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Dynamical friction in sliding condensed-matter systems

J. E. Sacco, J. B. Sokoloff, and A. Widom
Department of Physics, Northeastern University, Boston, Massachusetts 02115

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A many-body system sliding at constant center-of-mass velocity relative to an external periodic potential (e.g., due to the channels in which the ions move in an ionic conductor) is studied as a possible model for dynamical friction and as a model for electrical conduction in superionic conductors and systems containing charge-density waves. The damping force is found to all orders in the center-of-mass velocity by calculating the lowest-order response to the external periodic potential (valid if this potential is weak) via the conventional fluctuation-dissipation theorem. This permits us to calculate nonlinear electrical conduction in the charge-density wave and superionic-conductor applications of the model.

I. INTRODUCTION

A large class of condensed-matter systems, such as adsorbed films,[1] charge-density-wave conductors,[2] and superionic conductors,[3] can be modeled as translationally invariant, interacting many-body systems subject to external, spatially periodic potentials. Some of the properties of such systems are determined by the geometric property of commensurability. If the internal length scales of the system, in the absence of the external potential, "fit well" into the minima of the external potentials, then the system becomes commensurate and can be characterized as follows: (i) The system is incompressible, i.e., it is difficult to change the internal length scales gradually since such compressional thermodynamic work would destroy the minimum-free-energy feature of being commensurate. (ii) The reversible thermodynamic work required to displace the system relative to the external potential is large. In what follows it will be natural to consider this latter property as exhibiting a large "static friction." If the system is incommensurate with the external potential, then the situation is greatly changed. It may require very little reversible work to displace the system relative to the external potential. The energy used to move one atom off its minimum-external-potential site is recovered somewhere else in the system by another atom falling into its minimum-external-potential site. Hence incommensurate systems exhibit small static friction.[4,5]

In previous work, the above notions[4,5] were examined in detail for the one-dimensional classical Hamiltonian

$$H=\sum_n (p_n^2/2M)+\sum_n v(|x_n-x_{n-1}|)+\sum_n \varphi(x_n)\,, \tag{1}$$

where $v(r)$ is a nearest-neighbor two-body potential, and $\varphi(x)$ is the external periodic potential. Here, the dynamical properties of this Hamiltonian will be studied. In particular, our interest is in the "dynamical friction" exhibited as the system is moved with uniform velocity relative to the external potential. Some of the results obtained for Eq. (1) are easily extended to higher dimensions. These cases will, in due course, be noted.

The physical considerations required to understand dynamical friction are the following: (i) When the system moves with uniform velocity relative to the external spatially periodic potential, the time variations of the potential as "seen" by the system density also become periodic. (ii) When these time scales become comparable to the periods of the dynamical excitations in the system, there is a resonant absorption of energy by the system. This large, irreversible heating yields a large dynamical-frictional-drag coefficient. (iii) If the time scales are not in resonance with the excitations of the system, then the dynamical-frictional drag is small. (iv) As the velocity of the system increases, the external frequency scales become much higher than those of the internal system excitations. Ultimately, the frictional drag monotonically decreases to zero with increasing velocity.

Our approach is similar to that used by Boriak and Overhauser[6] and by Lee and Rice[7] in their respective discussions of frictional drag of electrons and sliding charge-density waves due to impurities and of scattering of charge-density waves by free electrons. Our analysis considers damping due to phonon or phason excitations as well as electrons. In addition, we consider the general dependence of the frictional drag on velocity of the sliding system, which allows us to consider nonlinear conduction in the charge-density wave and superionic-conductor applications of our work. Also, we note different results for the damping force in one, two, and three dimensions.

In Sec. II, we consider the dynamics of a chain of atoms in the absence of an external potential. In Sec. III, we consider the force of friction of a sliding lattice in a periodic external potential. In Sec. IV, the drag coefficient is studied for the one-dimensional model. In Sec. V, the effects of dilute impurities on the force of friction are studied. In Sec. VI, quantum-mechanical and multiphonon effects are studied. In Sec. VII friction due to electronic excitation is discussed.

In the concluding section we will discuss the applicability of the model studied in this article to thin films, supersonic conductors, and conductors with charge-density waves which contribute to the conductivity. We will also consider in what sense this is a model for ideal dynamical friction between real crystalline surfaces.

II. DYNAMIC CORRELATIONS FOR $\varphi(x)=0$

We will first discuss the one-dimensional model that we considered in previous publications.[4,5] The exact results for zero external potential will be used in sections to follow.

The thermodynamic properties of chains described by Eq. (1) are determined by

$$d\mu=-s\,dT+l\,df\,, \tag{2}$$

where l is the mean spacing between particles, and f is the compression force applied at the ends of the chain (i.e., one-dimensional "pressure"). The following features of static correlations in $\varphi(x)=0$ chains were discussed in Ref. 5 for potentials $v(r)$, which are strongly repulsive at short distances and (convex downward) decreasing to zero as $r\to\infty$:

(i) The thermodynamic properties are determined by

$$\mu_0(f,T)=-k_BT\ln\int_0^\infty\left(\frac{dr}{\lambda}\right)e^{-[v(r)+fr]/k_BT}\,,\qquad \lambda=\left(\frac{2\pi\hbar^2}{Mk_BT}\right)^{1/2}. \tag{3}$$

(ii) The static structure factor

$$S(Q)=\lim_{N\to\infty}\frac{1}{N}\left\langle\sum_{n,m}^{N}e^{iQ(x_n-x_m)/k_BT}\right\rangle, \tag{4}$$

can be computed via analytic continuation of the chemical potential,

$$\mu_0(f-ik_BT,Q)=\mu_0(f,T)-k_BT\ln M(Q)\,, \tag{5a}$$

$$S(Q)=\mathrm{Re}\left(\frac{1+M(Q)}{1-M(Q)}\right). \tag{5b}$$

(iii) In the low-temperature limit, the structure factor is identical to that of a "harmonic solid" with spring constant $K_0=v''(l_0)$, i.e.,

$$K_0^{-1}=-\lim_{T\to0+}(\partial l_0/\partial f)_T=-\lim_{T\to0+}(\partial^2\mu_0/\partial f^2)_T\,, \tag{6a}$$

$$S(Q)=\left(\frac{\sinh(k_BTQ^2/2K_0)}{\cosh(k_BTQ^2/2K_0)-\cos(Ql_0)}\right)\quad(T\to0+)\,. \tag{6b}$$

(iv) The coherence length $\xi(T)$ [i.e., the "reciprocal width" of the first "quasi-Bragg peak" in $S(Q)$ shown in Fig. 1] diverges as $T\to0$ (the "melting" temperature) according to

$$\xi(T)\sim l_0(K_0l_0^2/k_BT)^{\nu}\quad(T\to0+)\,, \tag{7}$$

with a critical index $\nu=1$.

Here, we shall extend the above results to the dynamical structure factor

$$S(Q,\omega)=\int_{-\infty}^{+\infty}\frac{dt}{(2\pi)}e^{i\omega t}\lim_{N\to\infty}\frac{1}{N}\left\langle\sum_{n,m}^{N}e^{iQ[x_n(t)-x_m(0)]}\right\rangle. \tag{8}$$

From the fact (discussed above) that the static correlations in the chain as $T\to0+$ are identical to those of a simple harmonic solid, one might guess that the same holds true for dynamic correlations. This can be proved for the phonon spectrum by using the method of moments.

The Hamiltonian in Eq. (1) for $\varphi(x)=0$, together with Eqs. (4) and (8), implies the following classical (rigorous) sum rules:

$$\int_{-\infty}^{+\infty}S(Q,\omega)\,d\omega=S(Q)\,, \tag{9a}$$

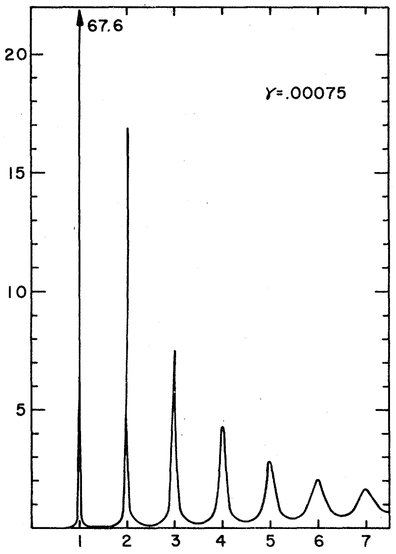

FIG 1. Static structure factor $S(Q)$ (dimensionless). The quasi-Bragg peaks occur at $Q=2\pi n/l_0$. The index n is shown as the abscissa. The quantity γ is $k_BT/2Kl_0^2$.

$$\int_{-\infty}^{+\infty} \omega^2 S(Q,\omega)d\omega = (k_B T Q^2/M), \tag{9b}$$

$$\int_{-\infty}^{+\infty} \omega^4 S(Q,\omega)d\omega = 3\left(\frac{k_B T Q^2}{M}\right)^2 + 2\left(\frac{k_B T Q^2}{M^2}\right) \times \int_0^{\infty} v''(r)[1-\cos(Qr)]dW(r), \tag{9c}$$

where $W(r)$ is the probability that the distance between two neighboring particles is less than r, i.e.,

$$dW(r) = (dr/\bar{\lambda})e^{[\mu_0 - v(r) - fr]/k_B T}. \tag{10}$$

The low-temperature limits,

$$\lim_{T\to 0+}[S(Q)/k_B T] = (Q^2/2K_0)[1-\cos(Ql_0)]^{-1} \tag{11a}$$

and

$$\lim_{T\to 0+}\int_0^{\infty} v''(r)[1-\cos(Qr)]dW(r) = K_0[1-\cos(Ql_0)], \tag{11b}$$

follow, respectively, from Eqs. (6b) and (10) using the method of steepest descent on the integral. Equations (9) and (11) are consistent, if and only if (as $T\to 0+$) the spectral function $S(Q,\omega)$ saturates the sum rules at a single frequency ω_Q, where

$$\omega_Q^2 = (2K_0/M)[1-\cos(Ql_0)]. \tag{12}$$

This can clearly be recognized as the phonon frequency in the near-neighbor harmonic approximation. More precisely, we have for Eq. (1) with $\varphi(x)=0$ the low-temperature result $(Q\neq 0)$

$$\lim_{T\to 0+}[S(Q,\omega)/S(Q)] = \tfrac{1}{2}[\delta(\omega-\omega_Q)+\delta(\omega+\omega_Q)]. \tag{13}$$

The asymptotic form of $S(Q,\omega)$ for small but finite T can be computed in more detail by evaluating Eq. (8) with the particle velocities $\{v_n(t)\}$ treated as Gaussian, random stochastic variables. This is valid in the harmonic approximation,[8] i.e.,

$$S(Q,\omega) = \sum_{m=-\infty}^{\infty} e^{imQl_0}\int_{-\infty}^{+\infty}\left(\frac{dt}{2\pi}\right)e^{-i\omega t}\, e^{-Q^2 W(|m|,t)} \quad (T\to 0+), \tag{14}$$

where

$$W(|n-m|,t) = \int_0^t (t-t')\langle v_n(t')v_m(0)\rangle dt' + (k_B T/2K_0)|n-m|. \tag{15}$$

By expanding the velocity coordinates in normal classical phonon modes, it is easily shown that

$$W(|n|,t) = (k_B T/2K_0)|n| + (k_B T/M)\int_0^t (t-t')J_{2n}(\Omega t')dt', \tag{16}$$

where $\Omega = 2(K_0/M)^{1/2}$ is the maximum phonon frequency (i.e., Brillouin-zone boundary) and $J_n(x)$ is the nth-order Bessel function. This expression will be used in later sections.

III. FRICTIONAL DRAG

We will now write down general expressions for the frictional drag to lowest order in the external potential. The considerations in this section hold for arbitrary dimension d. It is well known that for a system with a translationally invariant Hamiltonian, the static fluctuation response theorem for the compressibility

$$\langle \Delta N^2\rangle = Nk_B T\chi_T, \tag{17a}$$

where

$$\chi_T = (\partial\rho/\partial P)_T = \rho^{-1}(\partial\rho/\partial\mu)_T, \tag{17b}$$

has a dynamic-fluctuation-response extension for the classical structure[9] factor

$$S(\vec{Q},\omega) = (k_B T/\pi\omega)\,\mathrm{Im}\chi(\vec{Q},\omega+i0^+), \tag{18a}$$

where

$$\chi(\vec{Q},\zeta) = \rho^{-1}(\delta\rho/\delta\mu)_{\vec{Q},\zeta}. \tag{18b}$$

Now suppose that the system is moving with velocity $(-\vec{v})$ relative to an external spatially periodic potential. From the system coordinates' Galilean frame, the external potential is represented as periodic in both space and time,

$$\Phi = \sum_n^N \varphi(\vec{r}_n - \vec{v}t) = \sum_{\vec{G}} \varphi_{\vec{G}}\left(\sum_n^N e^{i\vec{G}\cdot\vec{r}_n}\right)e^{-i\vec{G}\cdot\vec{v}t}, \tag{19}$$

where the $\{\vec{G}\}$ are the reciprocal lattice vectors of $\varphi(\vec{r})$.

To lowest order in φ, the heating rate due to this drift velocity $\vec{v}$ is determined by the dissipative part of the response function $\chi(\vec{Q},\zeta)$,[9]

$$\langle\dot{H}\rangle = \tfrac{1}{2}N\sum_{\vec{G}}|\varphi_{\vec{G}}|^2[\omega\,\mathrm{Im}\chi(\vec{Q},\omega+i0^+)]_{\vec{Q}=\vec{G},\ \omega=\vec{G}\cdot\vec{v}}. \tag{20}$$

It is convenient to describe this heating rate by a "drag" force $\vec{F}$ per particle

$$\langle\dot{H}\rangle = N\vec{F}\cdot\vec{v}, \tag{21}$$

and, further, to define a velocity-dependent friction coefficient $\vec{\Gamma}(\vec{v})$ by

$$\vec{F} = \vec{\Gamma}\cdot\vec{v}. \tag{22}$$

Equations (18a) and (20)–(22) imply that, to lowest order in φ, the frictional-drag coefficient as a function of velocity is

$$\vec{\Gamma}(\vec{v}) = \left(\frac{\pi}{2k_B T}\right)\sum_{\vec{G}}\vec{G}\vec{G}\,|\varphi_{\vec{G}}|^2 S(\vec{Q}=\vec{G},\ \omega=\vec{G}\cdot\vec{v}), \tag{23}$$

which can be computed in any model where the structure factor $S(\vec{Q}, \omega)$ is known. To treat a more realistic situation in which excitations from both a sliding system and "substrate lattice" contribute to the damping, we simply add to Eq. (23) a similar contribution due to the creation of excitations in the substrate lattice. This is valid in the context of perturbation theory.

Since we are considering the linear response to the external periodic potential $\varphi(\vec{r})$, which represents a stationary crystal lattice, rather than the response to an external electric field, we can treat linear and nonlinear electrical conduction on an equal footing in the charge-density wave and superionic-conductor applications of our theory. Our method is valid as long as the external periodic potential is weak, which is a reasonable assumption in many applications of the model (for example, see Ref. 10).

IV. FRICTION IN ONE DIMENSION

Static equilibrium considerations of a one-dimensional chain in the presence of a week, periodic, substrate potential have shown that the ground state ($T = 0$ K) of an incommensurate chain becomes continuously degenerate below some critical strength of the periodic potential.[5] The existence of this degenerate ground state allows for the possibility of a zero-frequency phonon mode since the chain can be translated quasistatically through a continuous sequence of equilibrium configurations at no cost in energy. Static considerations alone cannot provide for the actual existence but only for the possible existence of such a mode. What is needed is an investigation of the dynamical resistance to sliding.

The results of the previous sections provide an understanding of the dynamics of the zero-frequency mode at low temperatures. From Eqs. (13) and (23), the frictional-drag coefficient ($v \neq 0$) in the limit of zero temperature is

$$\Gamma(v) = \frac{\pi}{2mv^2} \sum_G G^2 |\varphi_G|^2 [\delta(Gv - \omega_G) + \delta(Gv + \omega_G)] . \tag{24}$$

Since it is virtually impossible to satisfy the above δ functions for most values of v, one sees that, to second order in the substrate potential, the dynamical friction $[F = v\Gamma(v)]$ essentially vanishes as the temperature approaches zero regardless of the degree of commensurability between the substrate and the lattice. The above selection rules would also occur in a quantum-mechanical treatment of one-phonon processes. It is simply a consequence of the fact that a moving periodic potential produces a harmonic potential of frequency Gv and wave vector G. For a classical system (unlike a quantum system) all multiphonon processes vanish as $T \to 0$. In particular, the two-phonon processes in $\Gamma(v)$ (in the classical system) are of order kT/Ka to some power, where K is the chain's force constant, and a its lattice constant. These processes produce low, broad "tails" for the one-phonon peaks which result in a weak frictional force at low temperatures. The one-phonon peaks themselves do not broaden with temperature, due to the absence of anharmonic effects in the model.

We have performed a molecular-dynamics computer calculation on such a chain of atoms, which is consistent with the above picture. The external potential $\varphi(x)$ was taken to be

$$\varphi(x) = \frac{W}{2}\left(1 - \cos\frac{2\pi}{a}x\right),$$

and the atoms interacted via a near-neighbor harmonic potential of force constant K. Thermal equilibrium was attained by including in the equation of motion (i.e., Newton's second law) for the chain a random force and a damping term.[11] The resulting Langevin equation was then integrated numerically until thermal equilibrium was attained, at which point the damping and random forces were turned off (i.e., the system was disconnected from the "thermal bath"). After this procedure was performed, the chain was generally left with a center-of-mass velocity. We studied the way in which the center-of-mass velocity of the sliding chain in the absence of an external force decayed with increasing time. The chain, whose center-of-mass velocity is shown as a function of time in Fig. 2, has Ka^2/W equal to 100, and the time is in units of $(ma^2/\pi W)^{1/2}$. The velocity shown is the average velocity over the previous time interval of 10 time units. The chain on which the calculation was performed has a period 100/77 times that of the sinusoidal potential. It is a chain which has a nearly degenerate ground state, and hence, slides freely at $T = 0$.[4] As is seen, there is virtually no damping of the center-of-mass velocity in the 200 unit-time interval that is shown, which for $m = 100$ a.u., $W = 0.4$ eV, and $a = 3 \times 10^{-8}$ cm corresponds to a time of 10^{-10} sec at a temperature of about 0.6 times the sinusoidal potential strength W. This is presumably because the damping due to two-phonon processes is at least of order $k_B T/Ka^2$, and hence is quite small since $k_B T/Ka^2$ is only $\frac{1}{100}$. The wiggles in the velocity versus time curve represent thermal fluctuations. Similar calculations were performed for $k_B T/W = 0.15$, 3.0, and 15.0. The $k_B T/W = 0.15$ run shows that the undamped free sliding which occurs at $T = 0$ seems to persist as the temperature is increased.

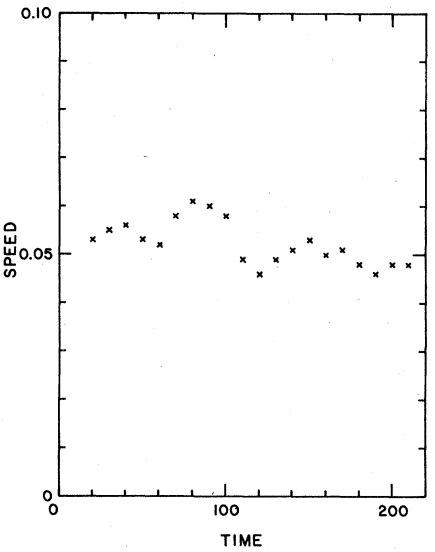

FIG. 2. Results of molecular-dynamics calculations. The center-of-mass speed for a one-dimensional chain (in units of substrate lattice constant per time unit) verus time in units of $(ma^2/\pi W)^{1/2}$.

The $k_B T/W = 3.0$ and 15.0 runs still show that the damping of the center-of-mass velocity is quite small although, of course, the thermal fluctuations increase with increasing temperature. The picture that arises is that at $T=0$ for a chain of atoms incommensurate with the sinusoidal potential with W/Ka^2 very small, there is neither static nor dynamic friction. As T increases, there still appears to be no static[5] friction and very little dynamic friction until $k_B T$ is comparable to Ka^2; the damping is determined by $k_B T/Ka^2$ and not $k_B T/W$. For chain and sinusoidal potential commensurate, there is strong static friction but little dynamic friction for $T=0$. At $T\neq 0$ there is no static friction (because the chain melts when $T\neq 0$, as pointed out in Ref. 5) and little dynamic friction until $k_B T$ is comparable to Ka^2.

V. SYSTEMS WITH DILUTE IMPURITIES

We saw in the previous section that a pure classical chain of atoms has practically no damping at low temperatures. In this section we consider the frictional effects of a dilute number of substitutional impurities in an otherwise pure system. The derivation of the drag function for the impure case is similar to that of the pure case with the external potential replaced by

$$\Phi_{\text{imp}} = \sum_{\vec{n}} \lambda_{\vec{n}} \varphi(\vec{x}_{\vec{n}}) \quad (\lambda = 0, 1)\,. \tag{25}$$

One finds the drag coefficient to be

$$\overleftrightarrow{\Gamma}_{\text{imp}}(\vec{v}) = \frac{\pi}{2k_B T} \sum_{\vec{G}} \vec{G}\vec{G}\,|\varphi_{\vec{G}}|^2 S_{\text{imp}}(\vec{Q}=\vec{G},\ \omega=\vec{G}\cdot\vec{v})\,, \tag{26}$$

where $S_{\text{imp}}(\vec{Q},\omega)$ is the Fourier transform of

$$G_{\text{imp}}(\vec{Q},t) = \frac{1}{N} \sum_{\vec{n},\vec{n}'} \lambda_{\vec{n}} \lambda_{\vec{n}'} \langle e^{i\vec{Q}\cdot[\vec{x}_{\vec{n}}(t) - \vec{x}_{\vec{n}'}(0)]} \rangle \tag{27}$$

(the above thermal average being taken in the absence of impurities). The impurities are taken to be randomly distributed so that G_{imp} may be averaged over impurity configurations. Since

$$\overline{\lambda_{\vec{n}} \lambda_{\vec{n}'}} = c(1-c)\delta_{\vec{n},\vec{n}'} + c^2\,, \tag{28}$$

where c is the impurity concentration, it follows when G_{imp} is so averaged that

$$S_{\text{imp}}(\vec{Q},\omega) = c(1-c) S_{\text{self}}(\vec{Q},\omega) + c^2 S_{\text{pure}}(\vec{Q},\omega)\,, \tag{29}$$

where

$$S_{\text{self}}(\vec{Q},\omega) = \frac{1}{N} \sum_{\vec{n}} \langle e^{i\vec{Q}\cdot[\vec{x}_{\vec{n}}(t) - \vec{x}_{\vec{n}}(0)]} \rangle\,. \tag{30}$$

There are two concentration regimes of interest: (i) $c=1$, which was studied in the previous section, and (ii) $c \ll 1$, where S_{imp} is dominated by the first term in Eq. (29). It is convenient to introduce

$$\begin{aligned} \overleftrightarrow{\gamma}(\vec{v}) &= \lim_{c\to 0} [\overleftrightarrow{\Gamma}(\vec{v},c)/c] \\ &= \frac{\pi}{2k_B T} \sum_{\vec{G}} \vec{G}\vec{G}\,|\varphi_{\vec{G}}|^2 S_{\text{self}}(\vec{G},\vec{G}\cdot\vec{v})\,. \end{aligned} \tag{31}$$

Treating the particle velocities as Gaussian random variables (which is appropriate at low temperatures) the auto-pair-correlation function is given by

$$G_{\text{self}}(\vec{Q},t) = \exp\left[-\vec{Q}\vec{Q} : \int_0^t ds\,(t-s)\langle \vec{v}_0(t)\vec{v}_0(0)\rangle\right], \tag{32}$$

where by translational invariance the velocity autocorrelation function is independent of $\vec{n}$. It is the behavior of this function that controls the dynamics of the system in the presence of dilute impurities. It will be seen that the time dependence of this correlation function is strongly dependent upon the dimensionality of the system through the low-frequency behavior of the phonon density of states.

We first consider a one-dimensional system. For small t, we take $n=0$ in Eq. (16) and expand $J_0(\Omega t')$ to zeroth order in t' to give

$$G_{\text{self}}(Q,t) \approx \exp - (Q^2 k_B T/2m)t^2 \quad (|t| \ll 1)\,, \tag{33}$$

which indicates that the particles behave like an ideal gas; whereas, for large t, the behavior of G is diffusive

$$G_{\mathrm{self}}(Q,t)\approx e^{-Q^2D|t|}\,,\quad |t|\gg 1 \tag{34}$$

where

$$D=k_BT/M\Omega\,. \tag{35}$$

To obtain this result, we have made use of some standard definite integrals involving Bessel functions. This "liquid-like" behavior is typical of a one-dimensional system that "melts" at $T=0+$. Since the short-time behavior of G_{self} controls the high-frequency behavior of $S_{\mathrm{self}}(Q,\omega)$, whereas the long-time behavior of G_{self} controls the small-frequency behavior of S_{self}, we find by Fourier transforming Eqs. (33) and (34) that S_{self} is Lorentzian for small ω and Gaussian for large ω. Which of the two line shapes dominates for the operable range of ω is clearly decided by the temperature. At $T\to 0$, classical systems cannot diffuse. Therefore, the line shape is "motionally narrowed" into a Lorentzian. By taking the Fourier transform of Eq. (34), it follows that for low temperatures

$$\gamma(v)\approx\frac{1}{2M\Omega}\sum_G G^4|\varphi_G|^2\frac{1}{(vG)^2+(DG^2)^2}\,. \tag{36}$$

The presence of a small concentration of substitutional impurities drastically alters the motion of the system. Taking, for example, the impurity potential to be

$$\varphi(x)=W\cos G_0x\,, \tag{37}$$

leads to a drag force

$$F(v)=\left(\frac{cG_0^2W^2}{2m\Omega}\right)\frac{v}{v^2+(DG_0)^2}\,. \tag{38}$$

At low velocities, where F is linear in v, the drag force can be quite large, since the slope is proportional to $1/T^2$. Thus, a few impurities at low temperature can effectively dampen the motion of the chain.

One notes in passing that this particular model of an impure chain compound produces a highly nonlinear set of voltage-current characteristics in the charge-density wave and superionic-conductor applications of the model. The drag force is, within constants, equal to the potential difference across the length of the sample, while the drift velocity is directly proportional to the current. Above a critical value of the current ($v>DG_0$) the differential resistance becomes negative. Similar behavior should be true for any liquid near a critical point. More will be said about this in the last section.

Consider a three-dimensional system in the isotropic Debye approximation. Here, the auto-pair-correlation function is[8]

$$G_{\mathrm{self}}(\vec{Q},t)=\exp\left[-\frac{3k_BTQ^2}{m\omega_D^2}\left(1-\frac{\sin\omega_Dt}{\omega_Dt}\right)\right]. \tag{39}$$

The short-time behavior of G_{self} is Gaussian as in one dimension (or two dimension), so that the high-frequency behavior of $S_{\mathrm{self}}(\omega)$ is Gaussian. The long-time behavior of G_{self} is bounded and oscillatory. The velocity autocorrelation function does not continuously increase, as in one dimension, but approaches a finite limit.[12] Although the Fourier transform of $G_{\mathrm{self}}(t)$ cannot be obtained analytically for all values of

$$\lambda=\frac{3k_BTQ^2}{M\omega_D^2}\,, \tag{40}$$

a low-temperature expansion is readily obtained by expanding $G_{\mathrm{self}}(t)$ in powers of λ and then performing the Fourier transform, i.e.,

$$S_{\mathrm{self}}(\vec{Q},\omega)=\frac{e^{-\lambda}}{\omega_D}\left[\delta\left(\frac{\omega}{\omega_D}\right)+\frac{\lambda}{1!}I_1+\frac{\lambda^2}{2!}I_2+\frac{\lambda^3}{3!}I_3+\cdots\right], \tag{41}$$

where

$$I_1=\begin{cases}\frac{1}{2} & \omega<\omega_D\\ \frac{1}{4} & \omega=\omega_D\\ 0 & \omega>\omega_D\,,\end{cases} \tag{42}$$

$$I_2=\begin{cases}\frac{1}{2}(1-\omega/2\omega_D) & \omega\leqslant 2\omega_D\\ 0 & \omega\geqslant 2\omega_D\,,\end{cases} \tag{43}$$

and

$$I_3=\begin{cases}\frac{1}{8}[3-(\omega/\omega_D)^2] & \omega\leqslant\omega_D\\ \frac{1}{16}(3-\omega/\omega_D)^2 & \omega_D\leqslant\omega<3\omega_D\\ 0 & \omega\geqslant 3\omega_D\,.\end{cases} \tag{44}$$

The first term in S_{self} representing "recoilless emission," arises from G_{self} being finite as $t\to\infty$ and does not contribute to the dynamical friction ($v>0$). The next term indicates that as $T\to 0$ the dynamical friction is

$$\vec{F}(\vec{v})=\frac{3\pi}{4M\omega_D^3}\times\sum_{\vec{G}}\vec{G}(\vec{v}\cdot\vec{G})G^2|\varphi_{\vec{G}}|^2\Theta(\omega_D-\vec{G}\cdot\vec{v})+O(k_BT)\,, \tag{45}$$

where $\Theta(\omega)=0$ if $\omega<0$, 1 if $\omega>0$, and $\frac{1}{2}$ if $\omega=0$. One sees from this "one-phonon" result that the frictional force (i) depends upon the form of the impurity potential, (ii) varies linearly with the velocity up to some critical velocity, and (iii) vanishes thereafter. The latter characteristic arises from the absence of multiphonon processes in a classical system at zero temperature.

The two-dimensional system within the Debye approximation is anomalous. The velocity autocorrelation function is quadratic in t for small times and is logarithmic in t for large times. The functional form of

$$G_{\mathrm{self}}(\vec{Q}, t) = \exp\left[-\gamma\int_0^{\omega_D t} dx\left(\frac{1-\cos x}{x}\right)\right], \tag{46}$$

where

$$\gamma = \frac{4k_B T Q^2}{m\omega_D} \tag{47}$$

is Gaussian for small times and varies as $(\omega_D t)^{-\gamma}$ for large times. This power-law behavior introduces a dynamical, critical index into the problem since G_{self} ceases to be integrable when $\gamma \leqslant 1$. The Fourier transform $S_{\mathrm{self}}(\omega)$ then exhibits critical behavior (a Stanley-Kaplan transition[13,14]) as a function of γ (see Fig. 3): For $\gamma > 1$, $S_{\mathrm{self}}(\omega)$ is bounded and approaches a finite value (dependent upon γ) with zero slope as $\omega \to 0$. For $\gamma = 1$, $S_{\mathrm{self}}(\omega)$ diverges logarithmically as $\omega \to 0$. For $\gamma < 1$, $S_{\mathrm{self}}(\omega)$ diverges as $\omega^{-(1-\gamma)}$ as $\omega \to 0$. As γ approaches zero, the leading term of S_{self} (and subsequent terms) can be computed by breaking up the time integration into two parts. One chooses a $t_{\max}$ such that (i) $G_{\mathrm{self}}(t_{\max})$ takes its asymptotic form, and (ii) $\gamma G_{\mathrm{self}}(t) \ll 1$ when $|t| \lesssim t_{\max}$. The first integral $(0 < t \leqslant t_{\max})$ can be expanded in powers of γ while the second integral $(t_{\max} \leqslant t < \infty)$ can be evaluated analytically. One finds to leading order that

$$S_{\mathrm{self}}(\vec{Q}, \omega) \approx \frac{\gamma}{2|\omega|}\begin{cases} 1 & |\omega| < \omega_D \\ \frac{1}{2} & |\omega| = \omega_D \\ 0 & |\omega| > \omega_D \,. \end{cases} \tag{48}$$

This one-phonon result indicates that the dynamical friction [see Eqs. (22) and (23)] at zero temperature is a constant up to some critical velocity and vanishes thereafter (step function). To leading order in γ for nonzero temperature (i.e., for $\gamma \neq 0$) the force of friction (i) rises sharply ($F \sim v^{\gamma}$ where $\gamma \ll 1$) from zero to the one-phonon constant value, (ii) remains essentially constant up to the one-phonon critical velocity, and (iii) drops sharply (Gaussian-multiphonon tail) thereafter. Above the critical temperature (i.e., the temperature at which γ becomes 1), the force of friction rises linearly with velocity, then turns over and drops. This behavior is characteristic of an ordinary fluid.

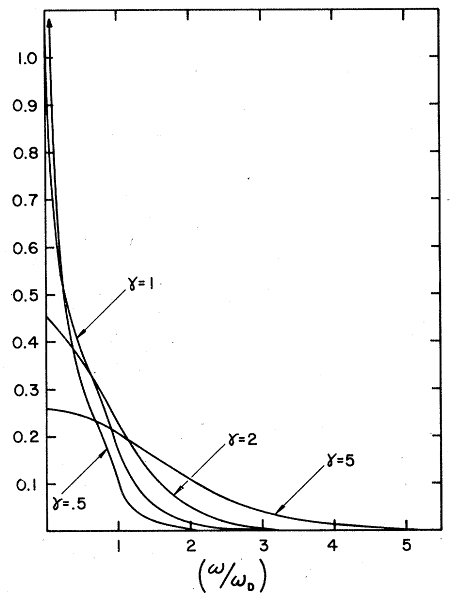

FIG. 3. $S_{\mathrm{self}}(Q, \omega)$ (dimensionless) for the two-dimensional Debye model versus ω/ω_D for various values of γ.

Actually, our model for impurities does not include impurities in the external potential which must be included if we are to apply this model to hollandite,[3] for example. Therefore, let us consider a model in which the impurities reside in the external potential rather than in the chain of atoms. In particular, our model will now consist of a chain of harmonically bound atoms interacting with an external potential of the form

$$\sum_n \Phi(\vec{r}_n) = \sum_j \lambda_j \varphi(\vec{R}_n - \vec{R}_j)\,, \tag{49}$$

where $\vec{R}_n$ is the position of an atom in the moving lattice (i.e., distortable chain in the one-dimensional problem), $\vec{R}_j$ is the position of an impurity (in the nondistortable external potential), and λ_j is defined as previously. The fluctuation-dissipation-theorem arguments used previously now give the following expression for the friction coefficient Γ:

$$\overleftrightarrow{\Gamma}(\vec{v}) = \left(\frac{\pi}{2k_B T}\right) c(1-c) \sum_{\vec{k}} \overleftrightarrow{\mathrm{kk}}\varphi(\vec{k}) S_{\mathrm{pure}}(\vec{k}, \omega = \vec{k}\cdot\vec{v}) \tag{50}$$

for dilute impurities. This is simply Eq. (23) with $\vec{k}$, any general wave vector, replacing the reciprocal lattice vector $\vec{G}$. Calculations for the three-dimensional Debye model using this expression give results not qualitatively different from Eq. (45).

For the one-dimensional model at $T = 0$, we may simply integrate Eq. (24) over G (treating G as any wave vector and not a reciprocal lattice vector) to obtain $F(v) \sim 1/v$. Since we do not have a rigorous expression for S_{self} in the one-dimensional model for general temperature, we cannot give a rigorous expression for Γ in this new model. To get an idea of how Γ behaves for low values of v at low temperatures, consider S_{pure} for the one-dimensional Debye model. In this model

$$S_{\text{pure}}(\vec{k},\omega)=\int dt\, e^{i\omega t}\sum_{nn'} e^{-g_{nn'}(\vec{k},t)}e^{ik(R_n-R_{n'})}, \tag{51}$$

where[8]

$$g_{nn'}(k,t)=\frac{k^2k_BT}{2m}\,\frac{l}{\pi}\int_0^{\pi/a}dq\left[\frac{\sin^2\frac{1}{2}q(R_{nn'}-ct)}{c^2q^2}+\frac{\sin^2\frac{1}{2}q(R_{nn'}+ct)}{c^2q^2}\right], \tag{52}$$

$R_{nn'}=R_n-R_{n'}$, l is the chain's lattice constant, and c is the speed of sound. In the low-temperature limit [k is kept from getting too large by the $|\varphi(k)|^2$ factor in Eq. (50), which falls off with increasing k], g is sizable only when either $|R_{nn'}|\gg ct$ or $|R_{nn'}|\ll ct$. For the low-frequency limit we are interested in the long-time behavior, but of course $R_{nn'}$ can still become much greater than ct. Thus, we may replace g by the asymptotic forms

$$g_{nn'}(k,t)\approx\frac{k^2k_BTlt}{4mc} \tag{53a}$$

for $t>R/c$ and

$$g_{nn'}(k,t)\approx\frac{k^2k_BTl}{4mc^2}\,|R_{nn'}| \tag{53b}$$

for $R>ct$. We now substitute these expressions into Eq. (52) and evaluate the integral over t to obtain

$$S_{\text{pure}}(k,\omega)\propto\sum_{nn'}e^{ikR_{nn'}}\left[\frac{\sin\left(\frac{\omega R_{nn'}}{c}\right)}{\omega}+\frac{\frac{k^2k_BTl}{4mc^2}}{\omega^2+\left(\frac{k^2k_BTl}{4mc^2}\right)^2}\right]\exp\left(-\frac{k^2k_BTl}{4mc^2}\,|R_{nn'}|\right). \tag{54}$$

Since the last factor in the above equation should fall off with increasing k much more rapidly than $|\varphi(k)|^2$ (because $k_BT\approx mc^2$) for all $R_{nn'}$ except $R_{nn'}=0$, we may neglect the k dependence of $|\varphi(k)|^2$ and simply integrate the above expression over k to obtain $\Gamma(v)$ for Eq. (50):

$$\begin{aligned}\int dk\,S_{\text{pure}}(k,\omega)\sim|\varphi(0)|^2\sum_{\substack{nn'\\ n\neq n'}}\frac{\sin\omega\frac{R_{nn'}}{c}}{\omega}\left(\frac{4\pi mc^2}{k_BTlR_{nn'}}\right)^{1/2}\exp\left(-\frac{mc^2|R_{nn'}|}{k_BTl}\right)\\ \times\sum_{nn'}\int dk\,|\varphi(k)|^2\frac{(k^2k_BTl/4mc^2)}{\omega^2+\left(\frac{k^2k_BTl}{4mc^2}\right)^2}\exp\left(-\frac{k^2k_BT|R_{nn'}|l}{4mc^2}\right)e^{ikR_{nn'}}+\int dk\,|\varphi(k)|^2,\end{aligned} \tag{55}$$

where the last term is part of the $R_{nn'}=0$ term. We are interested in the behavior of Eq. (55) for small v (and hence small ω). Each term in the first summation is well behaved for small ω and the summation converges rapidly because the summand falls off rapdily with increasing $R_{nn'}$. The last term obviously converges. The integral over k in the second term is well behaved for all $\omega\neq 0$. To study its behavior as v (and hence ω) approaches zero, we consider the contribution to the integral from a small region around $k=0$ and set $\omega=vk$. This contribution to the integral is

$$|\varphi(0)|^2\int_{-\epsilon/2}^{\epsilon/2}dk\frac{(k_BTl/4mc^2)}{v^2+\left(\frac{kk_BTl}{4mc^2}\right)^2}\propto\arctan\left(\frac{\epsilon k_BTl}{8mc^2v}\right),$$

where $\epsilon\ll 1/l$, which is well behaved as $v\rightarrow 0$. Since the summand falls off exponentially with $|R_{nn'}|$ for large $|R_{nn'}|$, the summation converges. Thus, we see from Eqs. (22) and (50) that $\Gamma(v)$ approaches a finite constant as $v\rightarrow 0$, and hence, the force of friction will be linear in v for small v. This, along with the fact that, for larger v, the force of friction must behave for low temperatures as a function of velocity as it does at $T=0$ (i.e., it must fall off as $1/v$), implies that the velocity dependence of $F(v)$ in this model is qualitatively similar to that given in Eq. (38).

VI. QUANTUM-MECHANICAL EFFECTS AND MULTIPHONON CONTRIBUTIONS TO THE FRICTION

In this section we will discuss briefly some quantum-mechanical effects that were neglected in the previous sections. Although we saw that multiphonon processes are not important at low temperatures in a classical system, because of quantum-mechanical effects, they can be as important at low temperatures in some cases as the contributions to the force of friction due to impurities, discussed in Sec. V.

For simplicity we will consider only the three-dimensional system in order to avoid the abnormalities caused by the lack of ordering in lower dimensions. Many of the qualitative conclusions, however, will probably also apply to such lower-dimensional systems. Also, all real crystals are three dimensional (even if the interface between two crystals sliding relative to each other is not).

If the quantum, instead of the classical, fluctuation-dissipation theorem is used in going from Eq. (20) to Eq. (23), the force of friction defined in Eq. (21) is given by

$$\vec{F} = \frac{\pi}{2\hbar}\sum_{\vec{G}}\vec{G}\,|\varphi_{\vec{G}}|^2[1-\exp(-\hbar\vec{G}\cdot\vec{v}/k_BT)] \times S(\vec{Q}=\vec{G},\,\omega=\vec{G}\cdot\vec{v})\,. \tag{56}$$

For $k_BT \gg \vec{G}\cdot\vec{v}$, this expression reduces to the result expressed by Eqs. (22) and (23).

In an isotropic harmonic model for a pure crystal $S(\vec{Q},\omega)$ is given by

$$S(\vec{Q},\omega) = \int dt e^{-i\omega t}\exp\left[-\frac{\hbar Q^2}{NM}\sum_{\vec{q}}\omega_{\vec{q}}^{-1}(2\langle n_{\vec{q}}\rangle+1)\right]\sum_{jl}\exp\left\{\frac{\hbar Q^2}{2NM}\sum_{\vec{q}}\omega_{\vec{q}}^{-1}[(2\langle n_{\vec{q}}\rangle+1)\cos\theta_{jl}(t)+i\sin\theta_{jl}(t)]\right\}, \tag{57}$$

where

$$\theta_{jl} = \omega_{\vec{q}}t + \vec{q}\cdot(\vec{R}_j-\vec{R}_l)\,.$$

Here, $\omega_{\vec{q}}$ and $\langle n_{\vec{q}}\rangle$ are, respectively, the frequency and mean occupation number of a phonon state of wave vector $\vec{q}$, N is the number of atoms in a crystal, M is the atomic mass, and $\{\vec{R}_j\}$ denotes the equilibrium positions of the atoms. As we mentioned earlier, one of the most important quantum-mechanical effects in this problem is the possibility of a sizable contribution from multiphonon processes at low temperatures. Thus, let us consider the two-phonon contribution to S at $T=0$. Following Kittel,[8] we expand the second exponential in Eq. (50) to second order, keeping only the terms with a time dependence $\exp(i\omega_q t)$, which result in phonon creation. After performing the sum on j and l we obtain

$$\left(\frac{\hbar^2Q^2}{2M}\right)^2\sum_{\vec{q}}\frac{1}{\hbar^2}\frac{1}{\omega_{\vec{q}}\omega_{\vec{Q}-\vec{q}}}\delta(\omega-\omega_{\vec{q}}-\omega_{\vec{Q}-\vec{q}})\langle n_{\vec{q}}+1\rangle\langle n_{\vec{Q}-\vec{q}}+1\rangle\,, \tag{58}$$

where $\langle n_{\vec{q}}\rangle$ becomes zero at $T=0$. A similar contribution was obtained earlier for charge-density waves by one of the authors.[15] Comparing the intensity of such two-phonon processes with the integrated intensity of the one-phonon processes found in Ref. 8, we see that the two-phonon process can be smaller by a factor of the order of

$$\hbar^2Q^2/M\hbar\omega_D\,, \tag{59}$$

where ω_D is the Debye frequency. If we take $Q=G\approx 10^8\ \text{cm}^{-1}$, $M\sim 10^{-22}$ g, and $\hbar\omega_D\approx 0.025$ eV, we find that this factor is $\approx\frac{1}{250}$. The strength of the impurity contribution to S [i.e., the first term in Eq. (29)] is of the order of the product of the impurity concentration and the intensity of the one-phonon contribution to S. Thus, we see that for very pure systems (i.e., systems with impurity concentrations below 0.4%) and temperatures small compared to the Debye temperature, it might be necessary to go beyond the classical treatment given in previous sections and include low-temperature multiphonon processes which do not occur in the classical treatment.

Another consequence of quantum mechanics follows from Eq. (56). For $\hbar\vec{G}\cdot\vec{v} > k_BT$, the force of friction might have a different velocity dependence that that predicted by Eq. (23). As we predicted for the one- and three-dimensional models with dilute impurities studied earlier, we would have a damping constant which limits to a constant at small velocities. This, of course, depends on the frequency dependence of S_{self} when quantum mechanics is used. From Ref. 8 we find that at $T=0$ in the Debye approximation

$$S_{\text{self}}(Q,t) = e^{-[W(0)-W(t)]}\,,$$

where

$$W(t) = \frac{\hbar Q^2}{2M}\frac{3}{\omega_D^3}\int_0^{\omega_D}\omega e^{i\omega t}d\omega\,. \tag{60}$$

Expanding $S_{\rm self}$ in powers of $W(t)$, which is valid for $\hbar^2Q^2/2M \ll \hbar\omega_D$, we obtain the one-phonon contribution from the first-order term, the two-phonon process in second order, etc. Expanding $S_{\rm self}$ $S_{\rm self}(Q,t)$ and Fourier transforming, we obtain for the one-phonon contribution

$$\frac{3\hbar Q^2}{2M}\frac{3}{\omega_D^3}\,.$$

This implies that at k_BT small compared to $\hbar\vec{\rm v}\cdot\vec{\rm G}$, the force of friction is still linear in v and, for v smaller than $k_BT/\hbar G$, it is quadratic. Higher-order terms in $S_{\rm self}(\vec{\rm Q},\omega)$ approach zero as ω approaches zero even faster than the first-order term and thus do not qualitatively change the above result. It should be noted that at room temperature the critical velocity $k_BT/\hbar G$ is of the order of 10^5 cm/sec. This critical velocity will, of course, decrease linearly with temperature. Thus, at temperatures well below the Debye temperature, the force of friction could actually be quadratic rather than linear in v for sufficiently small v. This could lead to interesting nonlinear conduction with current proportional to electric field.

Multiphonon processes in a pure crystal, which may be calculated from Eq. (58), are critically dependent on whether the system is commensurate or incommensurate. If it is commensurate, $\vec{\rm Q}=\vec{\rm G}$ is a reciprocal lattice vector common to both the sliding crystals. Since $\omega_{\vec{\rm G}-\vec{\rm k}}=\omega_{\vec{\rm k}}$, the two-phonon process is then proportional to the one-phonon density of states evaluated at $\omega/2$ for small ω. From Eq. (22), this implies that a contribution from two-phonon processes to the force of friction is linear in the velocity when $\hbar\vec{\rm G}\cdot\vec{\rm v}\gg k_BT$. For the incommensurate case, however, $\vec{\rm G}$ is a reciprocal lattice vector of the potential φ but not of the crystal in which phonons are being excited. Thus, since $\omega_{\vec{\rm G}-\vec{\rm l}}\neq\omega_{\vec{\rm l}}$, the two-phonon process will no longer vanish as ω approaches zero (at $T=0$). This implies that the force of friction is linear in v when $\hbar\vec{\rm G}\cdot\vec{\rm v}<k_BT$ and independent of v otherwise.

VII. FRICTION DUE TO ELECTRONIC EXCITATIONS

Since many systems with charge-density waves also have some free carriers, it is important to consider the contribution to the force of friction due to electronic excitations. This is the damping mechanism studied by Lee and Rice.[7] To treat this contribution, it is convenient to start with Eq. (20), using the single-band free-electron or Hartree-Fock susceptibility for the free carriers available to damp the motion of the sliding periodic potential (which can be taken to represent the charge-density wave),

$$\operatorname{Im}\chi(Q,\omega)=\frac{1}{N}\sum_{\vec{\rm k}}[f(\epsilon(\vec{\rm k}))-f(\epsilon(\vec{\rm k}+\vec{\rm Q}))]\delta(\epsilon(\vec{\rm k}+\vec{\rm Q})-\epsilon(\vec{\rm k})-\hbar\omega)\,, \tag{61}$$

where $\vec{\rm k}$ is summed over the Brillouin zone and $f(\epsilon(\vec{\rm k}))$ is the Fermi function. For small ω,

$$S_{\rm pure}(Q,\omega)=\frac{k_BT}{\omega}\operatorname{Im}\chi(Q,\omega)=k_BT\,\frac{1}{N}\sum_{\vec{\rm k}}\left(-\frac{\partial f(\epsilon(\vec{\rm l}))}{\partial\epsilon}\right)\delta(\epsilon(\vec{\rm l}+\vec{\rm Q})-\epsilon(\vec{\rm l})-\hbar\omega)\,. \tag{62}$$

At low temperatures compared to the Fermi energy, $-\partial f/\partial\epsilon$ is almost a δ function, which restricts $\epsilon(\vec{\rm k})$ to the Fermi surface. Thus, the electronic excitations contribute for small ω only if it is possible to have both $\epsilon(\vec{\rm k})$ and $\epsilon(\vec{\rm k}+\vec{\rm G})$ on the Fermi surface. This is impossible in one dimension, but is possible in two or three. Therefore, the electronic contribution in the absence of impurities does not contribute significantly in one dimension (it is zero if $T=0$), but does in two or three. To estimate the importance of the electronic damping mechanism in three dimensions we must compare it with the one-phonon term for an impure lattice (the dominant phonon contribution as was seen in previous sections). The one-phonon contribution to the force of friction is given by

$$F_p\sim\frac{c}{\hbar}\vec{\rm G}\left(\frac{3\vec{\rm v}\cdot\vec{\rm G}}{\omega_D^2}\right)|\varphi_{\vec{\rm G}}|^2\,. \tag{63}$$

The electronic contribution F_l is given by

$$F_l\sim\vec{\rm G}\left(\frac{\hbar\vec{\rm v}\cdot\vec{\rm G}}{\epsilon_F}\right)|\varphi_{\rm G}|^2\frac{1}{N}\times\sum_{\vec{\rm k}\in\text{ Fermi surface}}\delta(\epsilon(\vec{\rm l}+\vec{\rm G})-\epsilon(\vec{\rm k}))\,. \tag{64}$$

Thus the ratio of the two contributions is given by

$$\frac{F_l}{F_p}\sim\left(\frac{\hbar\omega_D}{\epsilon_F}\right)^2\frac{1}{c}\,. \tag{65}$$

Since $\hbar\omega_D/\epsilon_F\approx 1\%$ we expect the phonon contribution usually to dominate over the electronic contribution. The physical reason for this is that the phonons are distributed over a smaller energy range ($\approx\hbar\omega_D$) than that of the electrons ($\approx\epsilon_F$), and hence the phonons have a higher density of states. Of course for the charge-density-wave applications of this model it must be remembered that, if the phonons we are considering are really phasons, which have a very steep velocity[16] and hence a high value for ω_D, it is quite possible that the electronic contribution may be comparable to the phonon contribution.

VIII. CONCLUSIONS AND APPLICATIONS OF THE MODEL TO VARIOUS SYSTEMS

In this section we will conclude with a discussion of several systems to which the model should apply.

Hollandite is a one-dimensional ionic conductor in which mobile potassium ions move in separate one-dimensional channels.[3] The potassium ions in any one channel should be well approximated by a one-dimensional highly-correlated fluid (the ions do not solidify at low temperatures, because of the one-dimensional nature of the system). Perhaps TTF-TCNQ can also be treated as a one-dimensional system in a three-dimensional lattice if the phasons (i.e., phonons of the charge-density wave) are sufficiently one dimensional. The damping of the motion of the mobile ions in an applied external electric field is due to two sources the creation of excitations in the potassium-ion fluid and the creation of excitations in the rest of the lattice. As mentioned earlier, within the context of perturbation theory, these contributions may be simply added together. We expect the low-order perturbation theory used in this paper to be valid if the interaction between the potassium ions and the rest of the lattice is fairly weak, which it must be if the ions are to be mobile rather than pinned in place. The contribution to the force of friction due to excitations in the potassium system is given by Eq. (38), whereas the contribution due to the rest of the lattice (which is three dimensional) is linear in the velocity for low velocities, as predicted in Eq. (45). We then see that for low temperatures and low velocities, the potassium-ion contribution must be the dominant one. We find the steady-state current as a function of the applied field by setting the force due to the applied field equal to this expression for the friction and solving for the velocity (which is essentially the electric current). As pointed out earlier, this leads to highly nonlinear behavior with a negative-resistance region and a critical field above which a steady state is not possible since the system will accelerate (i.e., the current continually increases). Actually, it will not accelerate indefinitely, since the damping due to the rest of the lattice will eventually bring it back into steady-state motion. Nevertheless, one would expect a jump in the conductivity above this critical field. These effects are illustrated in Fig. 4. To find the temperature at which such anomalous nonlinear effects first appear, we add a term Γv to Eq. (38) to account for the damping due to the rest of the lattice. The anomaly will only occur if the resulting force of friction as a function of v has a relative maximum and minimum. Thus, we set dF/dv equal to zero and determine the temperature for which the resulting equation has solutions. Using $W=2$ eV, lattice constant $a=2.9$ Å, $\Gamma/m=150\ \mathrm{cm}^{-1}$, $m=$ the mass of a potassium ion, and $\Omega=0.4\times10^{14}$ rad/sec, which is determined from the force constant which is required to give the correct equilibrium configuration for the potassium ions,[17,3] we obtain a critical temperature of 6000 K. Thus, the effect should always be there at reasonable temperatures. To estimate the critical electric field at this temperature, we simply set $dF/dv=0$, solve for v, and substitute this value of v into the equation for F. We then divide the resulting force of friction per ion by the electrical charge of an ion to obtain an electric field of about 3×10^6 V/cm, a typical field for dielectric-breakdown phenomena. This critical field increases when the temperature is lowered. Thus, we expect an anomaly similar to the one illustrated in Fig. 4 when the electric field exceeds this value or higher for temperatures below 6000 K. Below this field, the conduction is linear, but the conductivity decreases to zero as T^2, as the temperature is reduced to zero [this result is obtained from Eq. (38)]. This is not because of the exact quantum-mechanical theorems which state that the conductivity is zero in a one-dimensional system.

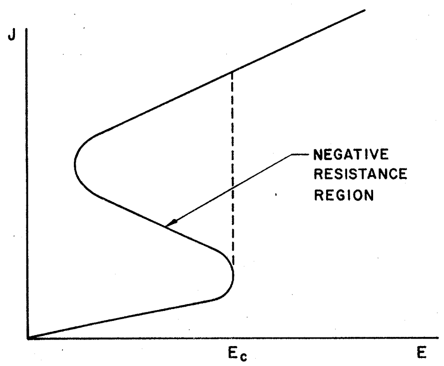

FIG. 4. Sketch of the electric current versus applied dc electric field for our model applied to hollandite. E_c is the electric field at which the system jumps into a highly conducting state. (At this point the current follows the dotted line.)

Rather, this is a classical result which states that only low-velocity steady-state motion of a chain of ions is possible until a rather high critical field is reached, at which point the system jumps into a more highly conducting state. The linear proportionality of the low-field conductivity with T^2 implies that the conductivity is not simply a thermally activated process; it most likely involves interesting collective effects. (Of course we can never measure the true dc conductivity of hollandite because of finite-channel-end effects, but the effects that we have predicted should be seen in relatively low-frequency ac conductivity.)

If the two-dimensional charge-density wave in a layered compound could be set in motion so as to contribute to the conductivity, our model predicts intrinsically nonlinear conductivity below the Stanley-Kaplan transition temperature, since the force of friction is not linear in the velocity. In particular, we expect a current proportional to $E^{1/\lambda}$, where λ is proportional to the temperature. Thus, the conduction should be nonlinear and quite low at low-temperatures. There are, however, theorems which state that a charge-density wave is not an ordered state in fewer than three dimensions, and it is not clear what effect fluctuations in the degree of order of the charge-density wave will have on the electrical conduction. Our two-dimensional model should also apply to monolayer thin films for which NMR might be an appropriate probe of the motion of the film.

The one system for which nonlinear conduction, due to charge-density waves breaking loose from their pinning potential, has been observed is $NbSe_3$.[18] In this system, where the charge-density waves are three dimensional, our three-dimensional model should be appropriate. Our model predicts a damping linear with the velocity, and hence a linear relation between current and applied field. Thus, we predict linear conduction once the charge-density wave becomes depinned, which is consistent with experiment.[18] Exactly how the charge-density wave becomes depinned is beyond the scope of this paper, as it requires a treatment of the intermediate behavior between static and dynamic friction. The correct mechanism could either be a quantum-tunneling mechanism as predicted by Bardeen,[19] or more likely, a mechanism involving the creation of dislocations, such as the mechanism which gives rise to the Peierls stress required to inelastically strain a solid.[20]

Perhaps a more speculative application of our theory is to friction between real solid surfaces. Friction between most surfaces is probably due to the shearing[21] of aspherities from the sliding surfaces (which results in wear). If, however, we consider sliding of ideal surfaces such as cleaved graphite crystals, our treatment might be applicable. Some of our ideas can perhaps be tested by studying the width of the rigid-layer modes in layered compounds, but this requires further study. Our theory does say something about how the force of friction depends on the excitation spectrum of the surfaces in contact, which might lead to some useful qualitative predictions about friction and lubrication, which requires further study.

We close with a brief discussion of the conventional result that the force of friction is proportional to the load. It is believed that the actual area of contact of aspherities of two surfaces in contact is proportional to the load (force pushing the two surfaces together).[21] Let us discuss friction due to creation of excitations rather than shearing of aspherities. We will assume that the microscopic interaction between the two sliding crystals [i.e., $\varphi(\vec{r})$] is repulsive, which should be the case for solids which do not chemically bind together. If the potential between a pair of atoms varies as $1/r^n$, where n is a large integer and r is the distance between the ions, we find that the resulting periodic interaction $\varphi(\vec{r})$ is almost proportional to the force pushing such a pair of atoms in the two surfaces in contact together (i.e., proportional to the load per atom or load per unit area of contact).

Since we stated before that the area of contact of the aspherities is also proportional to the load, $\varphi(\vec{r})$ should be nearly constant. However, since the expression for the force of friction that we obtained [i.e., Eq. (23)], has an overall proportionality to the area of contact, we expect the force of friction to be proportional to the load, the usual relationship.

ACKNOWLEDGMENT

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