Elastic properties of charge-density-wave conductors in combined ac and dc electric fields

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We calculate the Shapiro-type anomalies in the Young's modulus Y and internal friction δ of charge-density-wave conductors that are in combined ac and dc electric fields in the framework of perturbation theory. We use the classical model of two incommensurate, interacting, many-body systems proposed by Sneddon. It is found that this model produces the experimental results well at weak pinning. Some experiments are suggested.

The properties of charge-density-wave (CDW) conductors have been extensively[1] studied both by experimentalists and theorists. The widespread interest has not only focused on their nonlinear electrical properties, but also on their elastic properties, which depend sensitively on the motion of CDW, of the underlying crystal. The first measurements of the elastic properties of CDW conductors were carried out by Barmatz, Testardi, and DiSalvo.[2] They used vibrating-reed techniques[2,3] to measure the Young's modulus and the internal friction of $2H$-$TaSe_2$ and $2H$-$NbSe_2$ samples. Anomalies were found at the transition temperature T_0 of charge-density waves. These anomalies are due to a structural change of the underlying lattice at the transition temperature.

More recently, by using a modified vibrating-reed apparatus, Brill and Roark[4] and Mozurkewich *et al.*[5] demonstrated that bulk elastic properties of CDW conductors could be modified by an applied dc electric field E if it exceeded the threshold field E_T for the onset of nonlinear electronic conduction. In particular, an increasing CDW drift velocity resulted in a decrease in the Young's modulus Y and a corresponding increase of internal friction δ of the crystal. Bourne, Sherwin, and Zettl[6] also found sharp Shapiro-type anomalies in Y and δ in the presence of combined ac and dc electric driving fields.

The classical model[7] of two mutually incommensurate, interacting, dynamical, many-body systems proposed by Sneddon predicts the main experimental results of the elastic properties of CDW conductors in the electric dc fields. This model suggests that the internal CDW degrees of freedom play important roles in determining the elastic behaviors of CDW conductors. In this paper, we will examine whether Sneddon's model can predict the elastic properties of CDW conductors in combined ac and dc fields. We also explore the connections between ac-dc nonlinear interferences and Shapiro-type anomalies in Y and δ.

The classical model proposed by Sneddon consists of two mutually interacting incommensurate chains, one representing the CDW and the other representing the host crystal lattice. Each one distorts the other. Conservative and viscous interactions between the chains are used to represent the pinning forces that produce the CDW threshold and the dissipations that result from the CDW motion through the crystal, respectively. The dimensionless equation of motion can be written as

$$\begin{aligned} m\ddot{\Phi}_\alpha+\sum_\beta \Delta_\beta\Phi_{\alpha-\beta}+\mu\sum_j W(X_{\alpha,j})(\dot{\Phi}_\alpha-\dot{U}_j) &= -\mu\sum_j F(X_{\alpha,j})-E_L\ , \\ \sum_p D_p U_{j-p}+\sum_\alpha W(X_{\alpha,j})(\dot{U}_j-\dot{\Phi}_\alpha) &= \sum_\alpha F(X_{\alpha,j})+E\ , \end{aligned} \tag{1}$$

where m is the ionic mass. The inertia of the CDW is negligible, Φ_α and U_j are the displacements of particle α in the crystal lattice and particle j in the CDW, respectively, and $2\pi\alpha$ and Hj are their respective undisplaced positions. The coefficients Δ_β and D_p are the spring constants of the internal, harmonic restoring forces in the lattice and of the CDW, respectively. The function $W(x)$ is a weight function and represents the spatial range of the dissipative interactions. The force F between particle α and j depends on their separation $X_{\alpha,j}$. $E=E_0+E_1\sin(\omega t)$ is a combined ac and dc electric field acting on the CDW and $-E_L$ is the force that keeps the lattice stationary. The incommensurate limit is approached by considering M particles in the lattice chain, N in the CDW chain, and $M/N=H/2\pi\rightarrow$a fixed irrational, for example, $(\sqrt{5}-1)/2$. The parameter μ is a formal expansion parameter and will set to be 1 ultimately.

A general result of these equations, pointed out by Sneddon,[7] is that stationary solutions and the relative motion of the centers of mass of the two interacting chains could be described by the quasiperiodic forms

$$\begin{aligned} &\Phi_\alpha(t)=Q(y),\quad U_j(t)=vt+G(x)\ , \\ &y=2\pi\alpha-vt,\quad x=Hj-vt\ , \\ &Q(y+H)=Q(y),\quad G(x+2\pi)=G(x)\ , \end{aligned} \tag{2}$$

where $v=\langle \dot{U}_j \rangle$, the CDW center-of-mass velocity. Furthermore, the linear fluctuations of the dual interacting system, both static and stationary, are likewise characterized by the quasiperiodic forms similar to those given by Eq. (2).

In the stiff-lattice limit, i.e., $\Delta \gg F$ and $\Phi_\alpha=\langle \Phi_\alpha \rangle$, the crystal lattice simply produces a rigid incommensurate potential through which the CDW moves. Then Eq. (1) reduces to the equation for a single incommensurate chain that describes the motion of the CDW; some of its properties have been studied elsewhere.[8-11]

For comparison with experimental results, one should apply an external force $f_\alpha e^{-i\omega_0 t}$ on the lattice. In general, this force is small compared to Δ. One can then use perturbation theory to calculate δf and obtain the Young's modulus Y and internal friction δ. Expanding the equation of motion about the stiff-lattice limit by replacing

$$U_j \to U_j + u_j e^{-i\omega_0 t} ,$$
$$\Phi_\alpha \to \Phi_\alpha + \phi_\alpha e^{-i\omega_0 t} , \tag{3}$$
$$-E_L \to -E_L + f_\alpha e^{-i\omega_0 t} ,$$

we obtain the equations for u_j and ϕ_α:

$$m\omega_0^2\phi_\alpha + f_\alpha - \sum_\beta \Delta_\beta \phi_{\alpha-\beta} = -\mu \sum_j [F'(X^0_{\alpha,j}) - i\omega_0 W(X^0_{\alpha,j})](\phi_\alpha - u_j) + W(X^0_{\alpha,j})(\dot{\phi}_\alpha - \dot{u}_j) , \tag{4}$$

$$u_j - \langle u_j \rangle = \sum_\alpha [W'(X^0_{\alpha,j})(\dot{U}_j - \dot{\Phi}_\alpha) - F'(X^0_{\alpha,j}) + i\omega_0 W(X^0_{\alpha,j})](\phi_\alpha - u_j) - W(X^0_{\alpha,j})\dot{u}_j , \tag{5}$$

where $F'(x)$ and $W'(x)$ are the derivatives of the force $F(x)$ and weight function $W(x)$, respectively, and the equation of motion for the CDW is

$$U_j - \langle U \rangle + \sum_\alpha W(X^0_{\alpha,j})(\dot{U}_j - \dot{\Phi}_\alpha) = \sum_\alpha F(X^0_{\alpha,j}) + E_0 + E_1 \sin(\omega t) , \tag{6}$$

where ω_0 is the frequency of the linear fluctuations and

$$X^0_{\alpha,j} = X_{\alpha,j} - (\phi_\alpha - u_j)e^{-i\omega_0 t} = 2\pi\alpha + \Phi_\alpha - (Hj + U_j) \tag{7}$$

is the separation between particle α and j without the fluctuations. In the rigid-crystal-lattice limit, i.e., $\Phi_\alpha = \langle \Phi_\alpha \rangle = 0$, for the case $\phi_\alpha = e^{i2\pi\alpha q}$,[13] we have

$$U(x,t) - \langle U \rangle + \sum_\alpha W(x_\alpha)\dot{U}(x,t) - \sum_\alpha F(x_\alpha) = E_0 + E_1 \sin\omega t , \tag{8}$$

$$u(x,t) - \langle u \rangle + \sum_\alpha \{ W(x_\alpha)\dot{u}(x,t) - [W'(x_\alpha)\dot{U}(x,t) - F'(x_\alpha) + i\omega_0 W(x_\alpha)]u(x,t) \} = -\sum_\alpha [W'(x_\alpha)\dot{U}(x,t) - F'(x_\alpha) + i\omega_0 W(x_\alpha)] e^{i2\pi\alpha q} , \tag{9}$$

and

$$\delta f \equiv m\omega_0^2 + f - \Delta(q) = \frac{\mu}{M}\sum_{\alpha,j} \{ [F'(X^0_{\alpha,j}) - i\omega_0 W(X^0_{\alpha,j})](1 - u_j e^{-i2\pi\alpha q}) - W(X^0_{\alpha,j})\dot{u}_j e^{-i2\pi\alpha q} \} , \tag{10}$$

where $x = Hj$ and $x_\alpha = 2\pi\alpha - x - U(x,t)$. If the drift velocity v of the CDW is very large compared to the pinning force $F(x)$, one could solve Eqs. (8) and (9) perturbatively. For the sake of simplicity, we will consider the case $W(x) = 1/N$. Rewrite $U(x,t)$ as

$$U(x,t) = \frac{N}{M}\left[vt - \frac{E_1}{\omega}\cos\omega t \right] + g(x,t) .$$

Equation (8) becomes

$$\sum_p \frac{1}{M}[g(x,t) - g(x+Hp,t)] + \frac{M}{N}\dot{g} = \sum_\alpha F(x_\alpha) + E_0 - v . \tag{11}$$

It is obvious that $g(x+2\pi,t) = g(x,t)$; we can express $g(x)$ in terms of its Fourier components and have

$$g(x,t) = \sum_m g_m(t) e^{imx} ,$$
$$g_m(t) = \int e^{-\mathcal{K}_m(t-t')} F_m(t')dt' , \tag{12}$$

where constants $\mathcal{K}_m = N/M^2 \sum_p (1 - e^{impH})$ and Fourier components of the pinning forces

$$F_m = \frac{N}{M} e^{imu_0(t)} \frac{1}{2\pi} \times \int_0^{2\pi} dx\, e^{-imx} \sum_\alpha F[2\pi\alpha - x - g(x - u_0(t), t)] \tag{13}$$

with $u_0(t)=(N/M)[vt-(E_1/\omega)\cos\omega t]$. The CDW drift current I_{dc} is proportional to $\langle \dot U(x,t)\rangle_T=\langle \dot u_0(t)+F_0(t)\rangle_T$. Using the identity

$$e^{i(E_1/\omega)}\cos\omega t=\sum_l i^l J_l(E_1/\omega)e^{-il\omega t} \tag{14}$$

we then obtain

$$I_{dc}\propto E_0-\left[\frac{N}{M}\right]^2\sum_{m,l}J_l^2\left[\frac{mNE_1}{M\omega}\right]\frac{mNv/M+l\omega}{\mathcal{K}_m^2+(mNv/M+l\omega)^2}\operatorname{Im}\int_0^{2\pi}\int_0^{2\pi}\frac{dx\,dx'}{(2\pi)^2}\sum_{\alpha,\alpha'}F'(2\pi\alpha-x)F(2\pi\alpha'-x')e^{im(x-x')}, \tag{15}$$

where m and l are integers and $J_l(x)$ is the Bessel function of the first kind. Equation (15) is correct to the second order of perturbation of the pinning forces $F(x)$. The result is similar to the one obtained[12] using the Fukuyama-Lee-Rice (FLR) model[14] of random pinning. The second term in Eq. (15) is always negative. This term is minimized, and therefore I_{dc} is maximized, when $mvN/M+l\omega=0$. The differential resistance is also maximized at this point.

Equation (9) can be solved in a similar way. Let $u(x,t)=e^{iqx}h(x,t)$ (Bloch-wave-like solution), Eq. (9) becomes

$$\dot h(x,t)+\left[-i\omega_0+\frac{N}{M}\sum_\alpha F'(X_\alpha)\right]h(x,t)+\frac{N}{M^2}\sum_p[h(x,t)-e^{iqHp}h(x+Hp,t)]=\sum_\alpha\left[\frac{N}{M}F'(X_\alpha)-i\omega_0\right]e^{i(2\pi\alpha-x)q}. \tag{16}$$

The function $h(x,t)$ is a periodic function of x with a period of 2π. We can write $h(x,t)$ as

$$h(x,t)=\sum_m h_m(t)e^{imx}. \tag{17}$$

Generally speaking, q is the reciprocal vector of the lattice. Because of the incommensurability between the CDW and the underlying lattice, q will not be equal to the vector m of the Fourier-expansion component of $h(x,t)$. One could expand $h(x)$ perturbatively in the orders of $F(x)$ as

$$h^0(x,t),\ h^1(x,t),\ h^2(x,t),\ldots. \tag{18}$$

For large enough t the approximate solution is

$$h^0(x,t)=0, \tag{19}$$

$$h_m^1(t)=\frac{N}{M}\int_0^t dt'e^{-(\mathcal{K}_{m+q}-i\omega_0)(t-t')+iu_0(t')(q+m)}\int_0^{2\pi}dx\sum_\alpha e^{i2\pi\alpha q}F'(2\pi\alpha-x)e^{-ix(q+m)}. \tag{20}$$

Now we can calculate the quantity δf as defined by Eq. (10). The real and the imaginary parts of δf are proportional to Y and δ, respectively. To the second order of perturbation of pinning forces, we obtain the average of δf

$$\langle\delta f\rangle_T=\frac{1}{T}\int_0^T\delta f\,dt \tag{21}$$

to be

$$\begin{aligned}\langle\delta f\rangle_T=-i\omega_0-\frac{N}{MH}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\frac{dx\,dx'}{(2\pi)^2}&\left[F''(x)F(x')\sum_{l,m}e^{im(x'-x)}J_l^2\left[\frac{mNE_1}{M\omega}\right]\frac{\mathcal{K}_m-i(mvN/M+l\omega)}{\mathcal{K}_m^2+(mvN/M+l\omega)^2}\right.\\&+F'(x)F'(x')\sum_{l,m}e^{i(m+q)(x'-x)}J_l^2\left[\frac{(m+q)NE_1}{M\omega}\right]\\&\left.\times\frac{\mathcal{K}_{m+q}-i[(m+q)vN/M+l\omega-\omega_0]}{\mathcal{K}_{m+q}^2+[(m+q)vN/M+l\omega-\omega_0]^2}\right],\end{aligned} \tag{22}$$

where $T=2\pi/\omega$ and $F''(x)$ is the second derivative of $F(x)$. Like the differential resistance, Young's modulus Y and internal friction δ have sharp anomalies when $mvN/M+l\omega=0$. For a fixed ω, the drift velocity v of the CDW changes as one varies the dc bias current I_{dc}. Anomalies should be observed as v approaches

$$-\frac{M\omega l}{NM}, \tag{23}$$

where l,m are integers. These anomalies are similar to the Shapiro-type interferences in the electronic response. The magnitudes of these harmonic and subharmonic anomalies depend on the strength of the pinning forces $F(x)$ and on the amplitude of the ac field. In addition, Y and δ should also have sharp anomalies when v is equal to

$$-\frac{M(\omega l-\omega_0)}{N(m+q)}, \tag{24}$$

where l,m are integers. It seems that linear fluctuations are very important. They shift the positions of some of the anomalies approximately by $M\omega_0/N$; the magnitudes of the anomalies are given by the second term of Eq. (22). The experiments of Bourne, Sherwin, and Zettl did not show the effects of the linear fluctuations on elastic properties of the CDW conductors. We believe this kind of experiment would be very helpful for understanding both the electrical and the elastic properties of the CDW conductors.

A single-domain, classical model of two mutually incommensurate, interacting, many-body systems has been solved. It successfully accounts for the elastic properties of charge-density-wave conductors in the presence of combined ac and dc electric fields. Xiang and Brill[15] found that the electric-field-dependent anomaly in CDW conductors decreases with frequency, i.e., $\Delta Y/Y \propto \omega_0^{-p}$, where $p<1$. If the ac electric field is zero, i.e., $E_1=0$, the real part of Eq. (22) reduces to two parts. One part is independent of ω_0, while the other one, which is a function of ω_0, is equal to

$$\frac{N}{MH}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\frac{dx\,dx'}{(2\pi)^2}F'(x)F'(x')\sum_m e^{i(m+q)(x'-x)} \times\frac{\mathcal{K}_{m+q}}{\mathcal{K}_{m+q}^2+[(m+q)vN/M-\omega_0]^2}\,. \quad (25)$$

We see that the relative shift in the Young's modulus decreases as frequency ω_0 increases. This result, however, is different from the experimental data of Xiang and Brill. One possible reason could be that at low frequency the major contribution to the relative shift comes from the frequency-dependent part when the bias field is not much larger than the depinning threshold. It would be interesting to test experimentally whether $\Delta Y/Y$ approaches a limiting minimum value when the frequency becomes very high and the bias field is large. The shear modulus of CDW conductors has also been measured.[16] Like the Young's modulus, the shear modulus decreases as the depinning velocity v of the CDW increases. This result suggests that the interchain coupling of the lattice is affected by the motion of the CDW. We have not yet been able to construct a Hamiltonian that includes both the lattice interchain coupling of lattice and the lattice-CDW coupling. This is certainly an interesting and important problem, and is worth being explored.

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