Low-temperature strong pinning and creep of a three-dimensional charge-density wave

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Strong three-dimensional pinning and creep explain many low-temperature properties of systems with charge-density waves. Among these is the increase in the dielectric constant ε when the temperature increases and when the frequency decreases.

1. INTRODUCTION

Recent years have seen the discovery of many remarkable low-temperature properties of systems with waves of charge or spin density.[1,2] The temperature dependence of the dielectric constant has an additional sharp peak with a height that decreases as the frequency increases. The specific heat is a power function of temperature and is nonequilibrium, while the conductivity is of hopping origin. These properties suggest that the low-temperature state is glasslike.[3–5]

Any glass is characterized by the presence of metastable states separated by tall barriers. Usually calculating the energy of these barriers is extremely complicated. This paper considers a simple model of strong three-dimensional pinning, when sparsely spaced strong pinning centers (chain breakoffs) give rise to plastic deformations in charge-density waves (CDWs) and cause metastable states to form.

An impurity in such a metastable state acts on CDWs with a force directed against displacement.[6,7] On the other hand, impurities near which CDWs are in equilibrium act on CDWs with a randomly directed force dependent on the position of the impurity. After averaging over random position this force vanishes. In the event of strong pinning, the average force is proportional to the concentration of the impurities in a metastable state, while the barrier energy is independent of concentration and fairly low. Hence, only at extremely low temperatures can such metastable states survive long and determine the physical properties. As the temperature grows, their contribution to the force rapidly decreases and the dielectric constant grows.

This paper considers only the low-temperature region of strong pinning. The condition for strong pinning is found by applying the mean-field approximation to the interaction between chains. Unlike the one-dimensional case, in the three-dimensional the CDW–impurity interaction energy must exceed a critical value.[6,7] The neutral (dipole) or charged solitons generated on such impurities determine the low-temperature behavior of the specific heat and the dielectric constant.

2. DIPOLE SOLITONS

The systems considered below are three-dimensional objects. In the three-dimensional case there is a critical value for the energy of the CDW–impurity interaction below which an individual impurity does not give rise to pinning. We will see that this energy is equal to a quarter of the soliton energy. Impurities with an energy below this critical value lead only to weak collective pinning, when metastable states appear only as a result of the collective action of many impurities.[6–8] At these low temperatures we will be considering the collective-pinning force is exponentially small because of the long-range Coulomb interaction,[8] while the size of the region where there is short-range order is exponentially large. Hence, we can ignore collective pinning at low temperatures and consider the interaction with CDWs of individual sparsely spaced impurities.

Let us assume that an impurity interacts only with the chain on which it is situated. The interaction of a specified chain with the other chains will be taken into account in the mean field approximation. Quantitatively this approximation is justified by the fact the cell of the superstructure in the transverse direction is large (2×8 for TaS_3), which suggests that the interaction between chains is long-range. Thus, the problem is reduced to a one-dimensional one with the energy[8–10]

$$H=\frac{w}{8}\int dx\,(1-\cos\varphi+\varphi'^2/2)$$

$$+\sum_i V_i\{1-\cos[Qx_i+\varphi(x_i)]\}, \qquad (1)$$

where w is the energy of the interaction between chains, which is of the order of the transition temperature T_c, and V_i is the energy of the CDW–impurity interaction. For a weak interaction ($V<w$), V is the Fourier transform of the interaction potential. For a strong interaction, the impurity leads to CDW breakoff at the chain. The tunneling of CDWs through the impurity occurs in this case via virtual disintegration of CDWs into individual electrons[11] and their tunneling through the impurity or neighboring chains. The constant V provides a phenomenological description of these processes. When V is large, the impurity fixes the phase and $\varphi(x_i)$ is close to Qx_i.

An equation for the phase is obtained by minimizing H:

$$\frac{\delta H}{\delta\varphi}=\frac{w}{8}(-\varphi''+\sin\varphi)+\sum_i V_i\delta(x-x_i)\sin(Qx_i+\varphi)=0. \qquad (2)$$

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Without impurities the solution to this equation has the form of a soliton:

$$\tan \varphi/4=\exp[\pm(x-x_0)]. \quad (3)$$

Here, in one direction from the soliton the phase on one string differs by 2π from that on the other strings.[10] The energy of such a 2π-soliton is w, and the charge is $\pm 2e$.

Sparsely spaced impurities, the distances between which are much greater than the soliton size, can be considered independently. For one impurity a soliton solution (3) with energy w also exists, but now the soliton is pinned by the impurity and x_0 is not arbitrary but is found from the condition $\varphi(x_i) = Qx_i$.

Without impurities there is a state $\varphi \equiv 0$ with a zero net charge that energetically is more advantageous than a soliton. The solution $\varphi \equiv 0$ does not hold near an impurity; instead there exists a solution with a zero net charge but with a dipole moment:

$$\tan \varphi(x)/4=\tan(\psi/4)\exp(-|x-x_i|), \quad (4)$$

where $\psi = \varphi(x_i)$ is found from the matching condition

$$\pm(w/2)\sin(\psi/2)+V\sin(Qx_i+\psi)=0. \quad (5)$$

Solution (4) is a linear combination of two solitons of opposite sign with centers at the points $x = x_i \pm \ln \tan \psi/4$.

The energy of such a dipole soliton is

$$E_i=w[1\mp\cos(\psi/2)]+V[1-\cos(Qx_i+\psi)], \quad (6)$$

which is smaller than the energy w of a charged soliton. Equation (5) for ψ follows from the condition $\partial E/\partial\psi=0$.

3. THE CONDITION FOR STRONG PINNING

When the interaction is weak ($V \ll w$), an impurity perturbs CDWs only weakly, ψ is small (or close to $2\pi n$), and the interaction energy is

$$E=V(1-\cos Qx_i). \quad (7)$$

The force with which the impurity acts on CDWs is $\partial E/\partial x_i$. Averaging this force over the random distribution of impurities or over time yields zero. A finite value emerges only due to collective effects that are quadratically or exponentially small in the impurity concentration. An effect linear in the impurity concentration emerges only when the energy (6) is not a one-valued function of x_i, that is, Eq. (5) has two intersecting solutions. It can be verified that this occurs when $V > w/4$. For instance, when $V \gg w/4$ holds the impurity fixes the phase, $\psi = -Qx_i$, and the energy is

$$E_i=w\left[1\mp\cos\frac{Qx_i}{2}\right]-\frac{w^2}{8V}\sin^2\frac{Qx_i}{2}. \quad (8)$$

If we ignore the exponentially weak quantum-tunneling processes, the two branches intersect at $Qx_i = \pi$, and the energy depends on the history. For instance, when the wave propagates to the left, the position of the impurity relative to the wave, $x_i(t)$, increases as a function of time, and in Eq. (8) we must select the upper sign. When Qx_i approaches 2π, the two solitons of opposite sign move away from the impurity, and the energy approaches $2w$. The impurity–wave interaction force does not change sign and is given by the following expression:

$$F\equiv\frac{\partial E}{\partial x_i}=\frac{wQ}{2}\sin\frac{Qx_i}{2}. \quad (9)$$

After averaging over the period of x_i, we get the average force:

$$\bar{F}_i=\frac{wQ}{\pi}. \quad (10)$$

When the wave is displaced by $2\pi/Q$ this force performs work in order to create two outgoing solitons. The solitons never return, and further displacement of the wave by one period creates a new pair of solitons.

The same average force emerges for all impurities with energy $V > w/2$. As noted earlier, impurities with $V < w/4$ do not lead to the formation of metastable states and contribute nothing to the average force. Impurities satisfying $w/2 > V > w/4$ do not fix the phase and do not lead to the formation of solitons but do form metastable states with a zero net charge. The work spent on their formation is finite but smaller than w, with the result that the average force is smaller than the value given by Eq. (10).

The upper critical electric field is

$$eE_t=wn, \quad (11)$$

where n is the line density of strong impurities. For fields higher than E_t the CDWs move coherently and a periodic signal (narrow-band noise) is generated due to soliton creation.

4. CHARGE-DENSITY WAVE

Above we ignored thermal and quantum fluctuations. Allowing for such fluctuations causes the CDW to "creep" through the impurities at a field strength below the critical. The probability of quantum tunneling of CDWs through an impurity was found in Ref. 9 via the theory of instantons (or macroscopic quantum tunneling) and is given by the following relation:

$$\tau_0^{-1}\simeq\exp\left\{-2\left(\frac{m^*}{m}\right)^{1/2}\left(\ln\frac{V}{w}+C\right)\right\}, \quad (12)$$

where m^* is the effective soliton mass ($m^* \gg m$).

Impurities of large transverse size simultaneously pin many chains, that is, one or several superstructure cells. Such pinning has apparently been observed by the electron diffraction method.[12] For such multichain tunneling the effective mass m^* in Eq. (12) must be increased. Because of the large CDW mass (m^*) this probability is exponentially low, but it is important in the event of slow CDW motion. Quantum tunneling leads to repulsion of the branches of the spectrum, with the transitions between these branches described by the Landau–Zener theory. If the velocity of CDW motion is high, so that the time it takes the wave to travel one wavelength past an impurity is

short compared to τ_0, there is no quantum transition between the branches, so that it is proper to apply the classical approach to the problem. But if the velocity is low, then to within exponential accuracy the system remains on the lower level and these impurities provide nothing to the average force.

Thermal fluctuations play a similar role. If thermal equilibrium has time to establish itself while the CDW travels through the impurity, the average force is proportional to the lifetime of the metastable state and is weak. The time it takes thermal equilibrium to set in is determined by the activation energy, that is, the saddle point of Hamiltonian (1), or the maximum of energy (6). This maximum corresponds to the third solution of Eq. (5). For $V \gg w$ the solution is $\psi = -Qx_i + \pi$, and the energy is close to $2V$. Hence, the time for thermal equilibrium to set in is exponentially long:

$$\tau_T = \exp \frac{2V}{T}. \tag{13}$$

Here, as in Eq. (12), we have not written the pre- exponential factors, which are model-dependent.

Depending on the temperature and the interaction energy V, either thermal relaxation or quantum relaxation is important. Hence, it is logical not to solve the problem of macroscopic quantum tunneling but write an interpolation formula:

$$\tau^{-1} = \tau_0^{-1} + \tau_T^{-1}. \tag{14}$$

Creep processes give rise to a nonlinear current–voltage characteristic below the critical field.[13]

5. SPECIFIC HEAT

As noted earlier, impurities at points where Qx_i is close to π lead to states with close energies. These states provide the main contribution to low-temperature specific heat.[3] If the temperature is not too low ($T\tau_0 \gg 1$), quantum repulsion of levels can be ignored and the specific heat is given by the following formula:

$$C = \pi^2 n \nu T/3, \tag{15}$$

where n is the impurity density, and ν the density of states with equal energy. Equation (6) yields

$$\nu = \int \delta(E_1 - E_2) \frac{Q dx_i}{2\pi} P(V)\, dV$$

$$= \int_{w/4}^{\infty} \frac{P(V) dV}{16\pi} \left(1 - \frac{w^2}{16V^2}\right)^{-1/2}, \tag{16}$$

where $P(V)$ is the function determining the distribution of impurities with respect to their interaction energy. If this function falls off rapidly with increasing V, the temperature dependence may depart from linearity because impurities with a threshold energy $V = w/4$ yield a specific heat proportional to $T^{1/2}$. Above we assumed that the temperatures are not too low, and that in the course of the experiment there is time for the essential impurities with energies $V \approx w$ to establish a state of thermal equilibrium.

6. DIELECTRIC CONSTANT

The displacement φ of CDWs caused by a weak electric field E can be found from the equilibrium condition

$$eE = \sum_{i\alpha} f_{i\alpha}\, \delta E_{i\alpha}(Qx_i - \varphi)/\delta x_i, \tag{17}$$

where the energy of the two branches, $E_{i\alpha}$ ($\alpha = +, -$), is determined by Eq. (8), and the distribution functions $f_{\pm}$ satisfy the equation

$$\frac{\delta f_+}{\delta t} = \frac{1}{\tau_T}\left(f_+ \exp\left(\frac{E_+}{T}\right) - f_- \exp\left(\frac{E_-}{T}\right)\right). \tag{18}$$

Solving the system of equations (17) and (18) to first order in the displacement φ of the wave, we arrive at the following expression for the dielectric constant $\varepsilon = e\varphi S/E$:

$$\varepsilon^{-1} = \frac{nwS}{e^2} \int P(V) \frac{dV}{1 + (i\omega\tau_T)^{-1}}, \tag{19}$$

where S is the transverse size of a unit cell. Because of the exponential dependence of τ_T on V specified by Eq. (13), at low temperatures only impurities whose energy V is much higher than $T \ln \omega^{-1}$ contribute to ε. As the temperature rises or the frequency falls, the number of such impurities rapidly decreases and ε grows.

This temperature and frequency dependence of ε has been observed in experiments.[4,14,15] As Eq. (19) implies, in the low-temperature region ε depends only on the product $T \ln \omega^{-1}$. For instance, for an exponentially decreasing distribution function $P(V) \approx \exp(-V/V_0)$ we have

$$\varepsilon \approx \exp(+T \ln(\omega/\omega_0)/V_0).$$

As the temperature grows still further, the effects of collective pinning become important and lead to a decrease in ε as T increases.[16]

7. HOPPING CONDUCTIVITY

Below the Peierls transition temperature, a CDW is a glass. As in any glass, there are arbitrarily high barriers to overcome in order to reach a state of thermal equilibrium. A CDW in a weak field is pinned and there is no collective motion. Singular charges can serve as electric current carriers, and at high temperatures ($\approx T_c$) these charges are electrons. These excitations have a large energy gap Δ_p, and as the temperature is reduced the role of carrier switches to topological excitations, solitons with a lower energy w, which can be considered to be a dislocation loop with a small radius.[17]. At intermediate temperatures the conductivity is proportional to the number of free electrons[5]:

$$\sigma \approx \tau_0^{-1} \exp\left(-\frac{w}{T}\right). \tag{20}$$

The smallness of the pre-exponential factor proportional to τ_0^{-1} (see Eq. (12)) is due to the low probability for solitons to pass through the impurity.[9] Hence at $T \approx T_c$ the soliton contribution to the conductivity is lower than that

of electrons, although the number of solitons is great. Only at temperatures noticeably below T_c does soliton conductivity take over. As the temperature drops still further, the number of free solitons decreases exponentially and hopping conductivity sets in. The solitons pinned to impurities do the hopping. Coulomb interaction between solitons results in the following expression for hopping conductivity:

$$\sigma \approx \exp[-(T_0/T)^{1/2}]. \quad (21)$$

The pre-exponential factor in this expression is proportional to the number of pinned solitons.

The energy w of such a soliton is higher than that of state (8) with zero net charge. But for V large and Qx_i close to π the difference in the energy of such states is small, with the result that the density of such pinned solitons decreases with temperature only algebraically.

Commensurability effects, so far not taken into account, lead to a situation in which the number of pinned charged solitons remains finite even at absolute zero. These effects make the chemical potential of charged solitons finite, although lower than w. The energy of charged solitons is $w \pm \mu$, and for solitons of the same sign can be lower than the energy of a dipole soliton (8). These charged solitons provide the principal contribution to the hopping conductivity.

8. CONCLUSION

We have examined a model of strong CDW pinning. In it impurities distort the wave only on the chains containing the impurities, while interaction with neighboring chains is taken into account in the mean-field approximation. Quantitatively the model provides a condition for strong pinning: the wave–impurity interaction energy must exceed a quarter of the energy of a 2π-soliton.

Deformation of neighboring chains may change the quantitative relations but has no effect on the quantitative picture. Here a strong impurity fixes a local value of the CDW phase. A charged soliton may appear on the impurity, but the formation of a dipole soliton, in which charges with different signs appear on opposite sides of the impurity, is energetically more advantageous. Two states that differ in the sign of the dipole moment generally have different energies. One is metastable. The existence of such states makes it possible to explain qualitatively many low-temperature properties of systems with CDWs, say, the increase in ε when the temperature rises and when the frequency decreases.

The decrease in ε brought on by a further increase in temperature[4,14,15] is related in this picture to the change in the nature of pinning.[16] At high temperatures the pinning is collective; each impurity gives rise to a weak elastic deformation of CDWs, short-range order exists in large regions containing many impurities, the average force is weak, and the barrier energy is high. At low temperatures strong single-center pinning, where sparsely spaced strong pinning centers (chain breakoffs) give rise to plastic deformations in CDWs, is more important. In this picture a CDW is assumed to be in a glassy state everywhere below the three-dimensional transition temperature T_c. This assumption is supported both by the theoretical reasoning that random pinning transforms any structure into a glassy state and by the experimental evidence about the existence of a hysteresis loop at high temperatures.[5] The picture does not require the hypothesis that there is an additional low-temperature glassy phase transition.[3,4]

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