Anomalous Ultrasonic Attenuation due to Sliding Charge-Density Waves

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The influence of sliding charge-density waves on the elastic properties of quasi-one-dimensional conductors is investigated. By use of an extension of the Fukuyama-Lee-Rice model the imaginary part of the self-energy of acoustic phonons is shown to diverge in lowest-order perturbation theory above the threshold field. With the summation of leading divergent contributions, the resulting expressions account well for the observed field dependence of the elastic modulus and the ultrasonic attenuation α. In particular, a $\omega^{3/2}$, instead of the usual ω^2, law is predicted for α above the threshold field.

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A large increase of the ultrasonic attenuation above the onset of nonlinear conduction has recently been observed in several quasi-one-dimensional conductors.[1-4] The observed increase by at least 2 orders of magnitude seems not to be a critical phenomenon restricted to the immediate neighborhood of the threshold electric field E_c. Experiments suggest rather that it is present throughout the sliding charge-density-wave region and changes only slowly as function of the electric field on a scale set by E_c.

The occurrence of the above effect is somewhat surprising: In the noncritical region, the ultrasonic attenuation is given by integrals over densities of states (phonons or order-parameter fluctuations). Since only a small number of excitations are substantially altered by the electric field (namely long-wavelength phase modulations of the order parameter), one expects only a small field-dependent effect on the ultrasonic attenuation. Things can, however, dramatically change if lowest-order perturbation theory for the elastic properties breaks down because of the occurrence of divergences. Such a case has recently been found in the ultrasonic attenuation in pure incommensurate systems.[5] A diverging viscosity has also been discussed[6] and experimentally verified[7] for the smectic-A phase of liquid crystals. Related to this are also predicted anomalies in $\mathbf{q}$ and ω space of an isotropic ferromagnet with more than one component for which, however, no clear experimental evidence has been found up to now.[8] In the following, it will be shown that similar anomalies occur in the elastic properties of charge-density-wave systems above the threshold field. These anomalies are due to infrared divergences in the perturbation expansion which are not confined to the thresholdlike critical divergences but exist for all fields above the threshold. I will restrict myself to these kinds of divergences and exclude critical divergences from discussion. In contrast to Ref. 9, low-lying phase modulations as well as randomly distributed impurities play an important role in the approach.

Let the phase of the complex order parameter of the charge-density-wave state be denoted by ϕ. Furthermore, Q_λ denotes the normal coordinates of acoustic phonons with branch index λ. It is convenient to split ϕ and Q into fluctuating and nonfluctuating parts: $\phi=\bar{\phi}+vt+\tilde{\phi}$, $Q_\lambda=\bar{Q}_\lambda+\tilde{Q}_\lambda$. Here, v is the velocity of the sliding charge-density wave and $\bar{\phi}$ can be put to zero. If the Fukuyama-Lee-Rice model[10-12] is used and if acoustic phonons and their coupling to charge-density waves are included, $\tilde{\phi}$ satisfies the following Langevin equation within a phenomenological approach:

$$\left(\frac{1}{\tau_0}\frac{\partial}{\partial t}-\nabla^2\right)\tilde{\phi}=e^*E-\frac{v}{\tau_0}-\sin(vt+\tilde{\phi})\zeta_1+\cos(vt+\tilde{\phi})\zeta_2-\frac{\partial F'}{\partial\tilde{\phi}}+\epsilon. \tag{1}$$

τ_0 is a relaxation time, e^* an effective charge, and E the applied electric field. ζ_i and ϵ are Gaussian noises due to impurity and thermal fluctuations, respectively. Their correlation functions are

$$\langle\zeta_i(\mathbf{x},t)\zeta_j(\mathbf{x}',t')\rangle=\delta_{ij}\delta(\mathbf{x}-\mathbf{x}')\gamma^2, \tag{2}$$

$$\langle\epsilon(\mathbf{x},t)\epsilon(\mathbf{x}',t')\rangle=2\Gamma\delta(\mathbf{x}-\mathbf{x}')\delta(\mathrm{t}-\mathrm{t}'), \tag{3}$$

where the angle brackets denote the corresponding averages. γ is proportional to the strength of the pinning potential and Γ is related to τ_0 by the Einstein relation. F' describes the interaction between the phase of the order parameter and acoustic phonons and reads in $\mathbf{k}$ space

$$F'=\sum_{\mathbf{k}\mathbf{q}\lambda}\frac{V(\mathbf{k}\lambda)}{\sqrt{V}}Q(\mathbf{k}\lambda)\phi(-\mathbf{k}-\mathbf{q})\phi(\mathbf{q}), \tag{4}$$

with

$$V(\mathbf{k}\lambda)=\frac{i\gamma^2}{4\pi\omega_A}\sum_{\alpha\beta}k_\alpha V_{\alpha\beta}e_\beta(\mathbf{k}\lambda). \tag{5}$$

$V_{\alpha\beta}$ is a symmetric tensor, V the volume of the solid, and $\mathbf{e}(\mathbf{k}\lambda)$ the polarization vector of the phonon $(\mathbf{k}\lambda)$. Origi-

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nally, the acoustic phonons coupled to product states of the amplitude of the order parameter.[5] Because the dynamics of the amplitude is fast compared to that of ϕ and Q_λ, I have integrated out the amplitude fluctuations in the free energy. This yields the effective coupling, Eq. (5), where ω_A denotes the frequency of amplitude fluctuations. F' consists, in general, of more terms which contain spatial derivatives of ϕ. These terms, however, do not give rise to singular contributions to the elastic properties and are therefore omitted. Finally, the Langevin equation for $\tilde{Q}$ reads in $\mathbf{k}$ space

$$\left[\frac{\partial^2}{\partial t^2}+\gamma \mathbf{k}^2\frac{\partial}{\partial t}+c^2(\hat{\mathbf{k}}\lambda)\mathbf{k}^2\right]\tilde{Q}(\mathbf{k}\lambda,t)=-\frac{\delta F'}{\delta\tilde{Q}(-\mathbf{k}\lambda,t)}+\eta(\mathbf{k}\lambda,t). \tag{6}$$

η is a Langevin force connected to the damping constant γ by the Einstein relation. $c(\hat{\mathbf{k}}\lambda)$ is the velocity of sound of the branch λ in the direction $\hat{\mathbf{k}}=\mathbf{k}/|\mathbf{k}|$.

Expanding the right-hand side of Eq. (1) in powers of $\tilde{\phi}$, Eqs. (1) and (6) can be solved by iteration. In this way an expansion of the phonon self-energy $\Sigma(\mathbf{k}\lambda,\omega)$ in powers of $V_{\alpha\beta}$ and γ^2 can be obtained. Diagrams for Σ which are singular for all v's are shown in Fig. 1. The shaded triangle in the first line is defined by the second line. The small black triangle in the first line represents the interaction $V(\mathbf{k}\lambda)$. The small black square in the second line stands for an effective four-point vertex defined by the third line in Fig. 1. The empty square represents the third-order expansion coefficient of the trigonometric functions in Eq. (1) in the field $\tilde{\phi}$. The wavy line denotes the correlation function of the first two terms on the right-hand side of Eq. (1) for $\tilde{\phi}=0$ which can easily be evaluated from Eq. (2). The solid lines denote the response function of phase fluctuations. The solid lines with a circle stand for the corresponding correlation function.

An analysis of its perturbation expansion suggests that the phase response function has, at long wavelengths and low frequencies, the form

$$G^{-1}(\mathbf{k},\omega)=r+i\omega/\tau+w^2\mathbf{k}^2. \tag{7}$$

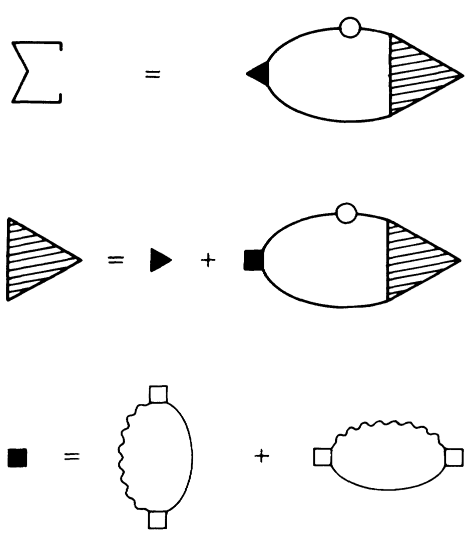

FIG. 1. Diagrams for the self-energy Σ of acoustic phonons.

The phase velocity w varies slowly with the electric field so that the free value $w=1$ can be used. Linear-response theory gives, moreover, for $v>0$ the relation

$$\frac{\partial v}{\partial(e^*E)}=\lim_{\omega\to 0}[i\omega G(0,\omega)]. \tag{8}$$

Equation (8) implies $r=0$ because v is a strictly increasing function of E and also $\tau=\partial v/\partial(e^*E)$. Keeping only zeroth- and second-order terms in the impurity concentration one obtains

$$\frac{1}{\tau}=\frac{1}{\tau_0}+\frac{\gamma^2}{8\pi(2\tau_0 v)^{1/2}}. \tag{9}$$

The leading correction to Ohm's law at high fields is the well-known term $\sim\sqrt{E}$. At small velocities we obtain $v\sim(E-E_c)^2$, i.e., a somewhat slower increase with E than in the infinite-range model[13] where the exponent is $\frac{3}{2}$. The difference between the two exponents is due to the fact that I do not take into account higher orders of the perturbation theory for τ and thus omit, in particular, terms which diverge for $v\to 0$.[14]

Taking the diagrams of Fig. 1 into account, I obtain for Σ

$$\Sigma(\mathbf{k}\lambda,\omega)=\frac{2V(\mathbf{k}\lambda)V(-\mathbf{k}\lambda)\Pi(\mathbf{k},\omega)}{1+u\Pi(\mathbf{k},\omega)}. \tag{10}$$

u is the expression for the black square in Fig. 1 and given by

$$u=-\gamma^2\Lambda/4\pi^2, \tag{11}$$

where Λ is a momentum cutoff. A more detailed analysis shows that the considered diagrams are sufficient if the space dimensions are not much lower than four [so that in the expansion of the trigonometric functions in Eq. (1) in powers of $\tilde{\phi}$ fourth- and higher-order terms are irrelevant and can be omitted] and γ is small (so that the lowest-order contributions in γ to τ and u are sufficient). Π is the expression for the bubble made up by one correlation and one response function. In the long-wavelength, low-frequency region it is given by[5]

$$\Pi(\mathbf{k},\omega)=\frac{ik_BT}{4\pi|\mathbf{k}|}\left\{\log\left[|\mathbf{k}|+\left(-\mathbf{k}^2+\frac{2i\omega}{\tau}\right)^{1/2}\right]-\log\left[-|\mathbf{k}|+\left(-\mathbf{k}^2+\frac{2i\omega}{\tau}\right)^{1/2}\right]\right\}. \quad (12)$$

Π exhibits singular behavior for small $\mathbf{k}$'s and ω's. In the limit $\omega\to 0$, $\mathbf{k}$ finite, one obtains

$$\Pi(\mathbf{k},0)\to k_BT/8|\mathbf{k}|. \quad (13)$$

The limit $\mathbf{k}\to 0$, ω finite, yields

$$\Pi(0,\omega)\to\frac{k_BT(1+i\,\mathrm{sgn}\omega)}{4\pi(|\omega|/\tau)^{1/2}}. \quad (14)$$

This hydrodynamic limit is important in connection with the viscosity and exhibits a square-root divergence. The same behavior is found for the limit $\mathbf{k}\to 0$, $\omega\to 0$, $\omega/|\mathbf{k}|=c(\hat{\mathbf{k}}\lambda)$ which is relevant for ultrasonic and Brillouin-scattering experiments.

The above divergences in Π for small $\mathbf{k}$'s and ω's are not restricted to the critical value $v\sim 0$, but persist over the whole sliding charge-density-wave region. They are due to the strong decay of acoustic phonons into product states of long-wavelength phase modulations of the order parameter. In view of these divergences the use of lowest-order perturbation theory for Σ [i.e., the neglect of Π in the denominator in Eq. (10)] is certainly not appropriate. For instance, Eq. (13) would imply that the static self-energy of acoustic phonons is $\sim|\mathbf{k}|$. An exact Bogoliubov inequality, however, tells us that this self-energy has to vanish for small $\mathbf{k}$'s at least as fast as $\mathbf{k}^2$. The set of diagrams generated by the first two lines in Fig. 1 represent the leading singular contributions which, if summed up, should yield a physically meaningful result.

Figure 2 shows the calculated real and imaginary parts of Σ from Eq. (10). The bars at the various quantities indicate that they have been made dimensionless by setting the bare phason velocity, the thermal energy, and the relaxation time τ_0 to one. The solid curves in Fig. 2 correspond to $\bar{\omega}=0.0002$. This frequency is so low that the approximate expression Eq. (14) for Π holds everywhere except for a very small interval of the electric field just above $\bar{E}_c$. As a result, both the real and imaginary parts of Σ vary very slowly with $\bar{E}$ excluding a small region just above $\bar{E}_c$. In this region, τ/τ_0 increases steeply from zero to one, causing a transition from the perturbative regime for Σ (where $u\Pi$ in the denominator can be neglected) to the collision-dominated regime where $u\Pi\gg 1$. For $\bar{\omega}=0.2$, the transition between the two regimes is very broad. The imaginary part of Σ again increases with an infinite slope at $\bar{E}_c$ as $(\bar{E}-\bar{E}_c)^{1/4}$. However, it reaches its maximum only slowly and then decays very smoothly towards higher fields. The real part of Σ is positive just above $\bar{E}_c$ and assumes then negative values towards higher fields. This means that the lattice first stiffens with increasing field and then softens. The reason for this behavior is rather simple: In the perturbative regime just above $\bar{E}_c$ we have $\Sigma\sim\Pi$ and the real part of Π is positive. At higher fields, the collision-dominated regime with $\mathrm{Re}\Sigma\sim 1/u$ is reached and u is negative according to Eq. (11).

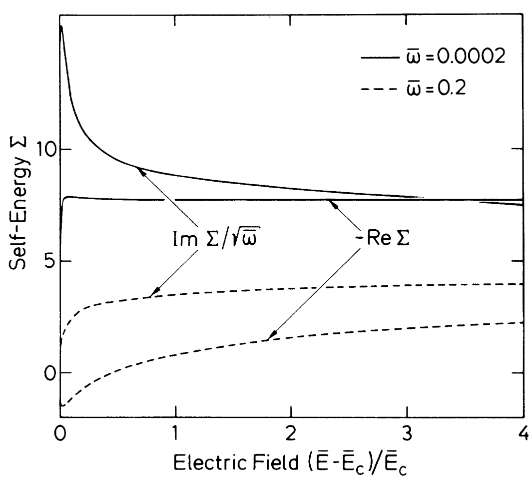

FIG. 2. Real and imaginary parts of Σ in arbitrary units as a function of the applied field for two frequencies $\bar{\omega}$ and $\bar{u}=-\frac{1}{3}$.

The above theory has several general implications which can be compared with experiment: (a) The changes in the elastic properties and the ultrasonic attenuation occur only for $\bar{E}\geq\bar{E}_c$. Below $\bar{E}_c$ there is no stiffening or softening of the lattice nor enhanced ultrasonic attenuation. The reason for this is that $\Sigma\to 0$ for $\bar{E}\downarrow\bar{E}_c$ because of $\tau/\tau_0\to 0$. $\tau/\tau_0\to 0$ means that v increases faster than linearly in $\bar{E}-\bar{E}_c$. Comparison with experiment shows that these properties of the above theory agree very well with the experimental observations; (b) The changes in the elastic properties set in discontinuously at $\bar{E}_c$. For instance, the slope of $\mathrm{Im}\Sigma$ is infinite at $\bar{E}=\bar{E}_c$. This discontinuous behavior is clearly seen in the experiments. An infinite slope is compatible with some of the experimental curves (for instance Fig. 6 of Ref. 3) but not with all of them (see, for instance, Fig. 4 of Ref. 3). The stiffening of the lattice just above $\bar{E}_c$ agrees with many of the experimental curves; (c) Both the real and imaginary parts of Σ vary slowly at higher fields on a scale determined by $\bar{E}_c$. This is in agreement with experiment and shows that the underlying phenomenon is not a critical property confined to the depinning transition at $\bar{E}_c$ but a property of the sliding charge-density-wave state; (d) The encountered divergences do not much affect the real part of Σ, but they do the imaginary part of Σ, causing an ultrasonic attenuation coefficient $\alpha\sim\bar{\omega}^{3/2}$ instead of the usual $\bar{\omega}^2$ law.

This makes the observed large enhancement of α and, at the same time, the rather small changes in the elastic properties above threshold understandable. Unfortunately, the available data do not admit a direct check of the predicted anomalous $\bar{\omega}^{3/2}$ behavior of α.

In conclusion, I have investigated the perturbation expansion for the change of elastic properties due to sliding charge-density waves in quasi-one-dimensional conductors. Using an extension of the Fukuyama-Lee-Rice model, I encountered divergences reminiscent of those in the longitudinal susceptibility of an isotropic ferromagnet with more than one component[8] or in the viscosity of smectic-A phases of liquid crystals.[6,7] As a result, the viscosity diverges at low frequencies like $\omega^{-1/2}$ due to the strong coupling of acoustic phonons to gapless phase modulations above the threshold field. If the most divergent diagrams are summed over, the theoretical field dependence of the elastic modulus and the ultrasonic attenuation agrees with the experiments.

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