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MODEL OF CHARGE DENSITY WAVE ELASTICITY

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We present a simple model of the elasticity of charge density wave conductors. The model is an extension of the Frenkel–Kontorova model which introduces elasticity into the normally rigid sinusoidal potential. All of the anomalies in Young's modulus and internal friction observed experimentally as a function of dc and ac applied electric fields, including those arising from ac–dc induced electronic mode locking, are qualitatively reproduced by this model.

Much theoretical work has been devoted in the past few years to understanding the novel electronic properties of several classes of charge density wave (CDW) conductors, for example TaS_3, $NbSe_3$, $(TaSe_4)_2I$ and $K_{0.3}MoO_3$ [1]. These materials undergo a phase transition from the metallic to the CDW state at temperatures as high as 265 K. In the CDW state, the electrical conductivity becomes field and frequency dependent, and a host of related nonlinear phenomena are exhibited. The unusual transport behavior is attributed to the collective response of the macroscopic CDW, and various phenomenological classical and quantum models have been suggested to account for the electronic aspects of CDW dynamics [1]. Recent experimental work by Brill and Roark [2] and Mozurkewich et al. [3] has shown that the *elastic* as well as the electronic properties of the CDW crystal are sensitive to applied electric fields. In particular, the Young's modulus (Y) and internal friction δ of the crystal are dramatically affected by CDW depinning induced by applied dc electric fields E_{dc} exceeding the threshold electric field E_T. Very recent experiments [4], show striking anomalies in Y and δ when the CDW is excited by combined dc and ac electric fields, i.e. for $E = E_{dc} + E_{ac}\cos(\omega t)$.

Current theories of CDW transport fail to account for the elastic properties of CDW's. We present here a classical phenomenological model which reproduces qualitatively all of the observed elastic anomalies of CDW conductors, including those arising from coupled ac and dc fields.

First we give a brief review of relevant sliding CDW phenomenology. When a dc electric field $E_{dc} > E_T$ is applied to a CDW crystal, the dc conductivity becomes field dependent, with the (dc) differential conductivity increasing smoothly. The current carried by the CDW is time dependent, with a coherent component called the narrow band noise (NBN) with fundamental frequency directly proportional to the CDW current (and hence to CDW drift velocity), and a broad band component with a $1/f$ like spectrum in the 0–100 kHz range. If large ac and dc electric fields are applied simultaneously to a CDW crystal, one observes interference effects (analogous to the Shapiro steps observed in Josephson junctions) between the NBN and the external ac field [5]. On such a step, the NBN frequency, and hence the CDW velocity, lock to the frequency of the external ac field over a finite range of dc bias, causing a dramatic increase in the differential resistance dV/dI.

Many CDW crystals (such as $NbSe_3$ and TaS_3) are thin, flexible fibers, and the elastic constants are thus most conveniently measured by exciting a flexural resonance and measuring the resonant frequency ω_r and "Q" of the mechanical response. Y and δ are determined directly from ω_r and Q.

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Original studies on TaS_3 by Brill and Roark [2] showed that, for dc fields $E_{dc} > E_T$, Y decreases smoothly and δ increases rapidly and saturates.

In more recent experiments by Bourne, Sherwin and Zettl [4], Y and δ were measured in the presence of ac fields alone and also in the presence of combined ac and dc fields. For ac fields only, Y was found to decrease even for small ac amplitudes $E_{ac} < E_T$. For combined ac and dc electric fields Shapiro steps in the electronic response were found to coincide with sharp anomalies in the elastic constants (fig. 3b). This is a very important result. If Y and δ were simply functions of average CDW velocity [2], then one would expect that when the CDW is electronically mode locked on a Shapiro step (and hence its drift velocity is constant) Y and δ would simply remain constant over the locked range of E_{dc}. Instead, during mode locking, Y and δ are observed to tend to their *low* field values.

A number of models have been proposed for the electronic properties of CDW conductors. The simplest of these is a "rigid particle" model first introduced by Grüner, Zawadowski and Chaikin [6] (GZC). In this highly oversimplified description, the CDW is represented by a single rigid particle in a rigid periodic potential. The equations of motion are isomorphic to those of the driven damped pendulum model or the resistively shunted Josephson junction in the RSJ approximation. This model exhibits a threshold field, narrow band noise for $E_{dc} > E_T$ and Shapiro steps for combined ac and dc electric fields. A most important *failure* of the GZC model is that it predicts a divergence in the dc differential conductivity near threshold which is not observed experimentally. The model says nothing about the elasticity of the CDW or lattice, since all entities are assumed rigid.

Many of the difficulties of the GZC model can be remedied by including internal degrees of freedom for the CDW [7, 9, 10]. Such degrees of freedom may be introduced by considering the CDW as composed of harmonically coupled discrete particles in a rigid potential, and the equations of motion resemble those of the Frenkel–Kontorova model [8], or the discretized sine-Gordon equation. These models are an improvement over GZC, and exhibit no divergence of the differential conductivity at threshold in the thermodynamic limit [9]. However, in these models lattice elasticity is again absent, since the potential in which the CDW moves is considered rigid.

In the first attempt to calculate the elastic properties of CDW crystals, Coppersmith and Varma considered a rigid CDW sliding through a deformable lattice [10]. Although an anisotropy was predicted for the velocity of sound, the effects are orders of magnitude smaller than the experimentally observed changes in Y and δ due to CDW depinning.

We present a classical model in which the CDW and the lattice are treated as coupled elastic media. This model is an extension of the Frenkel–Kontorova model, and we have incorporated elasticity into the sinusoidal potential (and hence the underlying lattice) by discretizing it into rigid units of mass M coupled harmonically by springs with spring constant K. The CDW is represented by discrete particles of mass m coupled harmonically to nearest neighbors by a spring constant κ. The mechanical analog of this model is shown schematically in fig. 1. This model can describe both commensurate and incommensurate cases, where in the former case the wavelength of the sinusoidal potential is equal to the equilibrium spacing of the particles in the lattice and CDW. The potential energy function for this system, assuming only nearest neighbor interactions, is

$$V = \sum_j \Big\{ \kappa/2(r_j - r_{j-1})^2 + K/2(x_j - x_{j-1})^2 + V\big(1 - \cos\left[Q(r_j - x_j)\right]\big)\Big\}, \qquad (1)$$

where r_j and x_j are respectively the (laboratory frame) positions of the jth CDW mass and jth lattice unit, V is the strength of the pinning potential and $Q = 2\pi/\lambda$ with λ the CDW wavelength. Applying Lagrange's equations, and adding inter-

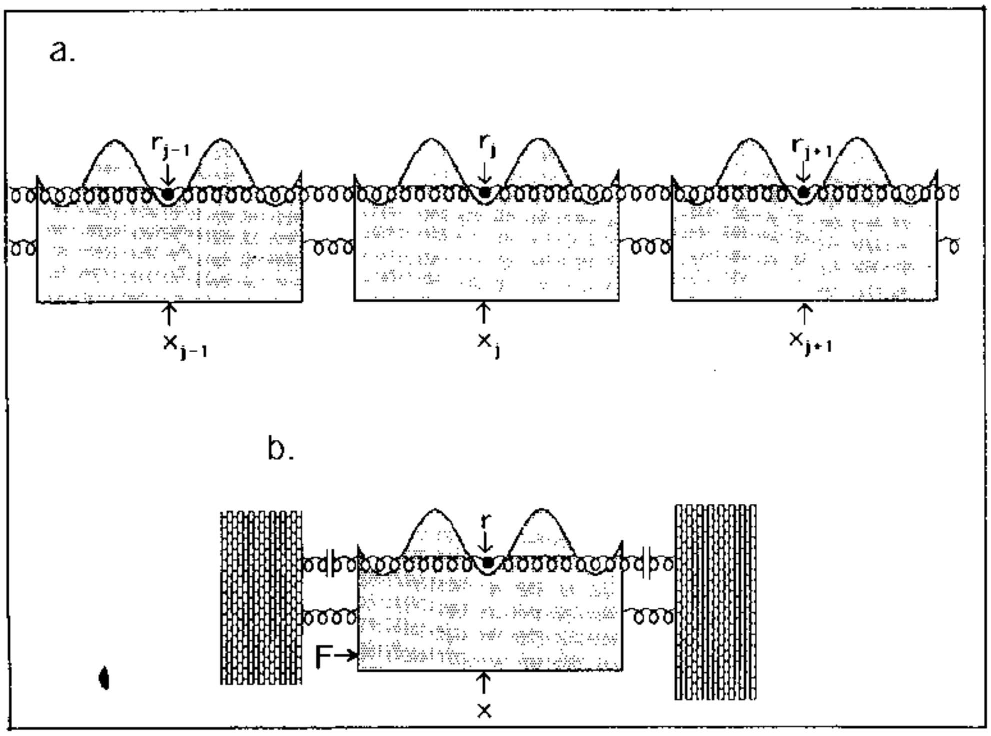

Fig. 1. Mechanical analog of our model, (a) for the infinite case and (b) showing the boundary conditions applied to reduce it to eqs. (4) and (5). The break in the CDW springs in (b) signifies that they respond only to ac excitations, thus allowing the CDW to slide. Y and δ are determined from the resonant frequency and amplitude of the response of x to the mechanical force F.

nal friction and forcing we derive the following equations of motion:

$$m\,\mathrm{d}^2r_j/\mathrm{d}t^2 + \gamma\,\mathrm{d}(r_j - x_j)/\mathrm{d}t + \kappa(2r_j - r_{j+1} - r_{j-1}) + QV\sin\left[Q(r_j - x_j)\right] = f(t), \tag{2}$$

$$M\,\mathrm{d}^2x_j/\mathrm{d}t^2 + \Gamma\,\mathrm{d}(2x_j - x_{j-1} - x_{j+1})/\mathrm{d}t + \gamma\,\mathrm{d}(x_j - r_j)/\mathrm{d}t + K(2x_j - x_{j-1} - x_{j+1}) + QV\sin\left[Q(x_j - r_j)\right] = F_j(t), \tag{3}$$

where Γ is the internal friction of the lattice, and γ is a frictional coupling between the CDW and lattice. f is the force applied to the CDW by external electric fields and F_j the mechanical forces applied to the lattice units. In the limit $K \to \infty$, eqs. (2) and (3) reduce to the discretized sine-Gordon equation [11].

These equations are analytically intractable, except if one considers small amplitude excitations and linearizes them [12]. We reduce the infinite set of eqs. (2) and (3) to the smallest set of equations that retain the essential physics of an elastic CDW interacting with an elastic lattice. This is accomplished by applying boundary conditions schematically illustrated in fig. 1b. The lattice is reduced to a single particle with its nearest neighbors nailed to the laboratory frame. The CDW is represented by a particle whose nearest neighbors are nailed to the CDW center of mass frame. The resulting equations of motion are

$$m^* \, \mathrm{d}^2 r/\mathrm{d}t^2 + \gamma_C \, \mathrm{d}(r-x)/\mathrm{d}t + k_C r + eE_T \sin[2k_F(r-x)] = e[E_{dc} + E_{ac}\cos(\omega t)], \quad (4)$$

$$M \, \mathrm{d}^2 x/\mathrm{d}t^2 + \Gamma \mathrm{d}x/\mathrm{d}t + \gamma_C \, \mathrm{d}(x-r)/\mathrm{d}t + K_L x + eE_T \sin[2k_F(x-r)] = F\cos(\omega_r t), \quad (5)$$

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 the Fermi wavevector. k_C and K_L parameterize respectively the total elasticity of the CDW and underlying lattice, and $F\cos(\omega_r t)$ is the mechanical force applied to the lattice. In order to allow the CDW to slide continuously through the lattice and retaining the periodicity of the pinning potential, we impose the restriction that k_C responds only to ac excitations.

We have solved eqs. (4) and (5) for a variety of dc and ac drives on an analog electronic computer built in our laboratory, keeping the mechanical force in eq. (5) small. The Young's modulus and internal friction are derived from the resonant frequency and amplitude of the response of x at ω_r: at the mechanical resonance, $Y \propto (\omega_r)^2$, and $\delta \propto (1/\text{amplitude}(\omega_r))$.

The results of our simulation with $E_{ac} = 0$ are presented in Fig. 2a. The parameters used are (in relative units) $eE_T = 0.76 \times 10^{-3}$, $2k_F = 6.28 \times 10^4$, $k_C = 2.85$, $K_L = 29.4$, $\gamma_C = .95 \times 10^{-3}$, $\Gamma_L = 10^{-3}$, $m^* = 4 \times 10^{-11}$, $M_L = 2 \times 10^{-5}$, and $\omega_r/2\pi \approx 200$. Fig. 2a shows that for $E_{dc} < E_T$, Y and δ are only weakly field dependent. For $E_{dc} \gg E_T$, Y saturates at a value smaller than for the pinned state, and δ saturates at a value larger than for the pinned state. These findings are consistent with experimental results displayed in fig. 2b. Discontinuous changes in $\mathrm{d}V/\mathrm{d}I$, Y and δ are evident in fig. 2a as E crosses E_T. Such divergences near

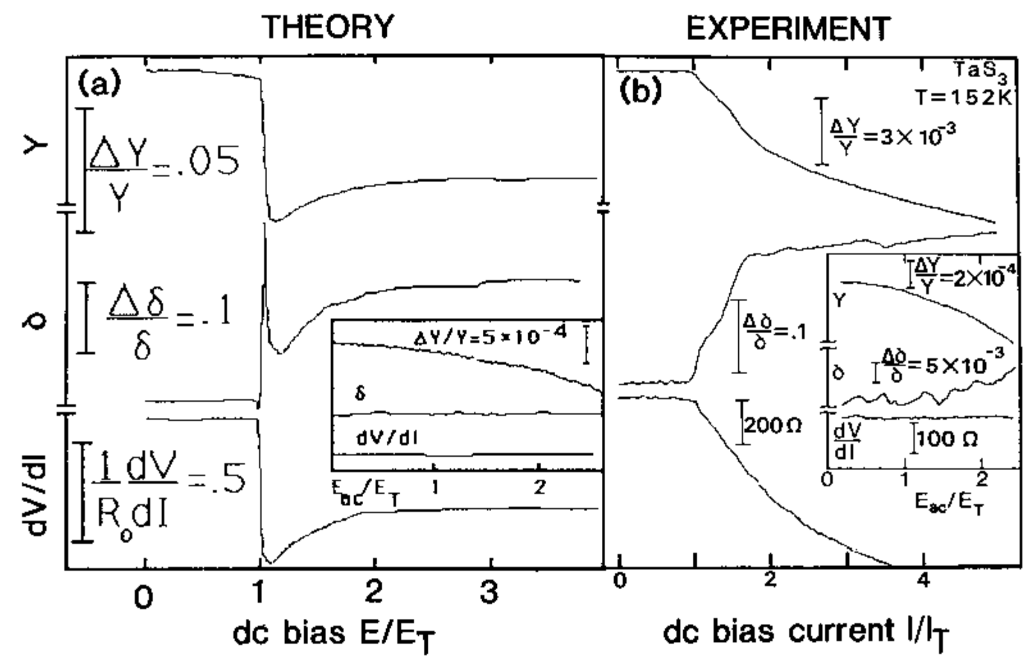

Fig. 2. Y, δ and $\mathrm{d}V/\mathrm{d}I$ as functions of dc bias for $E_{ac} = 0$ (a) as calculated from eqs. (4) and (5) and, (b) as measured in TaS_3 [4]. The insets show Y, δ and $\mathrm{d}V/\mathrm{d}I$ as functions of ac amplitude for $E_{dc} = 0$ (a) as calculated from eqs. (4) and (5), and (b) as measured in TaS_3 [4].

threshold are endemic to finite size classical models (such as GZC, mentioned above), and this suggests that eqs. (4) and (5) cannot be applied to Y and δ for E_{dc} close to E_T. We also note that the model parameters we have chosen compromise between those expected for a real CDW crystal and those accessible to our analog computer. In particular, the absolute size of the changes in Y due to CDW depinning are sensitive to the ratio k_C/K_L. A more realistic (smaller) value of this ratio would result in smaller elastic changes, consistent with experimental findings.

We have also solved eqs. (4) and (5) in the range of finite E_{ac}, with $\omega/\omega_r = 20$. The inset of fig. 2a illustrates that, for $E_{dc} = 0$, increasing E_{ac} from zero results in a smooth decrease in Y, and, within computational resolution, no detectable change in δ for very low E_{ac}. These results are in agreement with the experimental results for TaS_3 under similar conditions, shown in the inset of fig. 2b.

For finite E_{dc} and E_{ac}, eqs. (4) and (5) predict complete Shapiro step electronic locking, as shown in the dV/dI trace of fig. 3a. Also shown in the figure are Y and δ, calculated for the same set of drive parameters. It is clear that Shapiro step interference in the electronic response corresponds

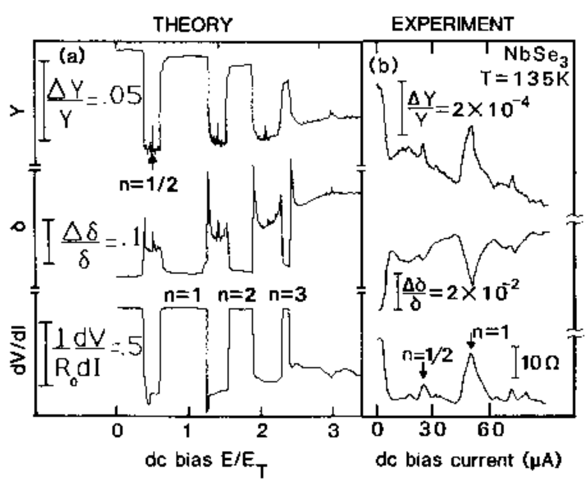

Fig. 3. Y, δ and dV/dI as functions of dc bias (a) as calculated from eqs. (4) and (5) with $E_{ac}/E_T = 3$, $\omega/\omega_r = 20$, and (b) as measured in $NbSe_3$ [4] with $\omega/2\pi = 2$MHz and $E_{ac}/E_T = 3.8$. The arrows identify interference structure.

in our model with striking anomalies in the elastic constants. In the Shapiro step region, both Y and δ tend to their respective values measured for $E_{dc} = 0$, as observed experimentally in fig. 3b. We also note the presence of harmonic and subharmonic structure in the calculated Y and δ, as observed experimentally.

The behavior of the elastic constants in our model has a simple interpretation. When $E_{ac} = 0$ and $E_{dc} < E_T$, there is a strong restoring force (with "spring constant" $2k_F eE_T \gg k_C$) due to the sinusoidal potential that forces $(x - r) \approx 0$. Thus the mechanical force acting on x must compress both springs k_C and K_L. When $E \gg E_T$ and the CDW is sliding, the restoring force due to the sinusoidal potential averages to zero (because the narrow band noise frequency $\gg \omega_r$), so a force applied to x compresses only K_L, and Y drops from its pinned value. Lifting the constraint $x - r \approx 0$ also increases the internal friction because now the damping term in eqs. (4) and (5) $\gamma_C\, d(x - r)/dt \neq 0$.

In the presence of both dc and ac fields, if the CDW is mode-locked, there is again an approximate constraint: $\langle d(r - x)/dt \rangle \approx$ constant if the average is taken over 1 cycle of the ac drive. Since $\omega \gg \omega_r$, the constraint is effective against the mechanical force and both k_C and K_L contribute to Y during mode-locking. However, when the CDW is unlocked the constraint vanishes and, as for a dc depinning, Y decreases and δ increases. On careful examination, it is apparent from fig. 3b that the constraint during mode-locking is not perfectly effective, since Y and δ do not return completely to their $E_{dc} = 0$ values despite the electronic mode locking being "complete" [13].

In conclusion, we have presented the simplest model that contains the essential features of a pinned, elastic CDW interacting with an elastic lattice, and all of the experimental results, with the exception of behavior near threshold, are reproduced. The divergences near E_T will almost certainly be remedied by the inclusion of more degrees of freedom in the reduction from the infinite set of eqs. (2) and (3).

The interaction of the CDW with the lattice is an area that requires further investigation. In fact, the Frenkel–Kontorova model has been applied to many condensed matter systems, notably superionic conductors, adsorbates on surfaces and 1-D magnetism [14]. In all of these systems, the assumption of a rigid potential is unrealistic. The extension of the Frenkel–Kontorova, or discretized sine-Gordon equation to include an elastic potential [15] is relevant to a wide variety of systems.

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References

[1] For a review, see G. Grüner and A. Zettl, Phys. Reports 119 (1985) 117.

[2] J.W. Brill and W. Roark, Phys. Rev. Lett. 53 (1984) 846.

[3] G. Mozurkewich, P.M. Chaikin, W.G. Clark and G. Grüner, in Charge Density Waves in Solids, Gy. Hutiray and J. Solyom, eds. (Springer, New York, 1985) p. 353.

[4] L.C. Bourne, M.S. Sherwin and A. Zettl, Phys. Rev. Lett. 56 (1986) 1952.

[5] A. Zettl and G. Grüner, Phys. Rev. B 29 (1984) 755; P. Monceau, J. Richard and M. Renard, Phys. Rev. Lett. 45 (1980) 43.

[6] G. Grüner, A. Zawadowski and P.M. Chaikin, Phys. Rev. Lett. 46 (1981) 511.

[7] J.E. Sacco and J.B. Sokoloff, Phys. Rev. B 18 (1985) 6549; L. Sneddon, Phys. Rev. Lett. 52 (1984) 65; L. Sneddon, Phys. Rev. B 30 (1984) 2974.

[8] J. Frenkel and T. Kontorova, Zh. Eksp. Theor. Fiz. 8 (1938) 1340; J. Phys. (USSR) 1 (1939) 137.

[9] D.S. Fisher, Phys. Rev. Lett. 50 (1983) 1486.

[10] S.N. Coppersmith, in Charge Density Waves in Solids, Gy. Hutiray and J. Solyom, eds. (Springer, New York, 1985) p. 206.

[11] S.N. Coppersmith and D.S. Fisher, Phys. Rev. B 28 (1983) 2566 and references therein.

[12] M.S. Sherwin and A. Zettl, unpublished.

[13] M.S. Sherwin and A. Zettl, Phys. Rev. B 32 (1985) 5536.

[14] L. Pietronero and S. Strässler, Solid State Commun. 27 (1978) 1041;
P. Bak, Rep. Prog. Phys. 45 (1982) 587;
A.R. Bishop, J. Phys. A 14 (1981) 883.

[15] Note that our discretization of the lattice and sinusoidal potential is distinctly different from introducing a deformable substrate into the Frenkel–Kontorova model. The latter approach is considered by T. Munakata, J. Phys. Soc. Japan 52 (1983) 1635, and T. Ishii, J. Phys. Soc. Japan, 52 (1983) 168.