# Frequency Dependence of Elastic Anomalies in Charge-Density-Wave Conductors

X.-D. Xiang and J. W. Brill
*Department of Physics and Astronomy, University of Kentucky, Lexington, Kentucky 40506-0055*
(Received 10 July 1989)

We have measured the frequency dependence of the Young's-modulus ($Y$) and internal-friction anomalies in the charge-density-wave conductors $NbSe_3$ and $TaS_3$. Measurements were made on overtones of flexural resonances using a helical resonator detector between 1 and 100 kHz. Temperature-dependent anomalies were independent of frequency, whereas electric-field-dependent anomalies decreased rapidly with frequency; e.g., $\Delta Y/Y \propto \omega^{-p}$, where $p \sim \frac{3}{4}$. These results are inconsistent with all models proposed of the elastic properties of the charge-density-wave state.

PACS numbers: 71.45.Lr, 62.20.Dc, 62.40.+i, 72.15.Nj

The dynamics of charge density waves (CDW's) has proven to be an extremely rich field; a number of surprising and novel phenomena, associated with the depinning of the CDW at a threshold electric field $E_T$, have been observed over the past decade.[1] A nagging theoretical question has been the explanation of the elastic anomalies observed at $E_T$: The Young's and shear modulus decrease with CDW depinning, while both increases and decreases are observed in the associated damping (i.e., internal friction), depending on the material and acoustic mode.[2-7] Mozurkewich *et al.*,[2(b)] Sneddon,[8(a)] and Sherwin and Zettl[8(b)] explained these changes as due to the decoupling of the elasticity associated with the CDW from that of the lattice as the CDW becomes depinned. In contrast, Maki and Virosztek[9] suggested that the elastic depinning anomalies reflect changes in the ability of the CDW to screen acoustic phonons. Most recently, Zeyher[10] considered increases in internal friction due to the decay of acoustic phonons into phase modes of the CDW. While each of these models has had some success, there are experimental observations in *qualitative* disagreement with each.[6] Because each model makes very different predictions regarding the frequency dependence of the elastic anomalies (see below), such measurements are desirable to illuminate the important physics governing these effects.

We have measured the frequency dependence of the Young's-modulus ($Y$) and internal-friction anomalies in $TaS_3$ and $NbSe_3$. Measurements were made as functions of both temperature through the CDW phase (i.e., "transition" anomalies) and electric field ("depinning" anomalies). Although previous measurements have suggested that depinning anomalies are much smaller at 10 MHz[4] than at 1 kHz,[3] it was not clear to what extent these results were sample dependent or how the anomalies varied at intermediate frequencies, and it was desirable to measure the frequency dependence of individual samples. We have now done this (for frequencies between 1 and 100 kHz) by measuring several overtones of flexural resonances. We find that while the transition anomalies are independent of frequency, the depinning anomalies (for both modulus and internal friction) fall continuously with frequency, in striking contrast to predictions of all three models.

$TaS_3$ and $NbSe_3$ are filamentary quasi-one-dimensional conductors. $TaS_3$ has a single CDW transition at 220 K and $NbSe_3$ has two, at 142 and 58 K;[1] in this paper, we will discuss its upper CDW transition only. The resistivity decreases rapidly when the applied electric field exceeds threshold, as the CDW becomes depinned and contributes to the dc conductivity.[1] The threshold electric field depends on the sample purity.[1,11] The low-frequency Young's moduli of $TaS_3$ and $NbSe_3$ are discussed at length in Refs. 3 and 6, respectively.

Flexural resonances were detected using a high-sensitivity helical resonator detection circuit described in detail in Ref. 12. Single crystals of $NbSe_3$ or $TaS_3$, of typical dimensions $3\,\text{mm}\times 10\,\mu\text{m}\times 3\,\mu\text{m}$, were glued down at one end with silver paint to a piezoelectric transducer, which was driven with an ac voltage phase locked to the sample resonance.[3,12] For temperature-dependent measurements, the second end of the sample was left free, while for voltage-dependent measurements it was clamped down (with an electrical contact) to a second transducer which could be driven with a dc voltage to vary uniaxial strain in the crystal. In the absence of such strain,[3] the flexural resonant frequencies are given by

$$f_n = a_n t/L^2 (Y/\rho)^{1/2}, \tag{1}$$

where $t$ is the thickness, $L$ the length, and $\rho$ the density of the crystal. The constants $a_n$ describe the overtones for the two different sets of boundary conditions.[13] Thus relative changes in $Y$ can be found from relative changes in the resonant frequency. The change in the reciprocal quality factor, $\Delta(1/Q)$, equals the change in internal friction.

Equation (1) holds only if negligible uniaxial stress (e.g., resulting from differential thermal contraction) is present.[3] We found, using the dc transducer, that bowing the sample only decreases resonant frequencies

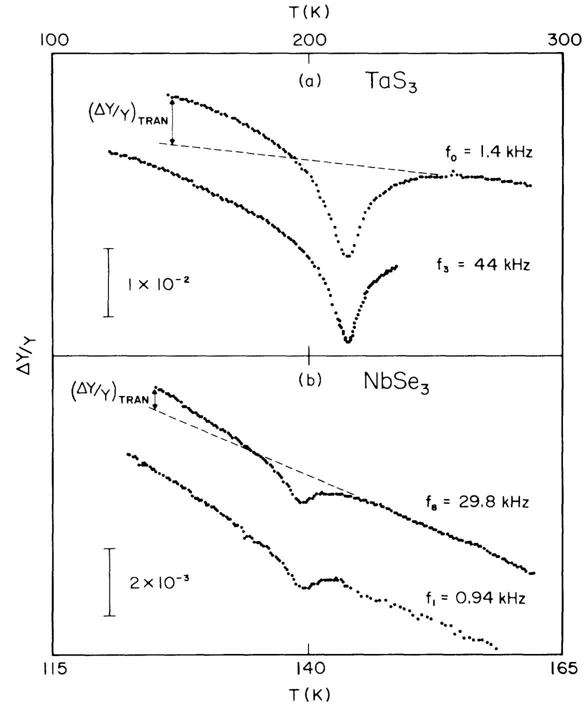


FIG. 1. Temperature dependence of the Young's moduli of (a) $TaS_3$ (sample No. 1) and (b) $NbSe_3$ (sample No. 5). Results are shown for two flexural modes, at frequencies $f_n$, of each; vertical offsets are arbitrary. The dashed lines are rough extrapolations of the pretransition behavior to low temperatures; the extra stiffening below the transitions, $(\Delta Y/Y)_{tran}$, is indicated.

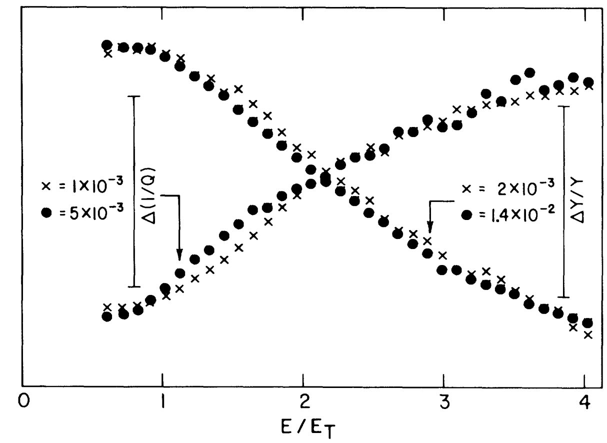


FIG. 2. Electrical field dependence of the Young's modulus and internal friction for $TaS_3$ (sample No. 2) at 103 K. Results are shown for the fundamental flexural mode (circles, $f_0=4$ kHz) and the third overtone (crosses, $f_3=48$ kHz).

slightly, and does not noticeably affect measured anomalies [i.e., $\Delta(1/Q)$ and $\Delta f/f$ are constant within $\pm 10\%$]. Pulling the sample increases the resonant frequencies rapidly and decreases the anomalies;[3,6] the correct transducer voltage was found by measuring the voltage dependence of the frequencies and adjusting the overtones to be in the correct ratios (within 20%[12]). Excessive Joule heating of the sample, which also affects its resonant frequency, was avoided by keeping the sample in a helium atmosphere.[3,6]

Transition anomalies in Young's modulus for both materials are shown in Fig. 1; results for two modes of a single crystal of each are shown. The modulus has a pronounced minimum, associated with fluctuations in the polarizability,[14] at $T_c$; below the transition, the modulus increases above its extrapolated pretransition value. This increase, $(\Delta Y/Y)_{tran}$, has been associated with the modulus of the CDW[8,11] and, alternatively, the inability of the pinned CDW to screen the acoustic phonons.[9] As seen in the figure, both the minimum and the subsequent increase are independent of frequency within the scatter ($\sim 10\%$); such was the case for all modes checked. This frequency independence is expected for the minimum;[14] the behavior of $(\Delta Y/Y)_{tran}$ will be discussed further below.

The electric field dependence of the modulus and internal friction for two modes of a sample of $TaS_3$ at 103 K are shown in Fig. 2; the behavior at low frequencies is similar to that previously reported, although none of the present samples had the broad maximum in $1/Q$ at $\sim 2E_T$ that is often observed.[3] At the higher frequency, the field dependences of the anomalies are similar in shape, but smaller in magnitude. A similar result is obtained for $NbSe_3$. The frequency dependences at 103 K of the depinning anomalies for all crystals measured are shown in Fig. 3. (Because the modulus saturates slowly with field for $TaS_3$,[3] we have for consistency defined the depinning anomaly as the value at $4E_T$; e.g., $|\Delta Y/Y|_{depin}=[Y(0)-Y(4E_T)]/Y(0)$. For $NbSe_3$, the anomalies are much sharper, but Joule heating is a greater problem,[6] so the anomalies are defined at $2E_T$.)

For $TaS_3$, the modulus depinning anomaly decreases with frequency roughly as $\omega^{-p}$, with $p\sim\frac{3}{4}$ for samples No. 2 and No. 6 and $p\sim\frac{1}{2}$ for sample No. 3. Extrapolation of the modulus anomalies to 10 MHz gives values of $\sim 2\times 10^{-4}$, similar to the ultrasonic results of Jericho and Simpson.[4] The internal-friction anomaly also decreases rapidly with frequency for low frequencies, but saturates at high frequencies at $\sim 10^{-3}$, also consistent with the ultrasonic results.[4] (Small kinks in the frequency dependence of the modulus are also observed at the internal-friction saturation frequencies.) The saturation frequency for sample No. 3 is $\sim 2$ kHz, a few times smaller than that of the other two samples. The quantitatively different behavior observed for No. 3 does not correlate with the threshold field, which is similar for all three samples. Sample No. 3 is considerably thinner, as shown in the legend, suggesting that the transverse coherence of the CDW might be important.[3,5] However, the depinning modulus anomalies measured for modes

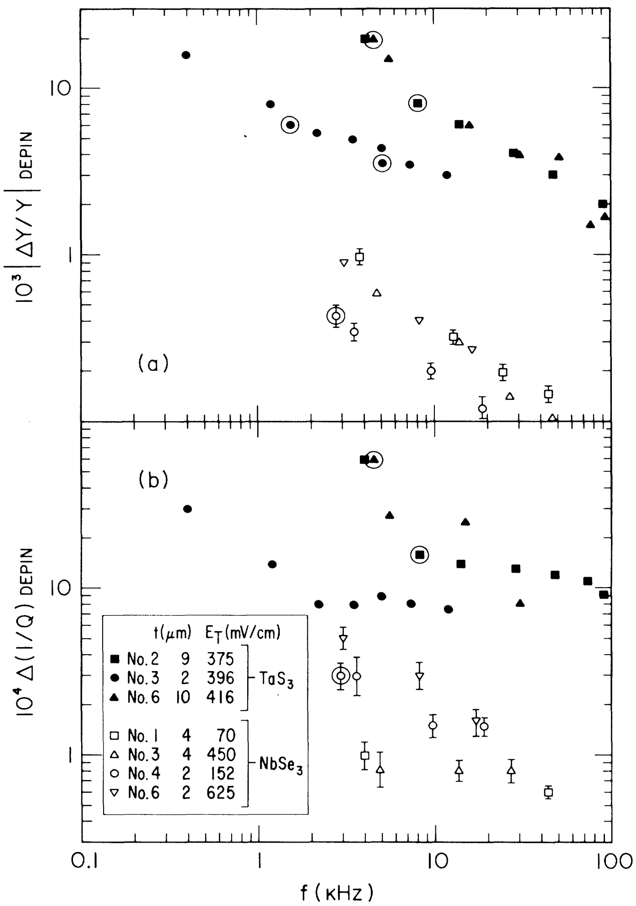


FIG. 3. Depinning anomalies, at 103 K, in (a) Young's modulus and (b) internal friction vs frequency. For $TaS_3$, the anomaly is measured at $4E_T$ and for $NbSe_3$ at $2E_T$. Threshold fields and thicknesses for all samples are listed in the legend. For symbols without error bars, the uncertainties are $\pm 10\%$. Circled symbols refer to modes for which the sample is oscillating in a direction perpendicular to the others.

for which the crystal vibrates in the perpendicular (and much thicker) direction, shown as circled data points, are consistent with the "parallel" anomalies, and so such a conclusion is dubious.[7]

For $NbSe_3$, the modulus depinning anomaly falls roughly as $\omega^{-3/4}$ for all four samples, but the magnitudes of the anomalies for sample No. 4 are half those of the others; this does not correlate with either thickness or threshold field. (We previously found that the shear-modulus anomalies were roughly proportional to threshold field.[6]) The small magnitude of the internal-friction anomalies, and resulting relatively large error bars and scarcity of data, make the frequency dependence unclear, but it seems to be similar to that of $TaS_3$, with a range of saturation frequencies from less than 5 kHz (for No. 3) to more than 20 kHz for No. 6.

Qualitative fits of the depinning anomalies by anelastic equations with relaxation times diverging at threshold[3,4] indicated that at least two very different relaxation processes were needed. In such a case, the shape of the anomalies should vary with frequency; i.e., as the frequency is increased, the changes in elastic properties should occur at higher fields. As shown in Fig. 2, such is not the case, and it does not appear that elastic relaxational models are appropriate descriptions of CDW depinning. Unfortunately, none of the present samples exhibited the peak in the internal friction most suggestive of relaxational behavior.[3,4]

As mentioned above, all proposed models for the elastic anomalies have some qualitative inconsistencies with previous low-frequency results, most of which have been discussed in Ref. 6; they also have different predictions for their frequency dependence. In the decoupling models,[8] the Young's-modulus depinning and transition anomalies are expected to be approximately equal (as usually observed at low frequency[2,3,6]) and independent of frequency. In the screening model,[9] they are also equal and should *both* disappear for phonon wave numbers $q > \Omega_p/v_F$, where $\Omega_p$ is the CDW depinning frequency and $v_F$ the Fermi velocity. For propagating flexural waves,[13,15] $\omega = tq^2(Y/12\rho)^{1/2}$, using the values $t \sim 3$ μm, $v_F \sim 5000$ km/s,[16] $\Omega_p/2\pi \sim 5$ GHz,[1] and $(Y/\rho)^{1/2} \sim 7.5$ km/s,[17] the anomalies should disappear for frequencies $\gtrsim 40$ kHz.[18] Zeyher[10] predicts that the depinning internal-friction anomaly should increase with frequency as $\omega^{1/2}$. In contrast, we observe that (i) the modulus depinning anomalies decrease with frequency, (ii) the modulus transition anomalies are independent of frequency, and (iii) the internal-friction depinning anomalies decrease with frequency below $\sim 10$ kHz and saturate at higher frequencies, in qualitative disagreement with all three models.

The fractional frequency power law observed for the modulus depinning anomaly, $|\Delta Y/Y|_{\text{depin}} \propto \omega^{-p}$, with $p \sim \frac{3}{4}$ for most samples, is reminiscent of the behavior observed for the ac conductivity of the *pinned* CDW, $\sigma \propto \omega^{p'}$, with $p' \sim 0.9$.[19] For the conductivity, the fractional power reflects CDW disorder and a wide distribution of CDW pinning frequencies.[19] The relationship of the two properties is unclear, however, especially in view of the rough frequency *independence* of the (pinned) transition anomaly.

We thank K. Maki and A. Virosztek for many illuminating discussions. This research was funded in part by the National Science Foundation (Solid State Physics Program) Grant No. DMR-86-15463.

[1]For a recent review, see G. Gruner, Rev. Mod. Phys. **60**, 1129 (1988).

[2](a) J. W. Brill and W. Roark, Phys. Rev. Lett. **53**, 846 (1984); (b) G. Mozurkewich, P. M. Chaikin, W. G. Clark, and G. Gruner, Solid State Commun. **56**, 421 (1985); (c) L. C.

Bourne, M. S. Sherwin, and A. Zettl, Phys. Rev. Lett. **56**, 1952 (1986); (d) A. Suzuki, H. Mizubayashi, and S. Okuda, J. Phys. Soc. Jpn. **57**, 4322 (1988).

[3]J. W. Brill, W. Roark, and G. Minton, Phys. Rev. B **33**, 6831 (1986).

[4]M. H. Jericho and A. M. Simpson, Phys. Rev. B **34**, 1116 (1986).

[5]X.-D. Xiang and J. W. Brill, Phys. Rev. B **36**, 2969 (1987).

[6]X.-D. Xiang and J. W. Brill, Phys. Rev. B **39**, 1290 (1989).

[7]Ronald Jacobsen and George Mozurkewich, Phys. Rev. B **39**, 10913 (1989).

[8](a) L. Sneddon, Phys. Rev. Lett. **56**, 1194 (1986); (b) M. S. Sherwin and A. Zettl, Physica (Amsterdam) **23D**, 62 (1986).

[9]K. Maki and A. Virosztek, Phys. Rev. B **36**, 2910 (1987); Synth. Met. **29**, F371 (1989).

[10]R. Zeyher, Phys. Rev. Lett. **61**, 1009 (1988).

[11]H. Fukuyama and P. A. Lee, Phys. Rev. B **17**, 535 (1978); P. A. Lee and T. M. Rice, Phys. Rev. B **19**, 3970 (1979).

[12]X.-D. Xiang, J. W. Brill, and W. Fuqua, Rev. Sci. Instrum. (to be published).

[13]L. D. Landau and E. M. Lifshitz, *Theory of Elasticity* (Pergamon, Oxford, 1986), 3rd ed.

[14]Y. Nakane, J. Phys. Soc. Jpn. **55**, 2235 (1986).

[15]Reference 9 in fact considers changes in the speed of *bulk*, longitudinal phonons with $\omega \propto q$. For such modes, both depinning and transition anomalies should decrease only at frequencies $\gtrsim 7$ MHz.

[16]J. Nakahara, T. Taguchi, T. Araki, and M. Ido, J. Phys. Soc. Jpn. **54**, 2741 (1985).

[17]J. W. Brill, Mol. Cryst. Liq. Cryst. **18**, 107 (1982).

[18]K. Maki and A. Virosztek (to be published) have also shown that the magnitudes and signs of the various elastic anomalies also depend on the relative sizes of $\omega/q^2$ and $D$, the diffusion constant (typically 1000 cm$^2$/s). For flexural waves, $\omega/q^2 < 100$ cm$^2$/s, so we are always in the "collisionless" limit.

[19]Wei-Yu Wu, L. Mihaly, George Mozurkewich, and G. Gruner, Phys. Rev. B **33**, 2444 (1986).