Dynamics of Charge-Density Waves Pinned by Impurities

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The model of Fukuyama and Lee for the pinning of the charge-density–wave phase by impurities is investigated. The current-current correlation function, related to the force-force correlation function by the equation of motion, is calculated to fourth order in the impurity concentration n_0. It is found that the threshold electric field is proportional to n_0^2, and above threshold, the resulting current is periodic in time, exhibiting many harmonics.

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Recent experiments on the conductivity of $NbSe_3$ and related compounds in the presence of an applied electric field have given strong support to the notion that the material exhibits a charge-density wave (CDW) pinned by impurities.[1-8] It has been found that there is a threshold electric field E_0 below which the CDW current vanishes. By varying of the impurity concentration n_0, E_0 has been found to vary approximately as n_0^2 for low concentration. Above threshold, the current is observed to have a periodic time dependence, with many harmonics of the fundamental frequency f (i.e., narrow-band noise); f is found to be proportional to the time-averaged current.[1-3]

Grüner, Zawadowski, and Chaikin[8] found that these and many other experimental facts concerning such systems can be qualitatively understood in terms of an overdamped oscillator with a periodic potential. The collective coordinate $x(t)$, which describes the center-of-mass position of the CDW, satisfies an equation of motion

$$\dot{x}/\tau + (\omega_0^2/Q)\sin(Qx) = eE/m, \tag{1}$$

where $m\omega_0^2/Q$ represents the strength of the pinning force.

Our goal is to investigate to what extent Eq. (1) can be derived from microscopic theory. We consider the Fukuyama-Lee model of a CDW in which the charge density is represented by

$$\rho(\vec{r}, t) = \rho_0 + \delta\rho\cos[\vec{Q}\cdot\vec{r} + \varphi(\vec{r}, t)], \tag{2}$$

where the wave vector $\vec{Q}$ is parallel to the x axis and incommensurate with the crystal lattice.[9] The amplitude $\delta\rho$ is taken to be a constant in $\vec{r}$ and t. The CDW is assumed to interact weakly with a random distribution of impurity or pinning sites, and is acted upon by a uniform electric field $E(t)$. The Hamiltonian is taken to be

$$H = C\int d^3r\{|\nabla\varphi(\vec{r}, t)|^2/2 + e^*E(t)\varphi(\vec{r}, t) + V_0\delta n(\vec{r})\cos[\vec{Q}\cdot\vec{r} + \varphi(\vec{r}, t)]\}, \tag{3}$$

where the coordinates x, y, and z have been scaled so that the coefficients of the x, y, and z derivative terms are equal; time is chosen such that the velocity C_0 of small-amplitude phase fluctuations is unity. The quantity $\delta n(\vec{r})$ is the fluctuation of the local density of pinning sites defined so that its spatial average vanishes, $\int\delta n(\vec{r})d^3r = 0$.

In these units, V_0 has the dimension of length. Within linear response theory, the deviation of the phase $\delta\varphi(\vec{r})$ produced by a defect centered at a point where $\sin\vec{Q}\cdot\vec{r} = 1$ is

$$\delta\varphi(\vec{r}) = V_0/4\pi r. \tag{4}$$

For weak pinning, we require $\delta\varphi(\vec{r}) \ll 1$. Therefore, V_0 is taken to be small compared to 4π times the "size" a of the impurity so that Eq. (4) holds only for $r > a$ with $\delta\varphi(\vec{r})$ saturating for $r < a$.

We assume relaxational dynamics for φ so that the equation of motion becomes

$$\dot{\varphi}(\vec{r}, t)/\tau = \nabla^2\varphi(\vec{r}, t) + V_0\delta n(\vec{r})\sin[\vec{Q}\cdot\vec{r} + \varphi(\vec{r}, t)] + e^*E(t), \tag{5}$$

where the damping rate $1/\tau$ is due to the CDW coupling to phonons, normal electrons, etc., but not to impurities. We define a collective variable $\Delta(t)$ as the spatially averaged phase:

$$\Delta(t) = \int d^3r\, \varphi(\vec{r},t)/\int d^3r, \tag{6a}$$

and the phase fluctuation $\delta\varphi$ as

$$\delta\varphi(\vec{r},t) = \varphi(\vec{r},t) - \Delta(t). \tag{6b}$$

We then define a coherence length ξ to be the size of the region over which the mean square fluctuation of $\delta\varphi(\vec{r})$ is $(2\pi)^2\gamma$:

$$\langle \delta\varphi^2\rangle_{i,s} \equiv \int^{\xi} d^3r\, \delta\varphi(\vec{r})^2/\int^{\xi} d^3r = (2\pi)^2\gamma, \tag{7}$$

where $\langle\cdots\rangle_i$ denotes an impurity average and $\langle\cdots\rangle_s$ denotes a spatial average, and where γ is of order unity. For the static case, one has

$$-\nabla^2\delta\varphi(\vec{r}) = V_0\,\delta n(\vec{r})\sin[\vec{Q}\cdot\vec{r} + \Delta + \delta\varphi(\vec{r})]. \tag{8}$$

Expanding $\delta\varphi$ in powers of V_0, one has to order V_0,

$$-\nabla^2\delta\varphi_1(\vec{r}) = V_0\delta n(\vec{r})\sin(\vec{Q}\cdot\vec{r} + \Delta) \tag{9a}$$

or

$$\delta\varphi_1(\vec{r}) = \int d^3r'\, G(\vec{r},\vec{r}\,')V_0\delta n(\vec{r}\,')\sin(\vec{Q}\cdot\vec{r}\,' + \Delta), \tag{9b}$$

where $G(\vec{r},\vec{r}\,') = |\vec{r} - \vec{r}\,'|^{-1}$ is the effective Coulomb potential. More generally, from Eq. (8) we see that $\delta\varphi$ behaves as an electrostatic potential, with the fluctuating quantity $V_0\delta n(\vec{r})\sin[\vec{Q}\cdot\vec{r} + \Delta + \delta\varphi(\vec{r})]$ acting as a charge density. An analogous calculation has been carried out by Efetov and Larkin,[10] and one finds to all orders in V_0,

$$\langle\delta\varphi^2\rangle_{i,s} = n_0V_0^2\int^{\xi} d^3r'/2(\vec{r} - \vec{r}\,')^2 = 2\pi n_0V_0^2\xi = (2\pi)^2\gamma \tag{10}$$

or

$$\xi = 2\pi\gamma/n_0V_0^2. \tag{11}$$

Equation (10) is represented by the diagram in Fig. 1. Equation (11) is consistent with the domain size found by Lee and Rice.[11] Since V_0 for weak pinning is small compared to the interatomic spacing, ξ is large compared to the mean spacing between impurity sites. For $n_0 = (10^{-3}–10^{-5})/a^3$, pinning strength $cV_0 \cong 0.01$ eV, and $E_F \cong 1$ eV, we have $\xi \cong 1 - 100$ μm. This value of V_0 used will be inferred below.

The length ξ is important for two reasons. Firstly, since we are expanding in terms of the dimensional parameter $n_0V_0^2 = 2\pi\gamma/\xi$, spatial integrals in diagrams must be cut off at distances

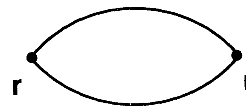

FIG. 1. Bubble diagram representing $\langle\delta\varphi^2\rangle_{i,s}$. The solid lines are Green's functions.

$|\vec{r} - \vec{r}\,'| \cong \xi$ so that trigonometric functions are expanded only for arguments $< 2\pi$. Secondly, one expects that regions having $\langle\delta\varphi^2\rangle_{i,s}$ which differ by an amount $\gg (2\pi)^2$ act as separate domains which are coupled by residual interactions, such as contact and long-range Coulomb forces.[12] While experimental results in different laboratories may lead to different interpretations regarding the coupling of domains, generally the narrow-band noise is consistent with the corresponding dynamics of a single domain.[1-4,8]

The observed noise spectrum of the CDW is proportional to the current-current correlation function

$$C_\alpha(t,t') = \dot{\Delta}_\alpha(t)\dot{\Delta}_\alpha(t') \tag{12}$$

for a fixed impurity distribution α. While this measured quantity depends upon the initial condition $\Delta_\alpha(t')$, we are free to choose the same value of $\Delta_\alpha(t') = \Delta(t')$ for all α. If one integrates Eq. (5) over a volume of size ξ, one finds

$$\dot{\Delta}_\alpha(t)/\tau = e^*E(t) + F_\alpha(t), \tag{13}$$

where

$$F_\alpha(t) = \langle V_0\,\delta n_\alpha(\vec{r})\sin[\vec{Q}\cdot\vec{r} + \Delta_\alpha + \delta\varphi_\alpha(r)]\rangle_s. \tag{14}$$

Since $F_\alpha(t)$ fluctuates randomly over the ensemble of impurity distributions, it follows that $\langle F(t)\rangle_i = 0$. Therefore, the impurity-averaged correlation function satisfies

$$C(t,t')/\tau^2 \equiv \langle\dot{\Delta}(t)\dot{\Delta}(t')\rangle_i/\tau^2 = (e^*E)^2 + \langle F(t)F(t')\rangle_i, \tag{15}$$

where the $\nabla^2\delta\varphi$ terms lead to negligible boundary terms. While $\langle F(t)\rangle_i = 0$, $\langle F(t)F(t')\rangle_i \neq 0$, and gives the desired pinning effects as well as internal damping effects. Since Δ gives the drift velocity of the CDW, we see that Eq. (15) expresses the current-current correlation function in terms of the external force and the pinning force-force correlation function.

Consider the adiabatic approximation in which $\Delta(t)$ and $\delta\varphi(\vec{r},t)$ vary sufficiently slowly in time that the zero-order Green's function can be approximated by its static limit $|\vec{r} - \vec{r}\,'|^{-1}$ in calculating diagrams. This requires $\omega/\tau \ll (2\pi C_0/$

$\xi)^2$. For $\tau \cong 10^{-11}$ sec and Fermi velocity $v_F \cong 10^8$ cm/sec, this approximation is valid for CDW drift velocities $v_d = \omega/Q < 10^2$ cm/sec, which is well obeyed near threshold,[8] but not at high velocities as considered by Sneddon, Cross, and Fisher.[13] The pinning force for a given Δ is given by Eq. (13) where $\delta\varphi(\vec{r})$ is given by Eqs. (5) and (6b). To order $(n_0 V_0^2)^4$, one finds

$$C(t, t')/\tau^2 = (e^*E)^2 + (n_0 V_0^2/2V)\cos\theta + (n_0 V_0^2/2)^2 V^{-1}(\cos 2\theta - \cos\theta)\, b + \ldots, \tag{16}$$

where $V = 4\pi\xi^3/3$, $\theta = \Delta(t) - \Delta(t')$, and $b = 4\pi\xi$ is the bubble shown in Fig. 1. Terms of order $(n_0 V_0^2)^n/V$ where $n = 3, 4$ have coefficients proportional to the diagrams shown in Fig. 2, which are proportional to ξ^{n-1}. We note that for a finite domain of size ξ given by Eq. (11), all of the terms in the expansion of $\langle F(t)F(t')\rangle_i$ through fourth order in $n_0 V_0^2$ are proportional to $(n_0 V_0^2)^4$. Presumably, a calculation to infinite order in V_0 would yield an infinite number of terms of the same overall order in $n_0 V_0^2$. Details of the above calculation will be presented elsewhere.

From Eqs. (15) and (16), the threshold field is given by

$$e^*E_0 = K n_0^2 V_0^4, \tag{17}$$

where K is a positive, real constant of order unity, obtained by minimizing Eq. (17) with respect to θ. Equation (17) was obtained by Lee and Rice,[11] employing a different procedure. Using the parameters given above and $e^* = \pi^2 e/2\hbar v_F$, $eE_0 \cong 10^{-2}$ eV/cm for $n_0 \cong 10^{-5}/a^3$, and taking $K = 1$, we infer $V_0 \cong a$, consistent with our weak-pinning assumption.

We can find the time dependence of Δ by integrating Eq. (16), treating t' as a constant. Rewriting Eq. (16), we have through fourth order in $n_0 V_0^2$

$$\langle \dot\Delta(t)\dot\Delta(t')\rangle_i/\tau^2 = (e^*E)^2 + \textstyle\sum_n A_n \cos(n\theta), \tag{18}$$

where the A_n are positive constants of order $(n_0 V_0^2)^4$. For $E < E_0$, Δ is a constant. For $E > E_0$, we may approximate $\dot\Delta(t')$ by its impurity-averaged root mean square value obtained by setting $t = t'$:

$$\langle \dot\Delta(t')^2\rangle_i/\tau^2 = e^{*2}(E^2 + E_1^2), \tag{19}$$

where we have defined the "maximum" field E_1 to be

$$(e^*E_1)^2 \equiv \textstyle\sum_n A_n. \tag{20}$$

We now have a first-order equation of motion for $\Delta(t)$, with the initial condition given by Eq. (19). If we naively keep only the first term in the Fourier expansion in terms of $\Delta(t)$ on the right-hand side of Eq. (18), we may solve for $\dot\Delta(t)$:

$$j(t) = \dot\Delta(t) \cong f[(E/E_0)^4 - 1]^{1/2}\{(E/E_0)^2 + \cos[f(t-t')]\}^{-1}, \tag{21}$$

where the fundamental "noise" frequency f is given by

$$f = \tau e^*[(E^4 - E_0^4)/(E^2 + E_1^2)]^{1/2}. \tag{22}$$

The static CDW current $j_{dc}(E)$ is the time average of $j(t)$, which in this naive approximation is found to be equal to f as given by Eq. (22). The amplitude of the nth harmonic is found by multiplying Eq. (21) by $\cos\{nf(t-t')\}$, and integrating over a full period of the harmonic time dependence.

We note that in general there are two electric field parameters in this formula. It is easy to show for the fourth-order calculation that we have performed that $E_1 > E_0$ for small γ. As the number of Fourier components of the right-hand side of Eq. (18) increases, the ratio E_1/E_0 also increases. We note that for E_1/E_0 near unity, the behavior of $j_{dc}(E)$ just above threshold is as $(E - E_0)^{1/2}$, contrary to experiment. However, as E_1/E_0 becomes large, the region of square-

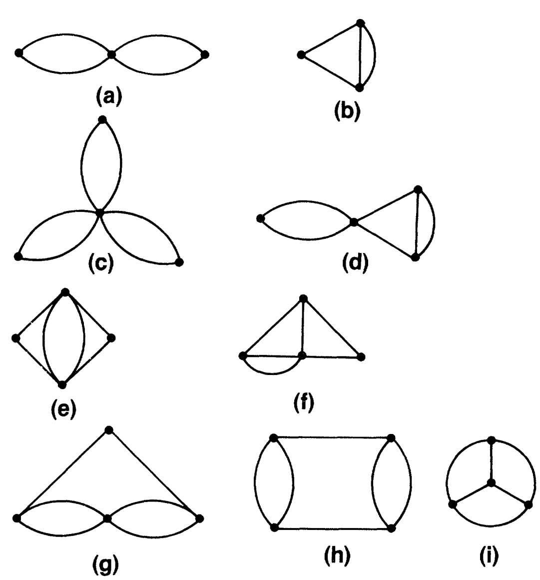

FIG. 2. Diagrams of third and fourth order in $n_0 V_0^2$ that contribute to the force-force correlation function.

root behavior near threshold becomes small. It therefore seems likely that a numerical calculation based upon Eq. (18) will give results that are in general agreement with experiment, if we treat γ as an arbitrary parameter.

Finally, there remains the more fundamental question of treating interdomain couplings, which in this calculation we have neglected. It is likely that dynamical (nonadiabatic) effects are essential in treating the domain-domain coupling. Presumably, such a treatment would lead to a correlated motion of the CDW in different domains, without destroying the narrow-band noise.

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