$ Hz (closed symbols) and 23.7 Hz (open symbols) are shown by the arrows.

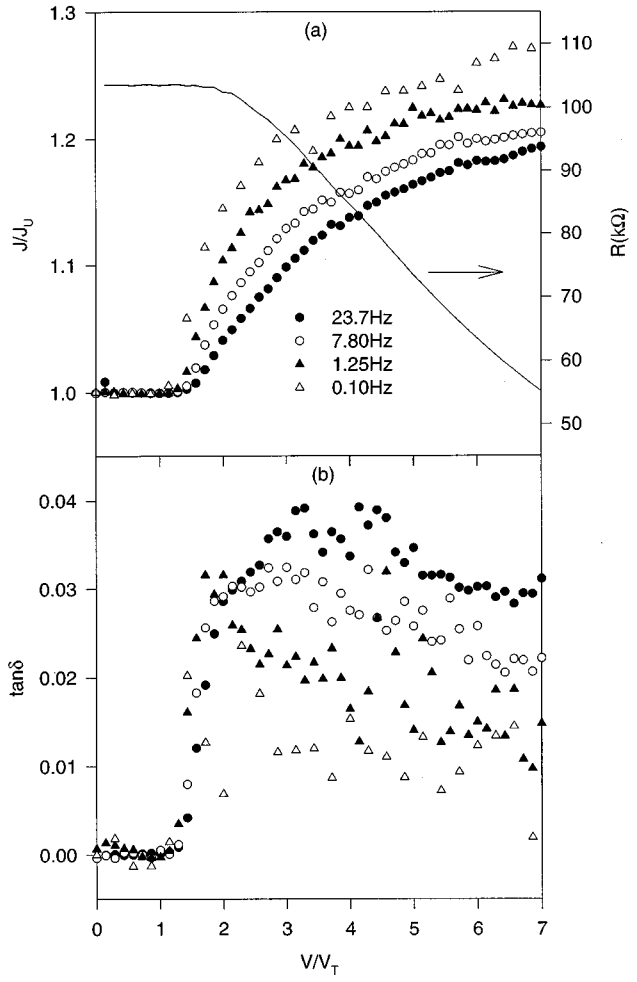

FIG. 3. Compliance and internal friction vs dc voltage at several values of f_θ, as listed, for sample No. 3 at $T=85$ K. The threshold voltage $V_T=70$ mV. Also shown is the resistance.

In Fig. 5 is plotted Im $J/J_U \equiv J/J_U \sin\delta$ versus Δ Re $J/J_U \equiv (J/J_U \cos\delta - 1)$ for several frequencies and voltages at 85 K; qualitatively similar results are obtained at 102 K. Clearly, the values for different voltages do not lie on the same locus, so that a basic conclusion of Ref. 18, based on examining Young's modulus data *at a single frequency*, is incorrect: it would be impossible to fit the compliance data to Eq. (4) with a single, voltage-independent relaxation strength. However, we have found that if we rescale the data for each voltage and plot $\mathrm{Im}J/J_U\Lambda(V)$ versus Δ Re $J/J_U\Lambda(V)$, where Λ is a real function of V, data for all voltages can be made to lie, within their scatter, on a single curve, as shown in Fig. 6, for both 85 and 102 K. The voltage dependence of the scale factor Λ, the effective, normalized relaxation strength, is plotted in Fig. 7.

In Fig. 6 are also shown a few plots of the HN expression [Eq. (4)], with $A=j_0\Lambda$, i.e.,

$$J(\omega,V)/J_U = 1 + j_0\Lambda(V)/\{1+[i\omega\tau(V)]^{1-\alpha}\}^{\beta}, \qquad (6)$$

where α, β, and j_0 are constants, chosen to approximate the data, and $\omega\tau$ is an implicit parameter. Fair fits are obtained with α between 0.6 and 0.66 and β between 1.4 and 2.9, but the best fits (solid curves) have $\beta=2.75$ and 2.00, at $T=85$ and 102 K, respectively, very different from the value found by Jacobsen *et al.*[18] for a single frequency.

For each ''fit'' shown in Fig. 6, the voltage dependence of the average relaxation time can be determined, and these are shown in Fig. 8. Although the magnitude of τ for any voltage varies by up to an order of magnitude, depending on the fitting parameters chosen, for all fits τ diverges as $(V/V_T-1)^{-p}$, with $p\approx 3$. Although there is some overlap, depending on the fitting parameters chosen, the best fits (shown by the solid curves in Fig. 6 and circles in Fig. 7) show that τ is 3–10 times longer at 85 K than at 102 K, consistent with the greater frequency dependence noted (when comparing Figs. 3 and 4) at the lower temperature.

In previous work on the Young's modulus,[9,26] it was found that while the voltage dependence of the softening varied considerably with temperature, the dependence of Re(Y) on CDW current ($I_{\mathrm{CDW}}\equiv I - V/R_0$) was a more universal function of temperature, and it was suggested that the CDW current controlled the elastic anomalies. As discussed above, the fact that the threshold for changes in shear compliance is below that for changes in resistance i.e., the com-

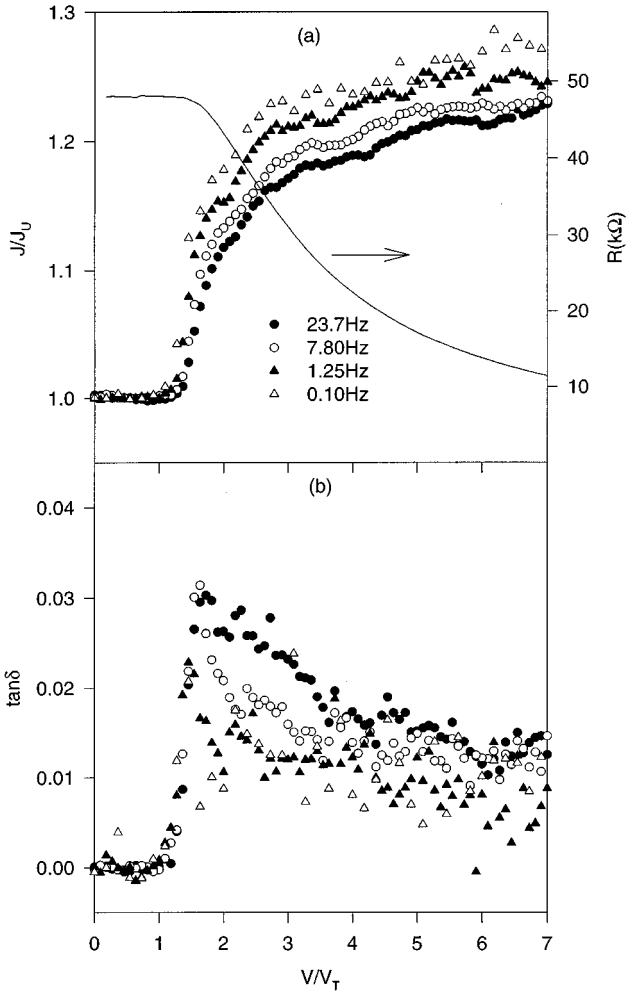

FIG. 4. Compliance and internal friction vs dc voltage at several values of f_θ, as listed, for sample No. 3 at $T=102$ K. The threshold voltage $V_T=55$ mV. Also shown is the resistance.

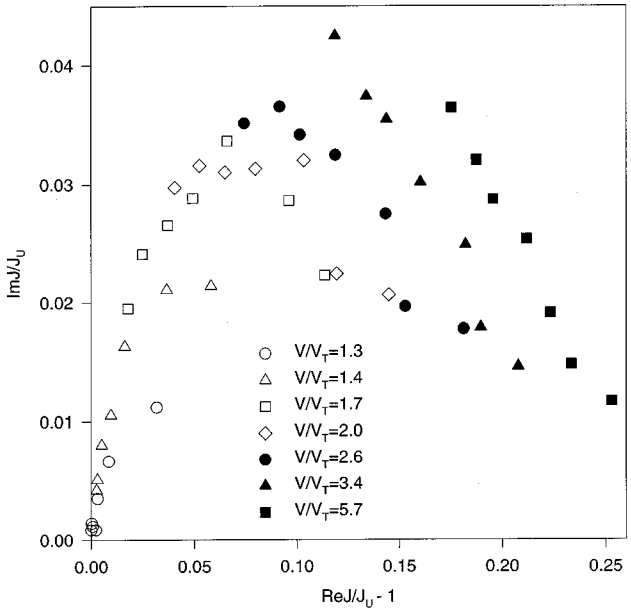

FIG. 5. Imaginary part of the compliance vs the change in the real part of the compliance for sample No. 3 at $T=85$ K, for the dc voltages shown.

pliance begins changing when the CDW current is still zero, means that this relationship must break down near threshold. It does hold for the compliance at larger voltages, as shown for $f_\theta=7.8$ Hz in Fig. 9(a). However, as shown in Fig. 9(b), the internal friction is clearly not a universal function of I_{CDW}, reflecting the fact that relaxation slows down with decreasing temperature. Therefore, there is no advantage in considering the dependence of the HN relaxation parameters on CDW current over voltage.

We need to emphasize that while qualitatively similar data have been obtained for several samples, the frequency and voltage dependences of the compliance anomaly of each sample are quantitatively different. Furthermore, these dependences change with time. The 85- and 102-K data shown above for sample No. 3 (Figs. 3–9) were taken 10 days apart, during which time the sample warmed to room temperature. We attempted to retake 102-K data two months later; during this two month period the sample was at room temperature (and had been briefly heated to 100 °C). As shown in Fig. 10, the compliance anomaly had changed remarkably while the threshold voltage increased by 50%. The data shown are for $f_\theta=3.3$ Hz; similar changes were observed at other torque frequencies. The increased size of the compliance anomaly at the later data indicates that the total relaxation strength had increased, while the second peak in the internal friction indicates that a new low-frequency relaxation process had turned on. We suggest that when aging, some silver paint diffuses into the sample, so that the distribution of CDW pinning centers becomes less homogeneous. We have observed similar double internal friction peaks on a few other samples. Considerable caution must therefore be used in comparing data taken at different times or on different samples.

CONCLUSIONS

We have shown that the frequency- and voltage-dependent elastic compliance can be fit to a Havriliak-Negami expression if one allows the relaxation strength to be voltage dependent, and have determined the voltage dependence of the relaxation strength and relaxation time; the latter varies as $\tau\sim(V-V_T)^{-p}$, with $p\approx3$, and grows (by up to an order of magnitude) between 102 and 85 K. This power law is much stronger than previously estimated ($p\approx1$) from higher-frequency measurements on a single mode.[4] In the context of the phase relaxation model, τ is the average time needed for phase coherent domains to readjust to a changing parameter such as stress; we are not aware of any prediction regarding its voltage dependence.

The dielectric relaxation (below threshold) of o-TaS_3 was previously fit[20] to the HN expression, with α between 0.5 and 0.6, similar to our results, but $\beta=1.35$, much less than our result, indicating a much more symmetric variation of Im(ϵ) with Re(ϵ) than obtained for the compliance. It must be emphasized, however, that the dielectric relaxation times near 100 K were ≈0.1 μs,[20] so that the dielectric measurements were sensitive to a much different regime of dynamics than we are probing. (Pinned dielectric relaxation times of the order of 1 s are observed only at $T\approx25$ K.[27]) We have not yet considered how to combine our present results with previous high-frequency measurements of the Young's

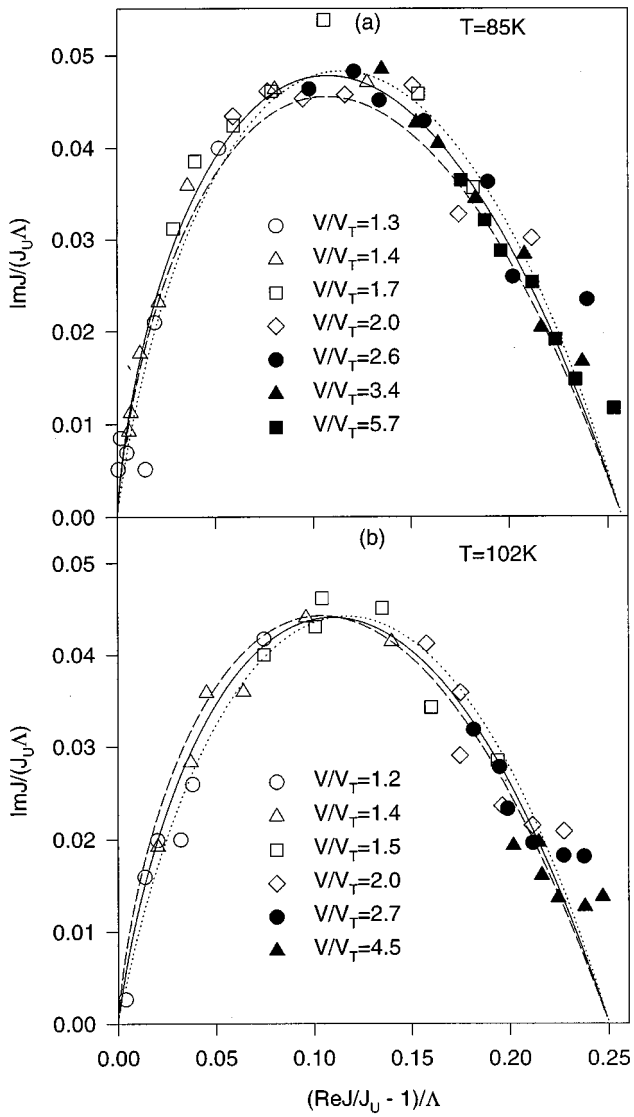

FIG. 6. Imaginary part of the compliance vs the change in the real part of the compliance, each normalized by the voltage-dependent factor Λ (shown in Fig. 7) for sample No. 3 at (a) $T=85$ K and (b) $T=102$ K. Also shown are plots of Eq. (6) with the following parameters: (a) solid curve: $j_0=0.257$, $\alpha=0.64$, $\beta=2.75$; dotted curve: $j_0=0.257$, $\alpha=0.61$, $\beta=1.8$; dashed curve: $j_0=0.257$, $\alpha=0.66$, $\beta=2.9$. (b) Solid curve: $j_0=0.250$, $\alpha=0.64$, $\beta=2.00$; dotted curve: $j_0=0.250$, $\alpha=0.61$, $\beta=1.4$; dashed curve: $j_0=0.250$, $\alpha=0.66$, $\beta=2.9$.

modulus,[8] which showed that the elastic anomalies vary as $\omega^{-3/4}$ above 1 kHz.

The voltage dependence of the relaxation strength has a natural explanation in terms of relaxation of velocity coherent domains, since the domain size diverges and the number of domains decreases as V approaches V_T from above.[17] However, since the elastic threshold is less than the resistance threshold, an interpretation in terms of CDW velocity, rather than CDW depinning, is difficult. While it is possible that this inconsistency is related to inhomogeneities in the sample, it is not clear how to correct for them.

On the other hand, the density of phase domains is not expected to vary with voltage, so that in the phase domain relaxation model, it is expected that the relaxation strength

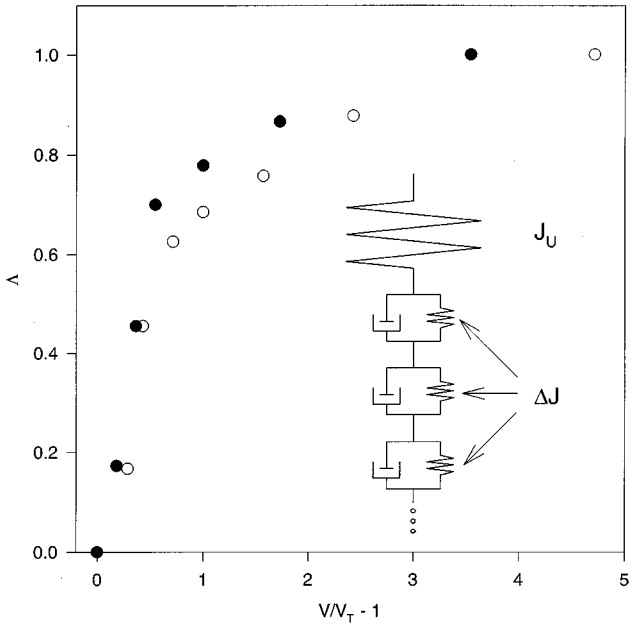

FIG. 7. Normalized relaxation strength Λ vs voltage for sample No. 3. Open circles: $T=85$ K; closed circles: $T=102$ K. Inset: mechanical model discussed in text.

would be independent of voltage. Indeed, this was one of the attractive features of the model when it was proposed.[10] Our results can be reconciled with the model, however, if one assumes that added to the distribution of relaxation strengths we obtain is a reservoir of domains with much longer relaxation times (e.g., $\tau>84$ s, the time of our previous quasi-static experiments[11]). Below threshold, all the domains are in this reservoir. As the voltage is increased, an increasing number of domains, indicated by the growth of the relaxation

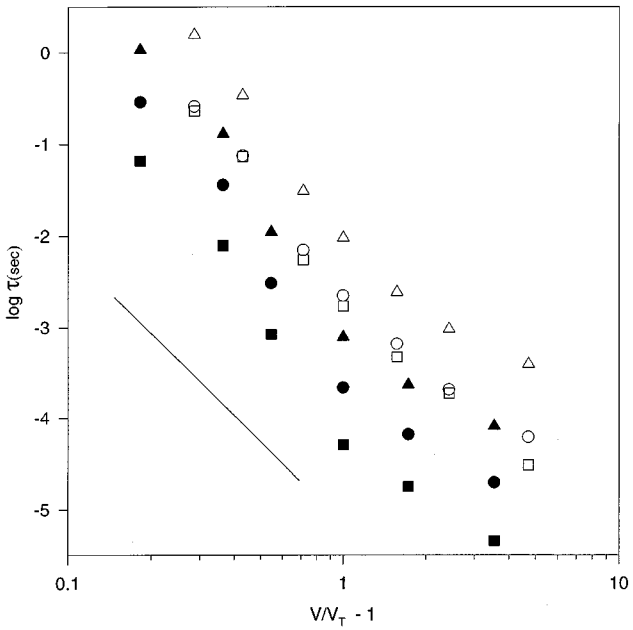

FIG. 8. Voltage dependence of average relaxation times for the fits to Eq. (6) shown in Fig. 6. Circles correspond to the solid curves in Fig. 6, triangles correspond to dotted curves, and squares correspond to dashed curves. Open symbols: $T=85$ K; closed symbols: $T=102$ K. The line with slope $=3$ is for reference.

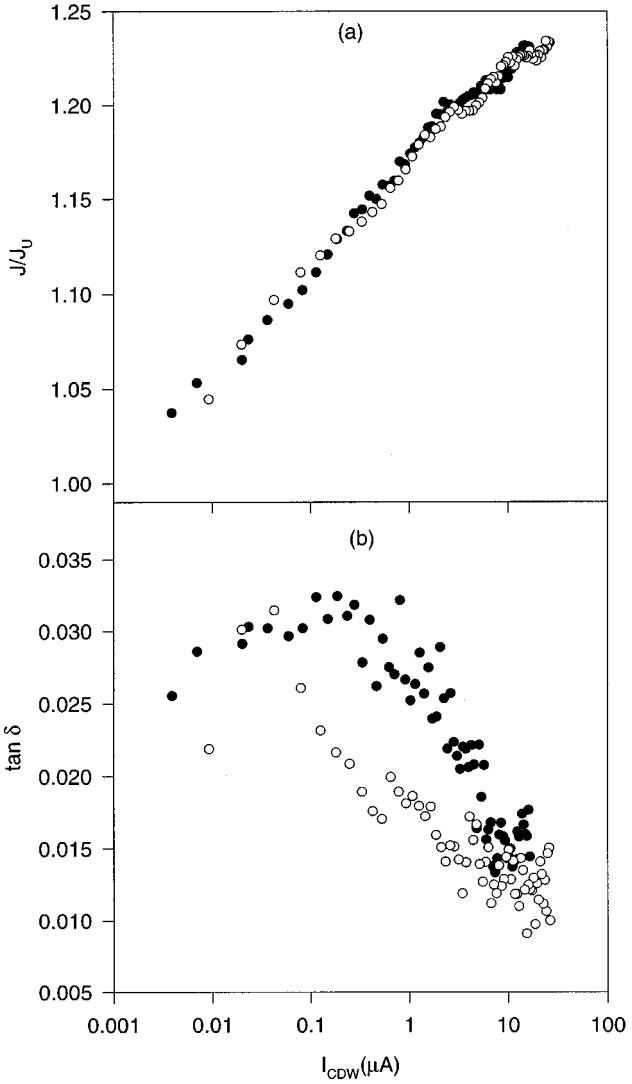

FIG. 9. Compliance (a) and internal friction (b) at $T=85$ K (closed circles) and $T=102$ K (open circles) vs CDW current for sample No. 3 at $f_\theta=7.8$ Hz.

strength, respond quickly enough to become observable, and are described by the HN distribution.

The phase domain model can be understood in terms of the Voigt anelastic model shown in the inset to Fig. 7. Here, the unrelaxed compliance, J_U, is that of the ''bare'' crystal; the series of small springs with parallel dashpots describe the elastic response of the phase domains; i.e., $\Delta J \equiv J(\omega,V) - J_U$. Below threshold, the viscosity of the dashpots is ''infinite,'' so that the small springs are clamped and the elastic response of the crystal is determined by J_U alone. As the

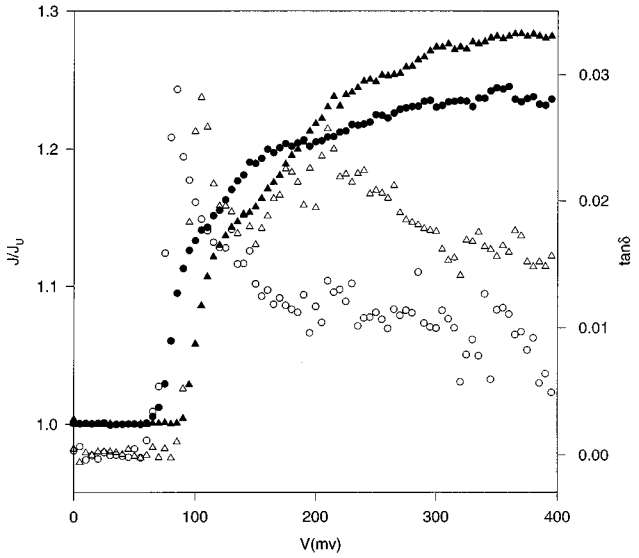

FIG. 10. Compliance (solid symbols) and internal friction (open symbols) for sample No. 3 at $T=102$ K and $f_\theta=3.3$ Hz. Data shown with triangles were taken 2 months after the data shown with circles, as described in text.

voltage is increased above threshold, the fraction $\Lambda(V)$ of the small springs becomes unclamped, with a time constant (proportional to the viscosity of the dashpot) distribution given by the HN expression. If the voltage is decreased below threshold when torque is applied to the sample, the CDW springs will be clamped in a strained position, giving rise to the strain memory effects discussed in Ref. 11.

In summary, we have studied changes in the complex shear compliance of o-TaS_3 observed with CDW depinning at low frequencies, 0.1 Hz$\leqslant f_\theta <$30 Hz of applied stress. We have shown that the strong dependence of the elastic anomalies on the frequency of current reversal, previously observed at higher f_θ, persists to low frequency, suggesting that even at very low frequencies, the anomalies must be due to the rearrangement of internal CDW structure, i.e., ''domains,'' rather than intrinsic to the sliding CDW state. We have then studied the frequency and (dc) voltage dependence of J for one crystal in detail, and shown that $J(\omega,V)$ can be fit to the Havriliak-Negami generalization [Eq. (6)] of simple Debye relaxation, and have used this fit to determine the voltage dependence of the relaxation time and strength.

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