

Irreversible phonon-free energy transport in disordered Coulomb systems

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(Submitted 26 May 1994)

Zh. Eksp. Teor. Fiz. **106**, 1169–1184 (October 1994)

A disordered Coulomb system in a random external potential is used as an example for an analysis of relaxation processes which occur in an ensemble of two-level systems with a dipole-dipole interaction. Phonon-free relaxation processes can occur only at temperatures $T > 0$. They are associated with the existence of a special class of elementary excitations: resonant bidipoles. At low temperatures the relaxation rate of these excitations is a linear function of the temperature, and the thermal conductivity has a $\kappa \sim T^{4/3}$ behavior. At ultralow temperatures, this behavior may outweigh the mechanism in which heat is transferred by phonons scattered by two-level systems, with $\kappa \sim T^2$. © 1994 American Institute of Physics.

1. INTRODUCTION

The primary relaxation mechanism in disordered semiconductors in which the mobility threshold is below the bottom of the conduction band—the mechanism responsible for the electrical conductivity—is the electron-phonon interaction. The phonons stimulate hops of electrons between different localized states, leading to a Mott law¹ $\sigma \propto \exp[-(T_0/T)^{1/4}]$. When the Coulomb gap is taken into account, the result is a Shklovskii–Éfros law² $\sigma \propto \exp[-(T_1/T)^{1/2}]$.

It has been shown³ that an irreversible motion occurs in a subsystem of localized electrons which is isolated (from phonons), because of an inelastic interaction. At low temperatures, this motion leads to a power-law dependence $\sigma \propto T^2$. A problem of independent interest is irreversible phonon-free energy transport in such a system (in the absence of a particle transport). The latter was first taken up in this formulation in Ref. 4, for the case of an ensemble of two-level systems interacting by an $R^{-\alpha}$ law with $\alpha > 3$. It was shown there that an irreversible energy transport occurs only at temperatures $T > 0$. It occurs by virtue of multiparticle (multiquantum) transitions, when a change in the state of the surrounding two-level systems leads to fluctuations in the transition energy at a resonant pair of two-level systems of interest. The results of Ref. 4 cannot be generalized directly to the $\alpha = 3$ case, since in that case we cannot draw a conclusion regarding the existence of an infinite resonant cluster. In the present paper we consider the case of an ensemble of two-level centers with an R^{-3} ($\alpha = 3$) interaction. As is shown in Sec. 2 of this paper, this is the case to which the model of an electrically neutral system of interacting electrons and holes in a random potential reduces at low temperatures.⁵ In Sec. 3 we describe a fluctuation mechanism for the onset of an irreversible motion, and we construct a self-consistent equation which describes this process. The R^{-3} interaction gives rise to an irreversible motion in a subsystem of resonant dipoles; this motion exists even at $T = 0$ in the case of a soft Coulomb gap (Sec. 4). In the case of hard Coulomb gap, a motion of this type arises only at a nonzero temperature, because of the existence of a special class of excitations: resonant bidipoles (Sec. 5). We call attention to

the important role played by excitations of this type in the propagation of sound in such systems, first studied theoretically in Ref. 6. In Sec. 6 we show that the energy-transport mechanism discussed in Sec. 5 leads to a phonon-free thermal conductivity. At sufficiently low temperatures, the thermal conductivity has a temperature dependence $\kappa \sim T^{4/3}$. We conclude the paper with a discussion of the results of the study, and we offer some qualitative considerations which indicate the existence of a hard Coulomb gap in a spectrum of charge excitations of a disordered Coulomb system. We set Planck's constant equal to unity: $\hbar = 1$.

2. RESONANT CLUSTERS IN DISORDERED COULOMB SYSTEMS

We consider the problem of a system of electrons in a random potential.⁵ We assume that the mobility threshold is well above the Fermi level μ . The localization centers of one-particle states, with a characteristic localization radius l , are half-filled and are distributed randomly, separated from each other by an average distance $a = 1$. We assume $l < a$.

To each localization center i of this lattice we assign a random potential ϕ_i (reckoned from the Fermi level), with a distribution function

$$g(\phi_1) = g_0 \theta((2g_0)^{-1} - |\phi_1|). \quad (1)$$

The energy of the multielectron system is thus the sum of the electron energy in potential (1) and the Coulomb interaction (V) between electrons. Here

$$V = U_0 \int \Psi^+(\mathbf{r}_1) \Psi^+(\mathbf{r}_2) |\mathbf{r}_1 - \mathbf{r}_2|^{-1} \Psi(\mathbf{r}_2) \Psi(\mathbf{r}_1) d\mathbf{r}_1 d\mathbf{r}_2, \quad (2)$$

where the operators $\Psi^+(\mathbf{r})$, and $\Psi(\mathbf{r})$ create and annihilate an electron at the point \mathbf{r} . the intrasite repulsion is assumed to be strong enough that no more than one electron can occupy a site. Under this assumption, we can restrict the analysis below to a model without spin. We assume

$$\gamma = g_0 U_0 \ll 1. \quad (3)$$

As the one-particle basis functions of the problem we initially choose the one-particle (one-site) localized eigenstates of an electron in potential (1). The complete Hamiltonian of the problem can then be written

$$H = H_0 + V, \quad (4)$$

$$H_0 = \sum_i \phi_i(n_i - 1/2) + 1/2 \sum_{i,j} U_{i,j}(n_i - 1/2)(n_j - 1/2),$$

$$V = V_1 + V_2,$$

$$V_1 = \sum_{i,j} U_{ij}^j c_i^+ c_j, \quad U_i^j = \sum_k U_{ik}^j(n_k - 1/2),$$

$$V_2 = \sum_{i,j,k,l} U_{kl}^{ij} c_i^+ c_j^+ c_k c_l.$$

Here the operators c_i^+ and c_i create and annihilate an electron in a localized state at site i , and we have $n_i = c_i^+ c_i$. Combinations of the type $(n_i - 1/2)$ in expression (4) explicitly ensure electrical neutrality of the system. In the sums in the expressions for V_1 and V_2 , the subscripts in a pair always have different values. The matrix element $U_{ij} \sim U_0/r_{ij}$ describes the Coulomb interaction between electrons in localized states. The quantity U_{ik}^j is the seed amplitude for the transition of an electron from site i to site j in the field of an electron at site k . An important role is played in this problem by the matrix elements for the simultaneous transition of two electrons from sites i, j to sites k, l ,

$$U_{kl}^{ij} = U_0 \int \Psi_k^*(\mathbf{r}_1) \Psi_l^*(\mathbf{r}_2) |\mathbf{r}_1 - \mathbf{r}_2|^{-1} \Psi_i(\mathbf{r}_1) \Psi_j(\mathbf{r}_2) d\mathbf{r}_1 d\mathbf{r}_2, \quad (5)$$

in the case in which the distances between the localization centers satisfy $r_{ik}, r_{jl} \ll r_{kl}, r_{ij}$. (Here $\Psi_k(\mathbf{r})$ are the wave functions of the base states.) We place the origins of coordinates for \mathbf{r}_1 and \mathbf{r}_2 at the points k and l , respectively. With an eye on an order-of-magnitude estimate, we ignore the angular dependence of the matrix elements, and we replace $|\mathbf{r}_1 - \mathbf{r}_2|^{-1}$ in (5) by the expansion

$$r_{kl}^{-1} + r_{kl}^{-2}(r_1 + r_2) + r_{kl}^{-3}(r_1^2 + r_2^2 + r_1 r_2) + \dots$$

By virtue of the orthogonality of the selected basis states, the first nonvanishing contribution to expression (5) comes from the third term of this expansion. As a result we find

$$U_{kl}^{ij} \sim U_0/r_{kl}^3 \exp(-(r_{ik} + r_{jl})/l). \quad (6)$$

Correspondingly, we can show that we have

$$U_i^k = U_0 \sum_l \exp(-r_{ik}/l) n_l / r_{kl}^2 \approx U_0 \exp(-r_{ik}/l). \quad (7)$$

Following Ref. 5, we recall a classification of elementary excitations in the system described by the Hamiltonian H_0 . The basis functions selected above are also eigenfunctions of the Hamiltonian H_0 . The simplest excitation, a *charge* excitation, arises in the transfer of an electron from some site i to infinity; it has an energy $-\varepsilon_i$, where

$$\varepsilon_i = \delta H / \delta n_i = \phi_i + \sum_j U_{ij}(n_j - 1/2). \quad (8)$$

As an electron goes from infinity to some vacant site i , the energy of the system increases by ε_i . Since the ε_i 's are measured from the Fermi level, all the sites with $\varepsilon_i < 0$ are filled in the ground state, while the others, with $\varepsilon_i > 0$, are vacant.

The transport of an electron over a finite distance r_{ij} leads to a *dipole* excitation of the system, with an energy

$$\omega_{ij}^j = \varepsilon_j - \varepsilon_i - U_0/r_{ij} > 0, \quad \varepsilon_j > 0, \quad \varepsilon_i < 0. \quad (9)$$

Such a pair of sites is a "dipole." If the electron is at site j , then the dipole is by definition in an excited state with an excitation energy ω_{ij}^j ; the dipole is instead in the ground state if the electron is at site i .

To a large extent, the low-temperature kinetic and thermal properties of this model are governed by low-energy charge and dipole excitations.

The seed density of charge excitations in (1) is constant and is governed entirely by the random potential ϕ . In a first approximation in the Coulomb interaction, the density of states of the charge excitations of the system, $g(\varepsilon)$, is found from the requirement that the system be stable with respect to dipole excitations, i.e., from the condition that the quantity ω_{ij}^j be positive. This density of states is

$$g(\varepsilon) = g_0 \begin{cases} (\varepsilon/\Delta)^2, & |\varepsilon| < \Delta, \\ 1, & |\varepsilon| > \Delta, \end{cases} \quad \Delta = U_0 \gamma^{1/2}, \quad (10)$$

where $\Delta = U_0 \gamma^{1/2}$ is the "soft Coulomb gap."

In the next approximation, the distribution function of the charge excitations is found from the condition that the system be stable with respect to the *simultaneous* excitation of two dipoles, i.e., from the condition that the quantity ω_{kl}^{ij} be positive:

$$\omega_{kl}^{ij} = \omega_k^i + \omega_l^j - E_d > 0 \quad (11)$$

$$E_d = U_{kl} + U_{ij} - U_{ki} - U_{jl} \approx U_0 r_{ik} r_{jl} / r_{ij}^3, \quad r_{ik}, r_{jl} \ll r_{ij}^3.$$

Condition (11) leads to a structural feature in the density of states of the charge excitations, i.e., to the formation of a *hard* Coulomb gap in the density of states. We thus have

$$g(\varepsilon) \propto \exp[-\Delta(|\varepsilon| \ln^{7/4}(\Delta/|\varepsilon|))^{-1}]. \quad (12)$$

Note, however, that while the existence of a soft Coulomb gap in the density of states has been repeatedly confirmed experimentally in measurements of the temperature dependence of the hopping conductivity, the existence of a hard Coulomb gap remains an open question.

In the present paper we are interested in the temperature region $T \ll \Delta$. For the problem solved below, only excitations with energy

$$\omega \leq T \ll \Delta \quad (13)$$

can be important. Accordingly, charge excitations with the density of states (12) are exponentially suppressed, and we will consider them no further.

The density of states of dipole excitations in the space of energies and dimensions is given by

$$F(\nu) = \int_0^\infty g(\varepsilon_1) \int_{-\infty}^0 g(\varepsilon_2) \delta(\varepsilon_1 - \varepsilon_2 - U_0/r - \omega) d\varepsilon_1 d\varepsilon_2, \quad (14)$$

where $\nu = (\omega, r)$. In the case of a soft Coulomb gap, at energies which satisfy condition (13), we have

$$F(\nu) = F_0(\nu) \sim \theta(\omega) g_0^2(U_0/r + \omega) \times \begin{cases} 1, & r < r_0 \\ [(U_0/r + \omega)/\Delta]^4, & r > r_0 \end{cases}, \quad (15)$$

where $r_0 = \gamma^{-1/2} \gg 1$. It follows that the density of states of dipoles with a small excitation energy falls off rapidly with increasing size of the dipole under the condition $r_\nu > r_0$. In the case of a hard Coulomb gap we have

$$F(\nu) = F_0(\nu) \exp(-G(\omega, r)), \quad G(\omega, r) \approx r/r_0 \ln^{1/4}((\Delta + \omega)/\omega). \quad (16)$$

Low-energy dipole excitations which satisfy condition (13) can be constructed at sites with large values $\varepsilon \gg \Delta$, which do not lie in the Coulomb gap; the excitation energy in (9) is kept small by means of the offsetting term U_0/r . A decrease in the density of states of dipole excitations as the result of a hard Coulomb gap, on the other hand, occurs to a far lesser extent.

A dipole (a pair of sites with $\varepsilon_i < 0$ and $\varepsilon_j > 0$) thus essentially forms a two-level system with a distribution function

$$F(\nu, T) = F(|\omega|, r) f(\omega), \quad \omega = \pm \omega_i^j, \quad (17)$$

where $f(\omega) = [1 + \exp(\omega\beta)]^{-1}$. The minus sign on ω in (17) corresponds to the case in which the dipole is in the ground state (the electron is at site i), while the plus sign corresponds to the case in which the dipole is in the excited state (the electron is at site j). In addition, if the system is not in thermodynamic equilibrium, then the function $f(\omega)$ in (17) may depend on the coordinate of the dipole. As a result, the concentration of excited dipoles may be thought of as a measure of the local temperature.

Up to this point we have actually been discussing excitations in a system described by the Hamiltonian H_0 . The hybridization of excitations due to the interaction V [see (4)] evidently leads to a tendency toward localization. This tendency is most noticeable in the case of resonant excitations,^{3,4,6,7,8} for which the matrix element for a transition between two states reaches a value on the order of the transition energy. Excitations of specifically this sort are responsible for delocalization and transport in the system. The interaction V_1 [see (4)] gives rise to hybridization of the one-site basis functions (the eigenfunctions of Hamiltonian H_0) and to a trivial renormalization of the energy of the charge excitations, (8), which can be ignored. If this hybridization is taken into account, it leads to a renormalization of the energies of the dipole excitations. This renormalization is important if the dipole is resonant, i.e., if its excitation energy, (9), is smaller than transition amplitude (7), so we have

$$|\omega_i^j| \leq |U_i^j|. \quad (18)$$

In this case we have the following expression for the renormalized excitation energy:

$$\tilde{\omega}_i^j = \sqrt{(\omega_i^j)^2 + (U_i^j)^2}. \quad (19)$$

The two-particle amplitude for transition (6) is simultaneously renormalized, so we have

$$\tilde{U}_{it}^{jk} \sim U_{it}^{jk} + \frac{U_i^j U_t^k}{\tilde{\omega}_i^j \tilde{\omega}_t^k} E_d \quad (20)$$

[see (11) regarding the meaning of E_d]. The derivation of the estimate (20) is precisely equivalent to the derivation of (6) and (7). In addition, we have made use of the orthogonality of the new hybridized wave functions (which do not have an exponentially small overlap integral). The two terms in (20) are generally of the same order of magnitude. However, the resonant dipoles will play an important role below. For a pair of such resonant dipoles the first term in expression (20) (which is proportional to the tunneling amplitude) can be discarded, since under condition (18) the second term is actually dominant. This second term does not contain exponentially small tunneling amplitudes.

Denoting by U_ν the transition amplitude (7) for a dipole of size r_ν , and integrating expression (15) over the frequency ω within the limits set by the resonance condition (18), we find an estimate of the size distribution of the resonant dipoles:

$$P_\nu \sim F(\nu, T) U_\nu. \quad (21)$$

Using (16) and (18), we can easily show that the hard-Coulomb-gap factor for these dipoles can be used in the form

$$G_\nu = (r_\nu/r_d)^{5/4}, \quad (22)$$

where the parameter $r_d = (r_0^4 l)^{1/5}$ can be interpreted as the maximum size of a resonant dipole.

Energy transport may be associated with a transition in pairs of dipole excitations if one of the dipoles is initially in the ground state, and the other in the excited state.^{3,4,8} Analysis of various possibilities for energy transport by this mechanism shows that it is necessary to consider pairs of dipoles, i.e., *bidipoles*. We wish to stress that these bidipoles are themselves elementary entities (two-level systems of a sort) for the kinetics of excitations in highly disordered systems.^{3,4,8}

We assume that for some dipole pair a and b , with parameters ν_a and ν_b , respectively, the energy of their interaction, E_d [see (11)], is small in comparison with the excitation energy (9) for each. We thus have

$$|\omega_a|, |\omega_b| \gg E_d. \quad (23)$$

In this case we can introduce a distribution function for a bidipole, $F(\nu, T)$, i.e., the joint distribution function of the two dipoles under consideration:

$$F(\nu, T) = F(\nu_a, T) F(\nu_b, T). \quad (24)$$

Here ν specifies a complete set of parameters which characterize the bidipole. In particular, it includes ν_a , ν_b , and the distance between these dipoles, R . [Wherever we can do so without causing any misunderstanding, we will use the same notation, $F(\nu, T)$ and U_ν , for the distribution function and

the transition amplitude. We can then speak in terms of the parameters of a resonant cluster, i.e., a resonant dipole or bidipole.] In general, each such bidipole can be in four states, and it is characterized by a spectrum of excitations Ω in addition to transition amplitude (20). We introduce the distribution function of a bidipole with a given excitation energy Ω . We can then write

$$F_\nu(\Omega, T) = F(\nu_a, T)F(\nu_b, T)F(\nu_b, T)\delta(\omega_a + \omega_b - E_d - \Omega). \quad (25)$$

By virtue of condition (23), we can omit the parameter E_d from the argument of the δ -function. For such bidipoles, the distribution function is obviously independent of the relative spatial orientation of the constituent dipoles. Furthermore, a bidipole is by definition resonant if the transition amplitude U_ν for it, given by (20), is such that the condition

$$|\Omega| < U_\nu \quad (26)$$

holds. The parameter U_ν is at most no greater than E_d [see (20)]. Accordingly, to the same accuracy we can discard the term Ω from the argument of the δ -function in (25). This means that dipole distribution function (25) depends only weakly (not at all, in the approximation under consideration here) on both the distance between dipoles and the excitation energy Ω . Carrying out the trivial integration over Ω in (25) within the limits specified in (26), we find an estimate of the distribution function of resonant bidipoles with a given transition amplitude U_ν :

$$P_\nu(T) = F(\nu_a, T)F(\nu_b, T)\delta(\omega_a + \omega_b)U_\nu, \quad U_\nu \sim U_0 \frac{r_a r_b}{R_\nu}. \quad (27)$$

We wish to stress that in this expression and also everywhere below we have $\nu = (\nu_a, \nu_b, R_\nu)$, where R_ν —the size of the bidipole—is the distance between dipoles a and b . It follows from (27) that resonant dipoles actually form a two-level system and are made up of dipoles with approximately equal transition energies (these energies differ by an amount no greater than U_ν), but one is in the ground state and the other in the excited state. The transition energy in such a bidipole is found to be of order U_ν . The transition itself is the exchange of an excitation between the dipoles.

In a resonant dipole, an electron is simultaneously at both sites (although with different probabilities). In a resonant bidipole, on the other hand, the electrons are smeared over the sites of the corresponding dipoles, even if neither of the dipoles is resonant.

3. DERIVATION OF A SELF-CONSISTENT EQUATION FOR THE RELAXATION RATE

We adopt the hypothesis that *irreversible motion* of electrons occurs between the sites—the localization centers—in the system, in accordance with the following scenario. An electron is initially localized at some *single* site. We assume that a fluctuation has arisen for random reasons of some sort (they will be clarified below). As a result, the energy at this site and that at some other site, close by and vacant, change in such a way that the sites form a resonant dipole. After a certain time, for the same random reasons, this pair of sites

ceases to be resonant, and the electron can, with a finite probability, become localized at the second site. The resonant dipole disappears in the process. Each such hop of an electron is accompanied by a change in the dipole moment and, correspondingly, in the field of the dipole under consideration. This is the random factor which causes the energy fluctuation at surrounding sites and leads to the creation and disappearance of resonant dipoles.

We turn now to the description of a possible scenario for the evolution of the transition energy at some pair of sites. We assume that this pair of sites is initially not in resonance. At a later time, as the result of a fluctuation, the energy of the site which corresponded to the excited state of the dipole at the initial time begins to decrease, while the energy of the second site (which originally corresponded to the ground state) begins to increase. (We are actually interested in the mismatch of the levels, i.e., the difference between these energies.) In the course of such a “motion” of the energy levels, this difference can then ultimately reach the resonant value, equal to the transition amplitude U_ν . There is a finite probability (of order 1/2) that this tendency in the direction of the motion of the levels will persist. We adopt this instant of time as the initial time for a study of the subsequent evolution of the mismatch in the energy of the dipole. After a certain time t_ν , this pair of sites ceases to be resonant. The mismatch in the energy levels, changing sign, becomes greater than U_ν in absolute value. As a result, the positions of the ground and excited states trade places spatially. If this process occurs fast enough to satisfy the condition

$$U_\nu t_\nu < 1, \quad (28)$$

then one can show that the probability for the transition of an electron to the other site is small in proportion as this parameter is small. The electron remains in its previous position, although the energy of the transition between sites changes sign. In the opposite case,

$$\Gamma_\nu \sim t_\nu^{-1} \leq U_\nu, \quad (29)$$

of a slow (adiabatic) fluctuation, the probability for a transition to the other (previously vacant) site is unity (to within exponential accuracy). Such a transition is accompanied by a fluctuational change, over a time t_ν , in the dipole moment of this pair of sites.

Condition (29) can also be interpreted another way. The very concept of a resonant cluster ν is meaningful only if the quantum-mechanical uncertainty in the energy, which stems from the finite lifetime of a resonant cluster, $t_\nu \sim \Gamma_\nu^{-1}$, is smaller than the transition energy of this cluster, which is on the order of the transition amplitude.

Resonant dipoles play a major role in charge transport.³ In energy transport, the onset and disappearance of resonant bidipoles also play an important role (ultimately, a dominant role). A fluctuation of the transition energy in a bidipole occurs as the result of a fluctuation in the energy at one of the dipoles. If the difference between the excitation energies of the dipoles reaches the transition amplitude of the bidipole, the latter becomes resonant.

We wish to stress that, until the dipole or bidipole becomes resonant, its interaction with the surroundings does

not change. Only during the time interval t_ν while the cluster is in resonance do its dipole moment and thus the interaction with the other dipoles and bidipoles change. It is for this reason that nonresonant clusters are not directly pertinent to energy transport. In the discussion below we will thus consider only resonant dipoles, and then only those resonant dipoles which satisfy condition (29).

We proceed now to the derivation of a self-consistent equation for determining the reciprocal lifetime Γ_ν of a resonant cluster (dipole or bidipole). This time is equal to the probability for the decay of the cluster per unit time (the subscript ν represents the complete set of parameters of the resonant cluster). A resonant cluster disappears or appears if an electron in another resonant cluster, ν' , at some distance $R_{\nu\nu'}$ from the first, undergoes a hop (if the cluster ν' decays). The decay of the resonant cluster ν' causes a fluctuation of the transition energy at cluster ν :

$$\Delta E_{\nu\nu'} \sim U_0 r_\nu r_{\nu'} / R_{\nu\nu'}^3. \quad (30)$$

Here r_ν and $r_{\nu'}$ are the sizes of the dipoles of clusters ν and ν' . Cluster ν ceases to be resonant if the energy fluctuation $\Delta E_{\nu\nu'}$ exceeds the transition amplitude of this cluster:

$$\Delta E_{\nu\nu'} > U_\nu. \quad (31)$$

Strictly speaking, a fluctuation of transition energy is accompanied by a fluctuation of the transition amplitude. However, it is easy to show, with the help of (6), (7), and (20), that the fluctuation of the latter can be ignored.

The probability Γ_ν for the decay of the resonant cluster ν per unit time is numerically equal to the total number of decays per unit time of the surrounding resonant clusters ν' which satisfy conditions (29) and (31):

$$\Gamma_\nu = \sum_{\nu'} \sum_{R_{\nu\nu'}} \Gamma_{\nu'} P_{\nu'} \theta(\Delta E_{\nu\nu'} - U_\nu) \theta(U_{\nu'} - \Gamma_{\nu'}). \quad (32)$$

Here P_ν is the distribution function of the resonant clusters [see (21) and (27)], and the function θ reflects conditions (29) and (31).

Let us look at some general properties of the solution of Eq. (32), which is a homogeneous nonlinear integral equation. We first note that there exists a trivial solution $\Gamma_\nu \equiv 0$. This circumstance indicates that there is no irreversible motion in the system and that the localization of excitations is complete. We are only interested in a nontrivial solution of Eq. (32) for which the relation $\Gamma_\nu \neq 0$ holds for at least certain values of ν . Let us assume that the condition $\Gamma_\nu \ll U_\nu$ holds for all ν . Since we have $\theta(U_{\nu'} - \Gamma_{\nu'}) \equiv 1$ in this case, Eq. (32) becomes a linear integral equation, which has no solutions other than the trivial one. Consequently, if a nontrivial solution of Eq. (32) exists, then the condition

$$\Gamma_\nu \approx U_\nu \quad (33)$$

must hold for certain values of ν .

4. ABSOLUTE ZERO ($T=0$); ROLE OF RESONANT DIPOLES

Let us assume that the temperature of the system is $T=0$. Since there are no bidipoles, the only mechanism for energy transport is then that of dipole excitations. We first analyze

the case of a soft Coulomb gap. We make use of expression (21) for the density of dipole excitations; that expression is conveniently approximated by

$$P_\nu = \mu' (g_0^2 U_0 / r_\nu) / (1 + \mu'' (r_\nu / r_0)^4), \quad \mu', \mu'' \sim 1. \quad (34)$$

Here we have assumed that the renormalized transition energy of the resonant dipole satisfies $\omega < U_0 \exp(-r/l)$. Substituting (34) into (32), we find

$$\frac{\Gamma_\nu U_\nu}{r_\nu} = \mu' \int \frac{d(r_{\nu'})^4}{r_0^4} [1 + \mu'' (r_{\nu'} / r_0)^4]^{-1} \theta(U_\nu - \Gamma_{\nu'}) \frac{\Gamma_{\nu'} U_{\nu'}}{r_{\nu'}}. \quad (35)$$

The right side of this equation is independent of ν . Consequently, the quantity

$$\psi = \frac{\Gamma_\nu U_\nu}{U_0^2 r_\nu} \quad (36)$$

is an invariant, independent of the type of dipole. We divide both sides of the last equation by this invariant (it is nonzero if there is irreversible motion in the system), and we evaluate the integral which remains. We cut this integral off at the parameter r_* , found from the condition for the vanishing of the argument of the θ function in (35). Using (33), (36), and (7), we find

$$\exp(-r_*/l) = \psi^{1/2} r_*^{1/2}. \quad (37)$$

As a result we find the estimate

$$\ln(1 + (r_*/r_0)^4) \approx 1 \quad (38)$$

from (35). From the latter relation we find

$$r_* = \xi r_0 (\xi - 1). \quad (39)$$

Using (7), (33), (36), and (37), we find the order-of-magnitude result

$$\Gamma_\nu \sim U_0 \exp[(r_\nu - 2r_*)/l]. \quad (40)$$

This relation indicates that irreversible motion may exist in the system, but it contradicts basic principles of thermodynamics. It will be analyzed in detail in the Conclusion of this paper.

We turn now to the existence of a nontrivial solution of Eq. (32) at $T=0$ in the case of a hard Coulomb gap. We first carry out the summation over $R_{\nu\nu'}$ in (32). Using (30), we find

$$\Gamma_\nu = \sum_{\nu'} \bar{R}_{\nu\nu'}^3 \Gamma_{\nu'} P_{\nu'} \theta(U_{\nu'} - \Gamma_{\nu'}), \quad (41)$$

where

$$\bar{R}_{\nu\nu'} = (U_0 r_\nu r_{\nu'} / U_\nu)^{1/3} \quad (42)$$

is essentially the radius of the resonant interaction of the clusters.

To solve Eq. (41), we multiply both sides by

$$P_{\nu'} r_{\nu'} \theta(U_{\nu'} - \Gamma_{\nu'}) \quad (43)$$

and carry out the summation over ν . For the invariant in (36), which does not depend on the parameters of the resonant cluster,

$$\psi = \frac{\Gamma_\nu U_\nu}{U_0^2 r_\nu} = U_0^{-1} \sum_{\nu'} P_{\nu'} \Gamma_{\nu'} r_{\nu'} \theta(U_{\nu'} - \Gamma_{\nu'}), \quad (44)$$

we then find the equation

$$\sum \frac{U_0 r_\nu^2}{A_\nu} P_\nu \theta \left(\frac{U_\nu}{U_0 r_\nu^{1/2}} - \psi^{1/2} \right) = 1. \quad (45)$$

We can find an upper estimate of the contribution of resonant dipoles to sum (45), taking P_ν from (10), (21), and (22). If we omit the θ function in it obviously this estimate can only increase. Finding our estimate then reduces to analyzing the integral

$$\gamma^2 \int_1^\infty dr r^3 \exp(-r/r_0 (\ln r_0 + r/l)^{4/5}). \quad (46)$$

If the parameter $r_1 = l \ln r_0$ is less than or equal to one, then we find $(l/r_1)^{4/5}$ as an upper estimate of this integral. If we instead have $r_1 \gg 1$, we find the estimate $(r_1/r_0)^4 + (l/r_0)^{4/5}$. Since the relation $l < r_0 / \ln r_0$ always holds, the upper estimate of integral (46) is actually $(l/r_0)^{4/5} \ll 1$. Equation (45) thus has no solution in the case of a hard Coulomb gap, with only resonant dipoles being taken into account (because the right side of this equation is always much less than unity). Accordingly, self-consistent equation (32) has no solutions other than the trivial solution $\Gamma \equiv 0$, and it indicates that there this no irreversible motion in the system.

5. TEMPERATURES $T > 0$; ROLE OF RESONANT BIDIPOLES

We first note that for $T > 0$ (46) is an upper estimate, so in the case of a hard Coulomb gap the conclusion that there is no irreversible motion in a system of dipole excitations—a conclusion reached in the preceding section of this paper—remains in force. As for the case of a soft Coulomb gap, we note that, formally, the dipole transport mechanism, which leads to relation (40), can outweigh the bidipole mechanism only at temperatures

$$T < U_0 \exp(-r_0/l) \quad (47)$$

[see (54) below]. We restrict the discussion below to higher temperatures, at which the dipole contribution can be ignored in comparison with the bidipole contribution.

Let us examine the relaxation which occurs at a temperature $T > 0$ and which is associated with bidipole excitations. Looking back at Eq. (45), we note that the summation in this equation is over all types (ν) of resonant bidipoles. Here r_ν is that size of one of the constituent dipoles at which a fluctuation of the transition energy occurs. Since we are restricting the discussion to the temperatures (47), the characteristic size of the dipoles of interest here, found from the condition $T = U_0 \exp(-r/l)$, is smaller than r_0 . Thus the energies of the charge excitations making up the dipole, $\sim U_0/r$, are large in comparison with the width of the Coulomb gap, U_0/r_0 . We can therefore ignore the effect of the Coulomb gap. At the temperatures of interest, we can also ignore the

anomalous diffusion associated with a one-particle delocalization of excitations by virtue of the behavior of the hopping amplitude, $\propto R^{-3}$ (Ref. 9).

Using (27), we now write Eq. (45) for resonant bidipoles in a different form, switching from summation to integration over the parameters of the resonant bidipoles:

$$\begin{aligned} & U_0 \int_1^\infty r'^4 dr' \int_1^\infty r''^2 dr'' \int_{-\infty}^\infty d\tilde{\omega}' \theta \\ & \times (|\tilde{\omega}'| - U(r')) \int_{-\infty}^\infty d\tilde{\omega}'' \theta \\ & \times (|\tilde{\omega}''| - U(r'')) \int_1^\infty R^2 dR \delta \\ & \times (\tilde{\omega}' + \tilde{\omega}'') F_1(\tilde{\omega}', r', T) F_1(\tilde{\omega}'', r'', T) * \theta \\ & \times \left(\frac{r'^{1/2} r'' U(r') U(r'')}{R^3 |\tilde{\omega}'| |\tilde{\omega}''|} - \psi^{1/2} \right) = 1. \end{aligned} \quad (48)$$

Here r' , r'' , $\tilde{\omega}'$, $\tilde{\omega}''$, $U(r')$, and $U(r'')$ are the dimensions, renormalized energies, and transition amplitudes of the dipoles making up the resonant bidipole of size R .

We first integrate over R and $\tilde{\omega}''$ in (48). As a result we find

$$\begin{aligned} & \psi^{-1/2} U_0^{-1} \gamma^4 \int_1^\infty r'^{7/4} dr' U(r') \\ & \times \int_1^\infty r''^2 dr'' U(r'') \int_{U(r')}^\infty (d\tilde{\omega}' / \tilde{\omega}'^2) f(\tilde{\omega}') f(-\tilde{\omega}'). \end{aligned} \quad (49)$$

For an order-of-magnitude estimate, we replace $f(\tilde{\omega}')$ by 1, and $f(-\tilde{\omega}')$ by $\exp(-\tilde{\omega}'/T)$. We find

$$\begin{aligned} & \int_{U(r')}^\infty \dots d\omega' = U^{-1}(r') \exp(-U(r')/T) + T^{-1} \text{Ei} \\ & \times (-U(r')/T) \sim U^{-1}(r') \exp \\ & \times (-U(r')/T) \{ \theta(T - U(r')) \\ & + T/U(r') \theta(U(r') - T) \}. \end{aligned} \quad (50)$$

It is easy to see that estimate (50) holds at large and small values of $U(r)/T$. We substitute (50) into (49), and we evaluate the remaining integrals separately for the regions $U(r') > U(r'')$ and $U(r') < U(r'')$. The integrals over the two regions are of comparable magnitude. Recalling that we have $T \ll U_0$, we find the following from (48)–(50):

$$\psi^{-1/2} \gamma^4 T / U_0 r_*^{11/2} \sim 1, \quad (51)$$

where

$$r_* = \max\{l, l \ln(U_0/T)\} \quad (52)$$

is a characteristic size of the dipoles. Over the broad temperature range

$$\Delta(r_0/l) \exp(-r_0/l) l \ll T \ll \Delta, \quad (53)$$

the relation $r_* < r_0$ holds. Also using (33), (36), and (51), we find

$$\Gamma_\nu \sim r_\nu U_\nu^{-1} \gamma^8 r_*^{11} l^4 T^2. \quad (54)$$

The existence of a finite lifetime supports the suggestion that there is an irreversible motion. Let us find some characteristic parameters of the problem.

Analyzing the region of parameter values which make the dominant contribution to the left side of (48), we easily find characteristic values of the energy ω_* of the dipoles making up the resonant bidipole and also the characteristic transition amplitude in them:

$$\omega_* \sim U_* \sim T. \quad (55)$$

Since the quantity ψ is an invariant, relation (33) holds for all $\Gamma_\nu \neq 0$. Using (43), (52), and (54), we then find the following result for the characteristic parameters of the bidipoles:

$$\Omega^* \sim U_2^* \sim \Gamma^* \sim \gamma^4 r_*^6 l^2 T. \quad (56)$$

The characteristic size of the dipole is given in (52). The characteristic size of the bidipoles follows from the relation

$$U_2^* \approx E_d(R^*) \approx U_0 r_*^2 R^{*3}. \quad (57)$$

Specifically, we find

$$R^* \sim (r_*^4 l^2 T^2 / U_0)^{-1/3}. \quad (58)$$

In temperature region (53) the parameter $\gamma^4 r_*^6 l^2$ is much less than unity. Inequality (23) thus follows from (55)–(58). Substituting the characteristic parameters found for the problem into (42), we find that the radius of the resonant interaction, (42), agrees with the estimate (58). Finally, estimating the concentration of resonant bidipoles with the characteristic parameters (55)–(58), i.e., c_* , from the relation

$$\sum P_\nu \theta(U_\nu - \Gamma_\nu) \sim c_* \sim R_c^{-3}, \quad (59)$$

we find an estimate for the characteristic distance between resonant bidipoles, R_c , which agrees with (58). The equality of the radius of the resonant interaction and the average distance between the resonant bidipoles is in total agreement with the proposition that the resonant bidipoles form a connected infinite cluster with a quasicontinuous spectrum of excitations.⁸

To conclude this section of the paper we note that, formally, expression (54) can be used to estimate the typical lifetime of a resonant dipole (this time is finite, of course, because of the interaction with resonant bidipoles). For this purpose we substitute the characteristic value for $U_\nu \sim T$ into (54), since it is clear that only warm dipoles can play a significant role. From the same relation we can find an estimate of the typical size of such a dipole, which agrees with r_* [see (52)]. The characteristic reciprocal lifetime found for a resonant dipole,

$$\Gamma \sim (\gamma^4 r_*^6 l^2)^2 T, \quad (60)$$

is considerably smaller than the reciprocal characteristic time of a bidipole, (56). This circumstance indicates that the resonant dipoles have a significant inertia, so they can be completely eliminated from consideration in solving problems of low-temperature kinetics.

6. LOW-TEMPERATURE PHONON-FREE THERMAL CONDUCTIVITY

It turns out that the process of heat transfer over macroscopic distances can be associated with the self-relaxation mechanism described above. We will see below that at very low temperatures the mechanism proposed here may outweigh the classical mechanism, in which heat is transferred by phonons scattered by two-level centers.

It was shown above that irreversible hops are executed for the most part by electrons of resonant bidipoles, for which the constituent dipoles have approximately equal excitation energies. One of the dipoles, say dipole a , must be in the ground state, while the other, b , is the excited state. If this bidipole becomes resonant at a certain initial time, then after a time $t_\nu \sim \Gamma_\nu^{-1}$ this bidipole ceases to be resonant, and there is a finite probability that the electrons in it will undergo an irreversible hop. The result is an exchange of excitations between dipoles, so dipole a goes into the excited state, and dipole b into the ground state. In this manner energy is transported over a distance on the order of the size of a resonant bidipole, R . After a certain time, excited dipole a can in turn, as a result of a fluctuation of the transition energy, form a resonant bidipole with some other dipole c and, with a finite probability, transfer its excitation energy to this other dipole. Microscopic diffusion energy fluxes thus arise in the system. At thermodynamic equilibrium, these fluxes cancel out.

If there is a temperature gradient or, equivalently, a gradient of dipole excitations in the system, then a directed, uncanceled macroscopic energy flux arises in the system. Let us calculate this flux through a unit area of the $z=0$ plane, oriented perpendicular to the temperature gradient. If some bidipole is to contribute to the energy flux through this area by means of the mechanism described above, the dipoles making up this bidipole must lie on different sides of the area. Let us assume that a resonant bidipole whose center is at z and whose ends are at $z \pm R_\nu/2$ forms at a certain time (R_ν is the size of the bidipole). We assume that the first dipole, at $z + R_\nu/2$, is in an excited state, with a locally equilibrium distribution function (17), determined by the temperature $T(z + R_\nu/2)$. We assume that the second dipole, at $z - R_\nu/2$, has a locally equilibrium distribution function (17) with a temperature $T(z - R_\nu/2)$.

According to the mechanism proposed in the preceding section of this paper, there is a finite probability for an exchange of excitations between dipoles during the lifetime of the resonant cluster under consideration. The contribution from this resonant bidipole to the steady-state energy flux is proportional to the probability for the formation of a local-equilibrium resonant bidipole of type ν (with a different dipole temperature!). This probability can be written in a form similar to that of (28). The flux itself is equal to the product of this probability and the magnitude of the energy transferred (which is numerically the same as the transition energy in the dipoles), $|\omega'_\nu| \sim |\omega''_\nu|$, divided by the transfer time $t_\nu \sim \Gamma_\nu^{-1}$:

$$g_\nu^-(z) = -F_1(-|\omega'_\nu|, r'_\nu, T(z + R_\nu/2)) F_2(|\omega''_\nu|, r''_\nu, T$$

$$\begin{aligned} & \times (z - R\nu/2) U_2(\nu) \cdot |\omega'_\nu| \delta \\ & \times (|\omega'_\nu| - |\omega''_\nu|) \Gamma_\nu \theta(U_2(\nu) - \Gamma_\nu). \end{aligned} \quad (61)$$

If, on the contrary, the dipole at $z + R\nu/2$ is in the ground state, and that at $z - R\nu/2$ is in the excited state, then the contribution from this bidipole to the energy flux across the selected area is

$$\begin{aligned} g_\nu^+(z) &= -F_1(|\omega'_\nu|, r'_\nu, T(z + R\nu/2)) F_2 \\ & \times (-|\omega''_\nu|, r''_\nu, T(z - R\nu/2)) \\ & \times U_2(\nu) |\omega'_\nu| \delta(|\omega'_\nu| - |\omega''_\nu|) \Gamma_\nu \theta(U_2(\nu) - \Gamma_\nu). \end{aligned} \quad (62)$$

The total flux q across this area is determined by all resonant dipoles for which the condition $-R\nu/2 < z < R\nu/2$ holds. We thus have

$$q = \sum_\nu \int_{-R\nu/2}^{R\nu/2} dz (q^+ + q^-). \quad (63)$$

For an order-of-magnitude estimate we ignore the fact that the axis of the bidipole generally does not coincide with the normal to the area under consideration. Assuming that the temperature gradient is weak, we find from (61)–(63)

$$q = -\kappa \partial T / \partial z, \quad (64)$$

where

$$\begin{aligned} \kappa &= \partial / \partial T \sum F_1 \\ & \times (-|\omega'_\nu|, r'_\nu, T) F_2(|\omega''_\nu|, r''_\nu, T) U_2(\nu) |\omega'_\nu| \delta(|\omega'_\nu| \\ & - |\omega''_\nu|) \Gamma_\nu \theta(U_2(\nu) - \Gamma_\nu) R_\nu^2. \end{aligned} \quad (65)$$

This sum can be estimated in the same way as in (32), for example. As a result we find

$$\kappa \sim (T/U_0)^{4/3} r_*^{22/3} U_0 \gamma^{16/3} l^{5/3}. \quad (66)$$

The temperature dependence in (66) can be understood easily by drawing on some very simple ideas from kinetic theory. According to these ideas, the thermal conductivity is related to the specific heat c , the length scale λ of the energy transport per elementary event, and the time scale τ of this transport by

$$\kappa \sim c \lambda^2 / \tau. \quad (67)$$

As was shown above, the most effective bidipoles are those whose dipoles have an excitation energy $\omega \approx T$. We can thus assume $c \propto T$ (see also the more rigorous discussion in Ref. 5). The quantity λ is naturally associated with the characteristic size of the bidipole, R_* . The time τ is estimated as the characteristic lifetime of a resonant bidipole. We thus find

$$\kappa \sim TR_*^2 \Gamma_* \sim T^{4/3}, \quad (68)$$

in qualitative agreement with (66).

7. CONCLUSION

Let us discuss the results derived above. It was shown in Sec. 3 that the lifetime of resonant dipoles is finite in a system with a soft Coulomb gap at $T=0$ [see (40)]. This result

can be interpreted in the following way. A finite relaxation time implies an irreversible motion, so the system is not strictly speaking, in a pure ground state. Consequently, the entropy and therefore the temperature are not actually zero. This conclusion indicates that the description of the ground state of a disordered Coulomb system in the scenario of a soft Coulomb gap is an extremely approximate one. The reason is that, in the soft-gap approximation, the equality $T=0$ means only that there are no excited dipoles in this system [see (17)]. The possible existence of bidipoles in excited states remains outside the scope of the stability condition (9). If, on the other hand, we require that there be no such excitations [see (11)], then we find a more accurate description of the ground state of the Coulomb system: a hard Coulomb gap in which there is no trivial solution at $T=0$, as was shown at the end of Sec. 4. This situation is in complete accordance with the basic principles of thermodynamics. We thus reach the conclusion that only the hard-gap approximation can correctly describe the properties of disordered Coulomb systems in the limit $T \rightarrow 0$.

The soft-gap approximation is sufficient to describe the kinetics at $T \neq 0$ due to resonant bidipoles, because the solution of Eq. (45) is insensitive to the presence of a Coulomb gap if the temperature is not too low.

We recall that the thermal conductivity associated with the transport of phonons scattered by two-level systems has a T^2 behavior. We would thus expect that the thermal-conductivity mechanism discussed in Sec. 6 would be dominant at sufficiently low temperatures, since the decrease occurs by the much slower law in (68). The crossover between the two mechanisms for thermal conductivity requires a separate discussion. A deformation interaction between electrons, also of a dipole-dipole nature, may prove important there.

We sincerely thank Yu. Kagan for numerous useful discussions.

One of us (I.Ya.P.) was working under the auspices of the INTAS program (Grant 83-285). The work of the other authors (L.A.M.) and (A.L.B.) was supported financially by the Russian Fundamental Research Foundation (Grant 93-02-2540).

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Translated by D. Parsons