Elastic Properties of Charge-Density-Wave Conductors: ac-dc Electric Field Coupling

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We have measured Young's modulus Y and internal friction δ of the charge-density-wave (CDW) conductors TaS_3 and $NbSe_3$, in the presence of combined ac and dc electric fields. In the pinned CDW state, the elastic properties are sensitive to even small ac-field amplitudes. In the sliding-CDW state, sharp harmonic and subharmonic anomalies are observed in Y and δ; these coincide with Shapiro-type interference in the electronic response. Our experiments demonstrate that internal degrees of freedom (incoherence) of the CDW condensate are intimately tied to the dc- and ac-field–induced elastic anomalies.

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It has been clearly established that the anomalous low-field ac conductivity and nonlinear dc conductivity of low-dimensional materials such as TaS_3 and $NbSe_3$ are the result of collective-mode charge-density-wave (CDW) dynamics.[1] Recently, it was demonstrated[2,3] that bulk elastic properties of CDW conductors are modified by applied dc electric fields E_{dc} exceeding the threshold field E_T for the onset of nonlinear electronic conduction. In particular, an increasing CDW drift velocity results in a decrease in Young's modulus Y and a corresponding increase and eventual saturation of internal friction δ of the crystal. No detailed model of CDW elasticity has made predictions[4] consistent with these experimental findings.

We have investigated the bulk elastic properties of the CDW materials orthorhombic TaS_3 and $NbSe_3$, in the presence of combined ac and dc electric driving fields. In the limit of zero ac-field amplitude, our results are in accord with previous measurements.[2,3] In the limit of zero dc field, we find that Y is modified by the applied ac field, even in the range of very small ac amplitude where the CDW remains always pinned. In the regime of combined ac and dc electric fields, sharp anomalies are observed in Y and δ; these coincide with Shapiro-type interference[5-7] in the electronic response. We present a model of CDW transport and elasticity which includes coupling of the CDW dynamics to the underlying lattice dynamics. A computer solution of the model for ac, dc, and combined ac and dc drive fields yields behavior for elastic constants of the crystal in agreement with our experimental findings.

Our experiments were performed on single crystals of $NbSe_3$ and orthorhombic TaS_3 grown by conventional vapor transport methods, of typical dimensions 1 mm$\times$2 μm$\times$5 μm. The samples were mounted with silver-paint contacts in a two-probe "clamped-clamped" configuration. Flexural resonances were excited by capacitive coupling to the crystal, and the mechanical response was monitored by an rf detection technique using a 600-MHz carrier. The crystal formed part of a phase-locked loop which allowed simultaneous recording of the frequency ω_r and amplitude A_r of the mechanical resonance. The Young's modulus of the crystal was obtained from the resonance frequency using the relation $Y = D\omega_r^2$, where D is a constant which depends on boundary conditions and crystal dimensions and density.[2] Changes in $1/A_r$ were directly related to changes in δ. Typical resonant quality factors for crystals used in these experiments were a few hundred. Our measurement system also allowed simultaneous measurement of the differential resistance dV/dI of the sample. On occasion, signal averaging of the data was performed to improve the signal-to-noise ratio. The samples were cooled in a liquid-nitrogen gas-flow cryostat, and the gas pressure inside the sample chamber was held at 5 Torr to reduce Joule self-heating of the crystals. Details of the experimental technique will be presented elsewhere.[8]

Figure 1 shows Y, δ, and dV/dI, as functions of dc bias, for a TaS_3 crystal in the CDW state. Data for various amplitudes of the applied ac field at frequency $\omega_{ac}/2\pi = 1$ MHz are shown. Curves A correspond to no applied ac field, and here the drop in Y and rapid increase in δ for dc bias values exceeding E_T are consistent with previous studies.[2,3] As the ac field amplitude is increased from zero, Fig. 1 shows that E_T, as determined from the initial break in the dV/dI curve, is decreased to a lower value E_T', again consistent with previous conductivity studies.[5-7] Curves B, C, and D in Fig. 1 demonstrate that the depression in E_T is reflected in the behavior of Y and δ as well. Both Y and δ begin to deviate at E_T' from their zero-dc-bias values (measured with ac applied), and thus, even with an applid ac field, Y and δ are insensitive to dc bias until the CDW becomes depinned and assumes a finite dc drift velocity at E_T'. Figure 1 also shows that δ saturates more quickly, as a function of dc bias, with increasing ac field amplitude. On the other hand, Fig. 1 demonstrates that the effect of an ac field on Y and δ is *not* just to rescale the effective dc bias field E_{dc}. For example, curves A–D show that, even for zero dc bias, an increase in ac amplitude E_{ac} results in a decrease in Y and an increase in δ, although dV/dI remains un-

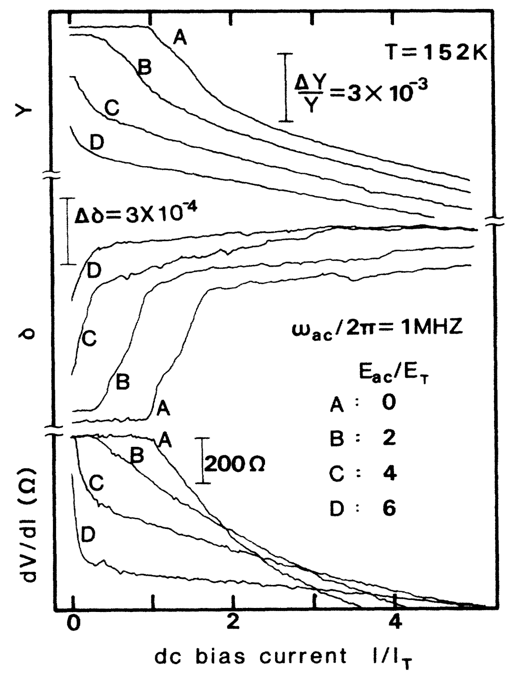

FIG. 1. Y, δ, and dV/dI as functions of dc bias in TaS_3. A 1-MHz ac field of amplitude E_{ac} is also present.

changed. It should be noted that curves $A-D$ in Fig. 1 have, within each group, *not* been displaced with respect to one another.

The detailed behavior of Y, δ, and dV/dI in the presence of an ac field alone (zero dc bias) is shown in Fig. 2. Here, for low-amplitude ac excitation, the CDW remains always pinned. Within experimental resolution, δ is roughly independent of ac amplitude for $E_{ac} < E_T$. On the other hand, Y changes smoothly as ac amplitude is increased from zero, with no evidence for a threshold behavior. Hence, in the pinned CDW state, *the elastic properties of the crystal are influenced even by low-amplitude ac dynamics of the CDW condensate.* As E_{ac} is increased from zero to E_T, there is a relative decrease in Y of approximately 5×10^{-5}. For $E_{ac}=2E_T$, an increase in δ of roughly 2×10^{-5} is observed.

An interesting consequence of superposed ac and dc electric fields in CDW conductors is Shapiro-step electronic interference, where sharp "peaks" are observed in dV/dI whenever the frequency of the ac field matches a harmonic or subharmonic of the intrinsic narrow-band noise frequency.[5-7] Shapiro-step interference is present in the dV/dI traces of Fig. 1, but somewhat difficult to identify due to the relatively poor resolution. Figure 3(a) shows the results of a very sensitive measurement of dV/dI, Y, and δ in the presence of combined ac and dc electric fields for a different TaS_3 crystal. The dominant (fundamental) Shapiro step is identified in the lower dV/dI trace with an arrow. Related anomalies are clearly observed in Y

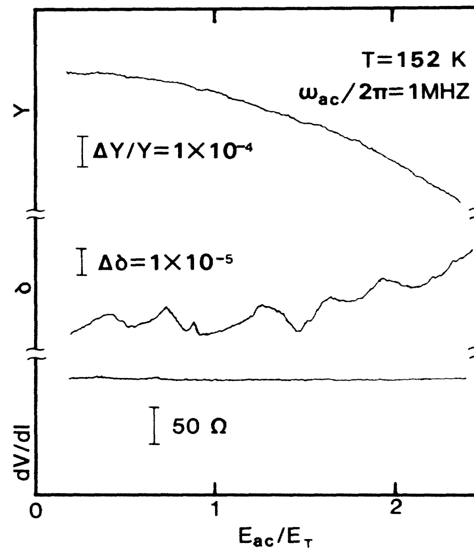

FIG. 2. Y, δ, and dV/dI as functions of ac field amplitude in TaS_3. No dc bias field is present. A low-amplitude field softens the crystal lattice.

and δ at the same values of dc bias. Figure 3(a) indicates that, during Shapiro-step electronic interference, *Y and δ tend to the values characteristic of the pinned CDW state.* This follows the behavior of dV/dI on the Shapiro step. Figure 3(b) shows similar interference data for a $NbSe_3$ crystal in the upper CDW state. Here, the interference effect in dV/dI is far more dramatic than that for TaS_3, and indeed the corresponding anomalies in Y and δ are striking. Both harmonic anomalies (identified by $n=1$, see Ref. 5) and subharmonic anomalies (identified by nonintegral values of n, see Ref. 6) are observed in Y and δ, as

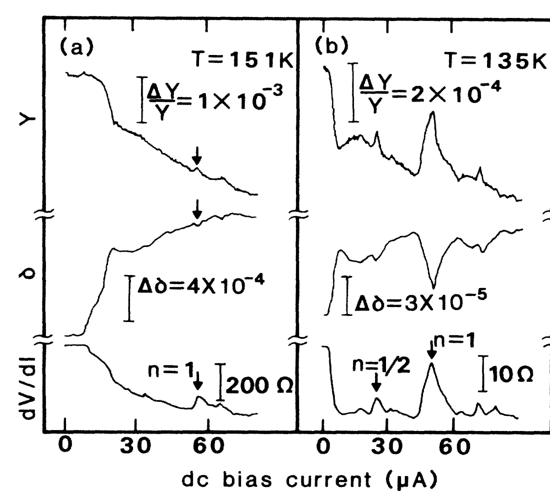

FIG. 3. Y, δ, and dV/dI as functions of dc bias. (a) TaS_3 with $\omega_{ac}/2\pi=1$ MHz, $E_{ac}/E_T=1.3$. (b) $NbSe_3$ with $\omega_{ac}/2\pi=2$ MHz, $E_{ac}/E_T=3.8$. The arrows identify interference structure for TaS_3.

well as in dV/dI. Measurements on other TaS_3 and $NbSe_3$ crystals indicate that there exists a direct correlation between the "strength" of the Shapiro-step electronic interference (i.e., the height of the peak in dV/dI, during interference, relative to the low-field resistance) and the size of the anomalies in Y and δ during interference. For "complete" electronic mode locking,[7] we expect Y and δ to attain exactly their zero field, $E_{dc}=E_{ac}=0$, values.

Figures 3(a) and 3(b) clearly demonstrate that the detailed behavior of Y and δ in the depinned CDW state does *not* depend only on the average CDW drift velocity. If that were the case, no anomalies (other than at best slight "plateau" structure) would be observed in Y and δ during Shapiro-step interference. Hence, ac-dc interference corresponds to more than simple electronic locking of the CDW drift velocity; there result profound changes in the elastic properties of the CDW crystal as well. As we discuss below, this appears to be related to a loss of internal degrees of freedom for the CDW condensate during interference, as has been suggested to account for unusual broadband-noise suppression on mode-locked Shapiro steps.[7]

Although a variety of models[1] have been advanced to account for the unusual properties of CDW conductors, most have been applied only to the electronic response. A difficulty in applying such models to the elastic response is that they in general treat the impurity pinning potential as immobile, and hence exclude the possibility of elastic-mode interactions between the underlying lattice and the dynamics of the CDW condensate. No model of CDW elasticity has been successful in accounting for the observed changes in Y and δ due to CDW depinning. Coppersmith[4] has considered the situation of a rigid CDW sliding through a deformable lattice. Although an anisotropic shift in the velocity of sound is obtained, the predicted effects are orders of magnitude smaller than elastic changes determined (at low frequency) by experiment.[2,3] This indicates that the CDW must be treated as deformable. Brill and Roark[2] suggest that the changes in Y and δ, due to CDW depinning, may reflect CDW-domain-wall relaxation, with the relaxation time decreasing with increasing CDW current. On the other hand, the large magnitude of the observed changes in δ would appear to rule out simple relaxation processes. Mozurkewich, Chaikin, Clark, and Grüner[3] have proposed that when the CDW is pinned to the lattice, its stiffness will add to the crystal stiffness, while a depinned CDW will not contribute to crystal stiffness. This approach yields an (order of magnitude) estimate that, changes in Y upon depinning should be comparable to CDW stiffness (computed, for example, from the Fukuyama-Lee model[9]), in rough agreement with experiment.[2,3]

Our experimental results appear to have a simple physical interpretation which points to the importance of inclusion of internal degrees of freedom in the description of elastic properties of CDW materials. Figures 1 and 2 demonstrate clearly that lattice softening in TaS_3 is not solely a result of CDW depinning and increasing CDW drift velocity. The strong E_{ac} dependence on Y for a pinned CDW shows that an ac field tends to decouple the CDW from the lattice. If an additionally applied dc field E_{dc} is increased past E_T', the CDW slides, adding further to the decoupling process. Naively, one might therefore expect that, on a mode-locked Shapiro step (where the CDW velocity is determined strictly by the external ac field frequency), the CDW dynamics would fully *decouple* from the underlying lattice, thereby allowing Y and δ during mode lock to attain their high-field (saturated) values. As previously discussed and demonstrated by Figs. 3(a) and 3(b), however, exactly the *opposite* effect occurs in TaS_3 and $NbSe_3$, i.e., on a model-locked step, the CDW couples *more* strongly to the lattice, and Y and δ tend to their *low-field* values. This effect implies that it is in fact CDW incoherence which is responsible for the dramatic changes in Y and δ due to CDW depinning.

On a mode-locked step, the electronic CDW drift velocity is fixed by the frequency ω of the external ac electric field, with $\omega >> \omega_r$. The anomalies in Y and δ indicate that the mode-locked CDW inhibits distortion of the lattice, presumably through a frictional coupling between the CDW and lattice. The connection between CDW electronic mode locking and elastic anomalies is well illustrated by a simple extension of the Grüner-Zawadowski-Chaikin (GZC) model[10] of CDW transport, where the CDW is represented by a rigid particle in a periodic potential. The GZC equation of motion for the CDW is

$$m\, d^2r/dt^2 + \gamma\, dr/dt + QV \sin(Qx) = e[E_{dc} + E_{ac}\cos(\omega_{ac}t)], \tag{1}$$

where m is the CDW mass, γ the CDW damping constant, $Q=2\pi/\lambda$ with λ the CDW wavelength, and V the strength of the impurity pinning potential. Equation (1) describes well the electronic Shapiro-step interference effects,[5] but cannot describe any elastic properties, as the lattice and CDW are here considered fully rigid. Allowing for CDW internal degrees of freedom and lattice elasticity, Eq. (1) may be extended to a simple set of coupled equations for the CDW *and* the lattice,

$$m^* d^2r/dt^2 + \gamma_C d(r-x)/dt + k_C r + eE_T \sin[2k_F(r-x)] = e[E_{dc} + E_{ac}\cos(\omega_{ac}t)], \tag{2}$$

$$M_L d^2x/dt^2 + \Gamma_L dx/dt + \gamma_C d(x-r)/dt + K_L x + eE_T \sin[2k_F(x-r)] = F\cos(\omega_r t), \tag{3}$$

where r and x are respectively the positions of the CDW center of mass and lattice. m^* is the total CDW effective mass, M_L the lattice mass, γ_C and Γ_L respectively the total CDW damping and internal lattice friction, and k_F is the Fermi wave vector. k_C and K_L parametrize respectively the total elasticity of the CDW and underlying lattice, and $F\cos(\omega_r t)$ is the mechanical force applied to the lattice which drives the resonance in the displacement x at frequency ω_r.[11] In the limit of infinite lattice mass and a loss of internal degrees of freedom, i.e., $M_L \to \infty$, k_C, $K_L \to 0$, Eqs. (2) and (3) reduce directly to Eq. (1), the rigid-particle GZC model.

In the framework of Eqs. (2) and (3), the behaviors of the elastic constants have a simple interpretation. For example, with $E_{ac}=0$ and finite $E_{dc} < E_T$, both k_c and K_L contribute to Y (there being an approximate constraint $r \approx x$) and hence Y is large. For $E_{dc} >> E_T$, only K_L contributes, and hence Y is small. Similarly, during mode locking (finite E_{ac} and E_{dc}), $\langle \dot{r} - \dot{x} \rangle$ is a constant, and hence k_c and K_L again contribute and Y is large. Indeed, detailed computer solutions[12] of Eqs. (2) and (3) show behavior for dV/dI, Y, and δ in striking qualitative agreement with the experimental results reported here for TaS_3 and $NbSe_3$, both for the cases of individually applied ac and dc electric fields, and combined ac and dc fields (mode locking). This supports further our conclusion that CDW incoherence is central to the behavior of CDW elastic properties, and that the unusual elastic anomalies associated with CDW depinning cannot be treated in any model in which $\langle \dot{r} \rangle$, the CDW drift velocity, alone dictates Y and δ.

Finally, we note that the strong coupling here implied between elastic and electronic response properties, and internal degrees of freedom, may have ramifications on the correct interpretation of Shapiro-step *electronic* interference in CDW conductors.

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[11]We here regard Eqs. (2) and (3) as phenomenological extensions of Eq. (1), where the CDW and lattice stiffness have been "lumped" into single parameters k_C and k_L, respectively. On a firmer theoretical footing, Eqs. (2) and (3) may be derived from a sine-Gordon model with discretized CDW and lattice units (M. S. Sherwin and A. Zettl, unpublished). Also note that, in order for the CDW to slide continuously through the lattice and to preserve the periodicity of the pinning potential seen by the CDW, k_C in Eq. (2) is restricted to respond only to ac excitations.

[12]Sherwin and Zettl, Ref. 11.