Low-temperature thermal conductivity of amorphous metals and alloys

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The Coulomb correction to the electronic thermal conductivity of amorphous metals and alloys is calculated taking into account the scattering of electrons by dynamical concentration excitations (DCE), which are introduced in order to describe the structural state of amorphous metallic systems. It is shown that at low temperatures the interference of inelastic electron–electron scattering and multiple elastic scattering of electrons by DCEs contributes to the thermal conductivity an amount comparable to its experimental values. The computed temperature dependence of the thermal conductivity reproduces the anomalous character of the function $k(T)$ for amorphous and metastable crystalline alloys at low temperatures. In the limit of low concentration of one of the components of the amorphous alloy the expression for $\delta k(T)$ is identical to the corresponding result obtained for an impure metal.

The universal character of the thermal conductivity of amorphous systems (AS) at low temperatures has recently been attracting the attention of many investigators.[1–9] Three types of temperature dependences of the thermal conductivity $k(T)$ are observed, irrespective of the type of chemical bond—covalent, ionic, or metallic: $k(T) \propto T^{2-a}$, $a \lesssim 0.3$, for $T < T_1$; $k(T) = \text{const}$ for $T_1 \lesssim T \lesssim T_2$; and, $k(T) \to k_{\text{cr}}(T) \propto T$ for $T > T_2$[1–9] [in Ref. 10 it is indicated that other types of temperature dependences $k(T)$ for $T > T_2$ are also possible]. The temperatures T_1 and T_2 for different types of amorphous systems (metals, dielectrics, semiconductors, polymers) differ by about one order of magnitude, namely, $T_1 \sim 1$–10 K, $T_2 \sim 10$–50 K.[1–9]

The thermal conductivity of metastable metallic alloys characterized by structural phase transitions $\omega \leftrightarrow \beta$ exhibit the same type of "glassy" behavior.[11] Being nondiffusive, $\omega \leftrightarrow \beta$ transitions are accompanied by atomic displacements of the order of 0.5 Å.[11] This enabled Lou to conclude in Ref. 11 that metastable metallic alloys have low-energy (as compared with phonon) atomic dynamics, similar to the dynamics of atoms in amorphous systems. This conjecture is corroborated by the fact that in these alloys not only the thermal conductivity but also the heat capacity, the electric conductivity, and the thermo-emf exhibit low-temperature "glassy" anomalies.[12–15] Neutron scattering experiments[16,17] and Mössbauer and x-ray spectroscopy[18,19] confirm the existence of coherent low-frequency lattice distortions in metastable alloys.

The low-temperature anomaly in the thermal conductivity of amorphous systems is interpreted with the help of a phenomenological model of two-level systems (TLS)[10,21] and different modifications of this model,[20] the microscopic soft-configuration model,[2–4] the quasiphonon model,[22] and other models.[9] The phonon thermal conductivity $k_{\text{ph}}(T)$ has been calculated within these theoretical models, and it has been shown that at very low temperatures ($T < 1$ K) $k_{\text{ph}}(T) \propto T^2$ (TLS model[20] and soft-configuration model[2–4]); in the temperature range $5 < T < 30$ K $k_{\text{ph}} = \text{const}$ (soft-configuration model[2–4] and quasiphonon model[22]); and, finally, at high temperatures [in the quasiphonon model $T_2 \sim 200$ K (Ref. 22)] $k_{\text{ph}}(T) \propto T$. In spite of the fact that the results obtained in Refs. 2–4, 10, and 20–22 are in good qualitative agreement with the experiments, it seems to us that the analysis of the low-temperature features of the thermal conductivity of amorphous metals and alloys performed on the basis of these results is incomplete. For metallic amorphous systems it is also necessary to calculate the electronic contribution to $k_{\text{ph}}(T)$, and it is well known[1,23] that in metals this contribution can be significantly greater than the phonon contribution to the thermal conductivity.

The problem of the electronic contribution to $k(T)$ for impure metals was addressed in Refs. 24–26. In Ref. 26 it was solved by the quantum kinetic equation method,[27] based on Keldysh's diagrammatic technique. This method has been used successfully to calculate temperature-dependent corrections introduced in the impurity conductivity by the electron–electron interaction (EEI)[28] and scattering of electrons by phonons.[29] The advantage of the method of Ref. 27 is that it can be easily extended to the case of any other long-wavelength scattering, since in Ref. 27 no assumptions other than isotropy of the metal and smallness of the phonon contribution to the damping of the electrons are made in the derivation of the kinetic equation.

The description of the low-temperature minimum of the electric resistance and the minimum in the electronic density of states at the Fermi level,[28] which are observed experimentally in metallic amorphous systems also,[1,21] is an unquestionable successful achievement of the theory of EEI in impure metals. It is obvious, however, that the EEI theory[28] cannot be applied in its original form to amorphous alloys in which the concentrations of the components are of the same order of magnitude. In these systems the electrons are scattered not by a random impurity but rather by cluster-type structural nonuniformities.[30] For this reason, in order to describe the electronic transport in metallic amorphous systems the EEI theory must be extended to the case of electron scattering by structural formations of the short-range-order type.

We proposed such an extension in Refs. 31–34, where it is shown that the anomalous low-temperature properties of metallic glasses (the electric resistance, the electronic heat capacity, and the thermo-emf) are determined by the interference of inelastic interelectronic interaction and multiple elastic scattering of electrons by DCEs. The DCEs are dy-

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namical concentration excitations of the electron–ion system, which are responsible for the cooperative nondiffusive rearrangement of local atomic configurations in metallic amorphous systems.[31–34]

Our calculation of the Coulomb corrections to the electric resistance and electronic density of states of amorphous metals and alloys[31,32] in the limit when the concentration of one of the components of the alloy $c \to 0$ agrees with the theory of EEI in impure metals.[28]

In the present paper we calculate the contribution of the Coulomb electron–electron interaction and multiple elastic scattering of electrons by DCEs to the electronic thermal conductivity of metallic amorphous systems.

A systematic exposition of the concept of DCEs as elementary excitations of amorphous metallic systems is given in Sec. 1. In Sec. 2 $\delta k(T)$ is calculated by the quantum kinetic equation method, as was done in Ref. 26 for impure metals.

1. ELEMENTARY EXCITATIONS IN AMORPHOUS METALLIC SYSTEMS

Consider an amorphous alloy formed by quenching from the liquid state (melt). At temperatures $T < T_g$ there exist in the system many structural states of regions (clusters) with different types of short-range order (SRO). A local minimum of the interatomic interaction potential can correspond to some of these states. When the quenched state is annealed (with increasing T) the state of the amorphous system becomes more ordered—the existing short-range-ordered regions relax into clusters, which are more "convenient" for the particular amorphous system at hand. The random position of the ions in such a system can be written as follows:

$$\mathbf{R}_l^N = \mathbf{R}_{0l}^N + \mathbf{u}_l^N + \boldsymbol{\xi}_l^N,$$

where $\mathbf{R}_{0l}^N$—the equilibrium position of an ion—is a "lattice" site of the short-range-ordered region (of the N-th cluster); $\mathbf{u}_l^N$ are the dynamical thermal displacements of ions with respect to $\mathbf{R}_{0l}^N$; $\xi_l^N = \vec{\mathscr{R}}_l \delta c(\mathbf{R}_l, t)$ are displacements induced by the dynamical fluctuations of the concentration $\delta c(\mathbf{R},t) = c(\mathbf{R},t) - c$ where $c(\mathbf{R},t)$ and c are the microscopic and macroscopic concentration of the components of the amorphous system. The quantity $\vec{\mathscr{R}}_l$ will be defined below.

The position of the ions in the Nth cluster can be represented conventionally as in Fig. 1. Different pairs (triplets, quadruplets, etc.) of chemically bound atoms can form in the amorphous system as a result of quenching. The change in ξ_l with time is associated with quenching and relaxation of the structure of the amorphous system.

In crystalline solids the ξ_l are static displacements, and their existence is demonstrated, for example, by diffuse scattering of x-rays.[35] The displacement ξ_l is essentially the displacement of an ion from $\mathbf{R}_{0l}$ due to the formation of a new chemical bond owing to quenching of some short-range order that is uncharacteristic for the structure given by the configuration $\{\mathscr{R}_{0l}\}$. The vector $\vec{\mathscr{R}}_l$ appearing in ξ_l is the static displacement of a single "defect"—atom (ion) in a position inconvenient for the given cluster. In the simplest case of a point defect $\vec{\mathscr{R}}_l$ can be represented in the form[35]

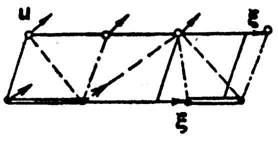

FIG. 1. Conventional representation of the position of ions in the Nth cluster: $\mathbf{u}_l$—temperature displacement of an ion relative to its equilibrium position; ξ_l—displacement due to dynamical concentration fluctuations.

$$\vec{\mathscr{R}}_l = \frac{\mathbf{R}_l}{R_l^3} \frac{1+\sigma}{12\pi(1-\sigma)} \frac{\partial V}{\partial \delta c_l},$$

where $\mathbf{R}_l$ is the position of the ion, σ is Poisson's ratio, and V is the atomic volume.

The concentration fluctuation $\delta c(\mathbf{R},t)$, characterizing the number of such "defects" in a cluster and the dynamical changes in ξ_l, can in turn be represented as follows:

$$\delta c_l(\mathbf{R}, t) = \frac{1}{\Omega} \int\int c_\mathbf{k}(\omega) \exp[-i(\mathbf{k}\mathbf{R}_l - \omega t)] d\mathbf{k}\, d\omega,$$

where $\mathbf{k}$ is a reciprocal lattice vector.

The average $\langle |c_\mathbf{k}|^2 \rangle$ over the observation time t can be taken as

$$\langle |c_\mathbf{k}|^2 \rangle_\omega = \langle |c_\mathbf{k}|^2 \rangle_\Omega = \langle |c_\mathbf{k}|^2 \rangle_{\{\mathbf{R}^N\}},$$

and, in particular, for binary systems[35]

$$\langle |c_\mathbf{k}|^2 \rangle_\Omega = c(1-c) \sum_l^N \alpha_l \exp(i\mathbf{k}\mathbf{R}_l),$$

where Ω is the volume of the system and α_l is the short-range-order parameter.

The configurational rearrangement of the uncharacteristic short-range-ordered regions, which is described by the variable $\delta c(\mathbf{R},t)$, is caused by transfer of electrons and, correspondingly, ions into a more favorable spatial position. As a result, there arises a new (more advantageous) chemical bond between the ions in a cluster. In metallic amorphous systems such a bond between ions is of a resonance character and can be realized by "resonance" $d(f)$-electrons. This is due to a characteristic feature of the electronic spectrum of disordered alloys. It is shown in Ref. 36 that narrow resonance fluctuation bands (FB), associated with the transfer of an atom of one type into "sites" of atoms of a different type, appear in the electronic spectra of disordered alloys together with a crystal-phase (CP) band. Therefore excited states, associated with disorder and maintained by external actions, appear in the electronic subsystem of disordered alloys. As the intensity of the external action decreases, the number of electrons in such an excited state decreases, and the fluctuation bands relax into crystal-phase bands. This releases sufficient energy for the ions to be transferred into more favorable spatial positions.

In our opinion, the electronic spectrum of amorphous metals should be characterized by a collection of different fluctuation bands, corresponding to all uncharacteristic configurations $\{\mathbf{R}_l\}$. For this reason, the relaxation of fluctuation bands into crystal-phase bands in amorphous systems can proceed both as a direct transition and sequentially—by a transition of one fluctuation band into another. The structural relaxation of metallic amorphous systems thus consists of a configurational rearrangement of uncharacteristic short-range-ordered regions, which is realized by a transfer of electrons and, correspondingly, ions into a more favorable spatial position. Such a cooperative atomic motion is nondiffusive and, evidently, analogous to the motion observed in alloys undergoing martensitic transformations.

We now determine the explicit form of the Hamiltonian H for the electron–ion system examined above:

$$H=H_i(\mathbf{R})+H_e(\mathbf{r})+H_{e-i}(\mathbf{R},\ \mathbf{r}). \tag{1.1}$$

Since we are concerned with amorphous systems at low temperatures, for which U_l can be assumed to be frozen out, we set $\mathbf{R}_l^N \approx \mathbf{R}_{0l}^N + \xi_l$.

The first term of the Hamiltonian (1.1) is the ionic Hamiltonian

$$H_i(\mathbf{R})=\sum_l \frac{M_l \mathcal{R}_l^2}{2}(\delta\dot{c}_l)^2+\frac{1}{2}\sum_{l,m} W(\mathbf{R}_l-\mathbf{R}_m)\delta c_l \delta c_m, \tag{1.2}$$

where M_l is the ion mass. If the δc_l form the concentration field, then the kinetic energy is $\propto \nabla^2 \delta c(\mathbf{R},t)$. Hence it is obvious that if the gradients of the concentration fluctuations in the system under consideration are small (weak nonequilibrium), then the kinetic energy of the ions can be neglected compared with the potential energy [second term in Eq. (1.2)]. If, however, $\nabla\delta c$ is large (strong nonequilibrium), then this contribution cannot be neglected.

It is interesting to make one other observation concerning the kinetic energy of ions. Since the term $M_l \mathcal{R}_l^2$ in $\Sigma_l M_l R_l^2 (\delta\dot{c}_l)^2/2$ is a certain moment of inertia, $\delta\dot{c}(l)$ can evidently be interpreted as an angular velocity. Then the motion of the ions in a transition to equilibrium is of a nondiffusive "rotational" character with displacement much less than the interatomic distance. As a result of this, the potential energy can be represented in Eq. (1.2) in the "harmonic approximation," where

$$W(\mathbf{R}_l^N-\mathbf{R}_m^N)=\left.\frac{\partial^2 U(\mathbf{R}_l^N-\mathbf{R}_m^N)}{\partial(\delta c_l)\,\partial(\delta c_m)}\right|_{\delta c=0}$$

are the "force constants," which determine the change brought about in the coupling forces by the concentration fluctuations. Here $U(\mathbf{R}_l^N-\mathbf{R}_m^N)$ is the pair interaction potential energy of ions located at the points $\mathbf{R}_l$ and $\mathbf{R}_m$ of the Nth cluster.

The electronic Hamiltonian H_e in the total Hamiltonian (1.1) has the form

$$H_e=\sum_i \frac{\mathbf{p}_i^2}{2m}+\sum_i V^{\text{eff}}(\mathbf{r}_i),$$

where $\mathbf{p}_i$ and m are the electron momentum and the electron mass, respectively, and $V^{\text{eff}}(r_i)$ is the effective single-particle potential acting on an electron at the point r_i.

Finally, the last term in Eq. (1.1) describes the interaction of the ions themselves with the electrons participating in the change in the interionic chemical bond:

$$H_{e-i}=\sum_{l,i}^{N,n} \delta c_l v(|\mathbf{r}_i-\mathbf{R}_l|),$$

where v is the electron–ion interaction potential. This interaction determines the change in the energy of the n electrons in the N-th cluster when the ions move from $\mathbf{R}_0^N$ to $\mathbf{R}_l^N$. This change in the energy is

$$\sum_{l,i} \left.\frac{\partial V_{\delta c_l}(|\mathbf{r}_i-\mathbf{R}_l|)}{\partial \xi_l}\right|_{\mathbf{R}=\mathbf{R}_0}\cdot\xi_l$$

$$=\sum_{l,i} \left.\frac{\partial V_{\delta c_l}(|\mathbf{r}_i-\mathbf{R}_l|)}{\partial(\delta c_l)}\right|_{\delta c_l=0} \equiv \sum_{l,i} \delta c_l v(|\mathbf{r}_i-\mathbf{R}_l|),$$

and corresponds to some "deformation potential" arising with a change in the position of the ions as a result of concentration fluctuations. The index δc_l in $V_{\delta c_l}(|\mathbf{r}_i-\mathbf{R}_l|)$ is inserted in order to underscore the explicit dependence of the electronic potential on the change in the atomic environment.

The classical equation of motion of the ions, which is associated with ξ_l, has the form

$$\sum_l M_l \mathcal{R}_l^2 \delta\ddot{c}_l=-\sum_{ll'} W(\mathbf{R}_l-\mathbf{R}_{l'})\delta c_l. \tag{1.3}$$

In the single-mode approximation (cluster with a given type of short-range order) we seek δc_l in the form

$$\delta c_l=\frac{1}{2}\left[\frac{c(1-c)}{N}\alpha_l\right]^{1/2}\{b_{\mathbf{k}_s}\exp[i(\mathbf{k}_s\mathbf{R}-\omega(\mathbf{k}_s)t)]+\text{c.c.}\},$$

where $\mathbf{k}_S$ is a "superstructure" vector corresponding to the given type of short-range order, and the energy $\omega(\mathbf{k}_S)$ is, in the case of resonance, the difference of the energy of the electrons in the fluctuation band $(\varepsilon_{\mathbf{k}+\mathbf{k}_S})$, associated with the presence of the given uncharacteristic type of chemical bond between ions, and the energy of electrons in the crystal-phase band $(\varepsilon_{\mathbf{k}})$, to which the amorphous system relaxes.

Then Eq. (1.3) assumes the form

$$\sum_l M_l \mathcal{R}_l^2 \alpha_l^{1/2} \omega^2(\mathbf{k}_s)$$

$$=\sum_{l,l'} W(\mathbf{R}_l^N-\mathbf{R}_{l'}^N)\alpha_l^{1/2}\exp[i\mathbf{k}_s(\mathbf{R}_l^N-\mathbf{R}_{l'}^N)]. \tag{1.4}$$

Introducing the dynamical matrix

$$D_l(\mathbf{k}_s)=\sum_l W(\mathbf{R}_l^N-\mathbf{R}_{l'}^N)\alpha_{l'}^{1/2}\exp[i\mathbf{k}_s(\mathbf{R}_l^N-R_{l'}^N)],$$

we obtain from Eq. (14) the scalar equation

$$\|M_l \mathcal{R}_l^2 \omega^2(\mathbf{k}_s)\alpha_l^{1/2}-D_l(\mathbf{k}_s)\|=0, \tag{1.5}$$

which enables us to find the dispersion relations for the frequencies $\omega(\mathbf{k}_S)$, if the form of the ion pair interaction potential energy is known. We note that, in contrast to the classical definition of the dynamical matrix in crystals, in the case of amorphous systems $D_l(\mathbf{k}_S)$ contains the short-range-order parameter $\alpha_{l'}$.

In the classical equation of motion of the ions (1.3) the quantity $M_l\vec{\mathcal{R}}_l\delta c(\mathbf{R}_l)$ plays the role of a generalized coordinate $\mathbf{Q}_l$ and $M_l\vec{\mathcal{R}}_l\delta\dot{c}(\mathbf{R}_l)$ plays the role of a generalized momentum $P_l = \dot{\mathbf{Q}}_l$. At low temperatures, in the case of strong nonequilibrium, when $\Delta\mathbf{Q}_l$ are small and $\Delta\mathbf{P}_l$ are large (i.e., $\nabla\delta c_l$ are large), the uncertainty relation $\Delta\mathbf{P}_l\Delta\mathbf{Q}_l \sim \hbar$ is obviously satisfied. Therefore, at low temperatures we can talk about the quantum motion of ions in amorphous systems. In order to describe this motion it is convenient to introduce field creation and annihilation operators φ^+ and φ, related with $\mathbf{P}_l$ and $\mathbf{Q}_l$ by the canonical transformation

$$\mathbf{Q}_l = \frac{M_l\vec{\mathcal{R}}_l}{2}[c(1-c)\alpha_l]^{1/2}(\varphi^+ + \varphi),$$

$$\mathbf{P}_l = -\frac{i}{M_l\mathcal{R}_l[c(1-c)\alpha_l]^{1/2}}(\varphi^+ - \varphi),$$

where $\mathbf{P}_l$ and $\mathbf{Q}_l$ satisfy the commutation relations $[\mathbf{Q},\mathbf{Q}] = [\mathbf{P},\mathbf{P}] = 0$ and $[\mathbf{Q},\mathbf{P}] = i\hbar$, while φ and φ^+ satisfy the condition $[\varphi^+,\varphi] = 1$.

Then we obtain for the concentration fluctuations

$$\delta c(\mathbf{R}_l,\ t) = {}^1/_2[c(1-c)\alpha_l]^{1/2}(\varphi_l^+ + \varphi_l). \tag{1.6}$$

With the help of the expressions presented for $\mathbf{P}_l$ and $\delta c(\mathbf{R}_l,t)$ we obtain from Eq. (1.2)

$$H_i = \sum_l A_l\varphi_l^+\varphi_l + \sum_{ll'}{}' \mathcal{M}_{ll'}\left(\varphi_l^+\varphi_{l'} + \frac{1}{2}\varphi_l^+\varphi_{l'}^+ + \frac{1}{2}\varphi_l\varphi_{l'}\right),$$

where

$$A_l \equiv -\frac{1}{c(1-c)}\frac{1}{M_l^3\mathcal{R}_l^2\alpha_l}$$

$$\mathcal{M}_{ll'} \equiv {}^1/_4 c(1-c)W(|\mathbf{R}_l - \mathbf{R}_{l'}|)\alpha_l\alpha_{l'}.$$

The unitary transformation[37]

$$\varphi_l = N^{-1/2}\sum_{\mathbf{k}}\{u(\mathbf{k})\exp(i\mathbf{k}\mathbf{R}_l)b_{\mathbf{k}} + v^*(\mathbf{k})\exp(-i\mathbf{k}\mathbf{R}_l)b_{\mathbf{k}}^+\}, \tag{1.7}$$

where the new Bose operators b_{k} and b_{k}^+ satisfy the commutation relations $[b_{\mathrm{k}},b_{\mathrm{k'}}^+] = \delta_{\mathrm{kk'}}$, diagonalizes the ionic Hamiltonian.[37] The transformation (1.7) is unitary, if

$$u^*(\mathbf{k})u(\mathbf{k}) - v^*(\mathbf{k})v(\mathbf{k}) = 1,$$

$$\sum_{\mathbf{k}}\exp[i\mathbf{k}(\mathbf{R}_l - \mathbf{R}_{l'})] = N\delta_{ll'}.$$

The diagonalized ionic Hamiltonian is

$$H_i = -\sum_{\mathbf{k}}E(\mathbf{k})v(\mathbf{k})v^*(\mathbf{k}) - \sum_{\mathbf{k}}E(\mathbf{k})b_{\mathbf{k}}^+b_{\mathbf{k}}.$$

Then the following conditions are satisfied:

$$\begin{aligned} &[A_l - E(\mathbf{k})]u(\mathbf{k}) + \mathcal{M}_l(\mathbf{k})[u(\mathbf{k}) + v(\mathbf{k})] = 0,\\ &[A_l + E(\mathbf{k})]v(\mathbf{k}) + \mathcal{M}_l(\mathbf{k})[u(\mathbf{k}) + v(\mathbf{k})] = 0, \end{aligned} \tag{1.8}$$

where

$$\mathcal{M}_l(\mathbf{k}) = \sum_{l'}\mathcal{M}_{ll'}\exp[i\mathbf{k}(\mathbf{R}_l - \mathbf{R}_{l'})].$$

The first term in the diagonalized ionic Hamiltonian corresponds to the "vacuum" state of the amorphous system, in which there are no excitations associated with the quantum motion of ions owing to dynamical short-range order. The Hamiltonian of these excitations (DCE) has the form

$$H_{\mathrm{DCE}} = \sum_{\mathbf{k}}E(\mathbf{k})b_{\mathbf{k}}^+b_{\mathbf{k}}, \tag{1.9}$$

where

$$E(\mathbf{k}) = \sum_l\{[A_l + \mathcal{M}_l(\mathbf{k})]^2 - \mathcal{M}_l^2(\mathbf{k})\}^{1/2}$$

is the energy of the DCE, which is determined from the solution of the system of equations (1.8).

According to the expression for $E(\mathbf{k})$, substituting the definitions of A_l and $\mathcal{M}_l(\mathbf{k})$, the DCE decay if $|A_l| < 2\mathcal{M}_l(\mathbf{k})$ or

$$\sum_{l'}W(\mathbf{R}_l - \mathbf{R}_{l'})\alpha_{l'}\exp[i\mathbf{k}(\mathbf{R}_l - \mathbf{R}_{l'})] > \frac{1}{c(1-c)M_l^3\mathcal{R}_l^2\alpha_l}.$$

Conversely, if $|A_l| > 2\mathcal{M}_l(\mathbf{k})$ or

$$\sum_{l'}W(\mathbf{R}_l - \mathbf{R}_{l'})\alpha_{l'}\exp[i\mathbf{k}(\mathbf{R}_l - \mathbf{R}_{l'})] < \frac{1}{c(1-c)M_l^3\mathcal{R}_l^2\alpha_l}$$

then the DCE are nondecaying excitations.

In the case $|A_l| \gtrsim 2\mathcal{M}_l(\mathbf{k})$ we can talk about real long-lived states in the amorphous system—low-energy DCE. They are of special interest for studying the properties of amorphous systems at low temperatures, since it has been shown experimentally that it is the low-energy excitations determined by the structural state of the amorphous system that are responsible for the anomalous behavior of the physical properties of amorphous systems at low temperatures. In Refs. 31–34 we proposed a theory of the anomalous electrical resistance, the thermo-emf, and the electronic heat capacity based on the DCE concept. The main results of Refs. 31–34 are as follows:

1. The contribution made to the temperature dependence of the electric conductivity by the interference of inelastic electron–electron scattering and multiple elastic scattering of electrons by DCE has the form

$$\frac{\delta\sigma(T)}{\sigma} \approx \frac{2{,}5\cdot 2^{1/2}}{6}\frac{T^{1/2}}{\nu_0 D^{3/2}}\left[1 + \left(\frac{T}{T_0}\right)^2\right]^{-1/4},$$

where ν_0 is the initial electron density of states at the Fermi level, D is the electron diffusion coefficient, and T_0 is a temperature, defined below, typical of a specific amorphous system. Figure 2 shows the $\delta\rho(T)/\rho$ dependence taken from Ref. 31.

2. The thermo-emf of nonmagnetic amorphous alloys, calculated taking into account the same scattering processes as for the electric conductivity, is defined as follows [Fig. 3 (Refs. 33 and 34)]:

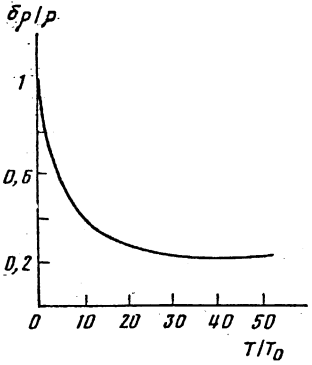

FIG. 2. Temperature dependence of the contribution $\delta\rho(T)/\rho$.[31,32]

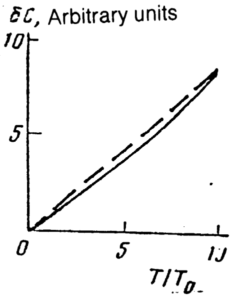

FIG. 4. Contribution $\delta c(T)$ to the electronic heat capacity.[31,32]

$$S(T) = \frac{k_B\pi}{6|e|}\Psi_{1/2}^{-1}\left(\frac{1}{4\tau T}\right)\Psi_{3/2}\left(\frac{1}{4\tau T}\right),$$

where

$$\Psi_\alpha(x) = \int_0^\infty \frac{dy}{y^{1/2}}\frac{1}{\mathrm{ch}^2(y+x)}\frac{1}{(y+x)^\alpha}.$$

3. The corresponding contribution to the electronic heat capacity

$$\delta C(T) = \tfrac{1}{3}\pi^2 k_B^2\,\delta\nu(\varepsilon_F)$$

is determined by the contribution of the indicated scattering processes to the electron density of states:[31,32]

$$\delta\nu(T) \approx \frac{2,6}{2^{3/2}\pi^2 D^{1/2}}T^{1/2}\left[1+\left(\frac{T}{T_0}\right)^2\right]^{-1/4}.$$

The temperature dependence δC (Ref. 31) is shown in Fig. 4. According to Figs. 2–4, it is at low temperatures $T \lesssim 50$ K, when the scattering of electrons by low-temperature DCE is significant, that all quantities computed in Refs. 31–34 are anomalous. For this reason it is of interest to calculate the temperature dependence of the electronic thermal conductivity associated with scattering of electrons by one another and by DCE.

2. COULOMB CORRECTION TO THE THERMAL CONDUCTIVITY

In order to calculate the contribution of the interference of inelastic electron–electron scattering and multiple elastic scattering of electrons by DCE to $k(T)$ on the basis of the quantum kinetic equation, it is necessary to determine the temperature Green's functions of the electrons and DCE in the amorphous system. In Refs. 31 and 32 we showed that the temperature Green's function of DCE is defined as the Fourier transform of the correlation function $d(x-x') = \langle \hat{T}\gamma(x)\gamma(x')\rangle$, where $x = (\mathbf{r},t)$, $\hat{T}$ is the chronological operator, and $\gamma(\mathbf{r},t) = \Sigma_{\mathbf{R}}g(\mathbf{r}-\mathbf{R})\cdot\delta c(\mathbf{R},t)$. Here $g(\mathbf{R}) = V_A(\mathbf{R}) - V_B(\mathbf{R})$, where $V_{A(B)}(\mathbf{R})$ are the "site" potentials of ions of species A (B), and $\delta c(\mathbf{R},t)$ is defined in Eq. (1.6). At low temperatures, when low-energy DCE are important, their temperature Green's function has the form[31,32]

$$\mathscr{D}(\mathbf{k},\omega_c) = \frac{c(1-c)}{\rho_0}|\mu|\pi\frac{\Delta^2(\mathbf{k})}{\omega_c}\delta(E_{\mathbf{k}}), \tag{2.1}$$

where ρ_0 is the density of the material, $\Delta(\mathbf{k}) = \sqrt{c(1-c)/2N}\,g(\mathbf{k})$, $g(\mathbf{k})$ is the Fourier transform of $g(\mathbf{R})$, N is the number of atoms in the volume, μ is the DCE chemical potential, and $E_{\mathbf{k}}$ are the eigenvalues of the diagonalized DCE Hamiltonian (1.9).

The temperature Green's function of the electrons in amorphous metals and alloys, neglecting the interaction of the electrons with one another, has the form

$$G_0(\mathbf{p},\varepsilon_n) = \left[i\varepsilon_n - \xi(\mathbf{p}) + \frac{i}{2\tau}\,\mathrm{sign}\,\varepsilon_n\right]^{-1}, \tag{2.2}$$

where

$$\frac{1}{\tau} \propto \int\frac{d^3\mathbf{p}'}{(2\pi)^3}|g(\mathbf{p}-\mathbf{p}')|^2\delta c(\mathbf{p})\delta c(\mathbf{p}')[i\varepsilon_n - \xi(\mathbf{p}')$$
$$+ i\delta\,\mathrm{sign}(|\mathbf{p}'| - \mathbf{p}_F)]^{-1}.$$

Here $\xi(\mathbf{p}) \equiv \mathbf{p}^2/2m - \mu_0$; $\mathbf{p}$, ε_n and μ_0 are the momentum, energy, and chemical potential of electrons with mass m; τ is the electron momentum relaxation time, taking into account scattering of electrons by DCE; $\delta c(\mathbf{p})$ and $g(\mathbf{p}-\mathbf{p}')$ were defined above.

In Keldysh's technique employed in the quantum kinetic equation method[27] the Green's function, the self-energy of the electrons, and the interelectron interaction potential are represented by the matrices

$$\hat{G} = \begin{pmatrix} 0 & G^A \\ G^R & G^c \end{pmatrix}, \quad \hat{\Sigma} = \begin{pmatrix} \Sigma^c & \Sigma^R \\ \Sigma^A & 0 \end{pmatrix}, \quad \hat{V} = \begin{pmatrix} 0 & V^A \\ V^R & V^c \end{pmatrix}. \tag{2.3}$$

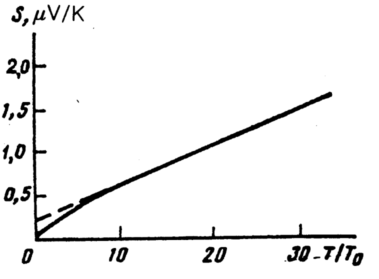

FIG. 3. Thermo-emf of amorphous metals and alloys as a function of temperature.[33,34]

The heat flux is expressed in terms of the electron Green's function as follows:[26]

$$\mathbf{q}=-k\nabla T=\int\frac{d^3\mathbf{p}\,d\varepsilon}{(2\pi)^4}\varepsilon\mathbf{v}s(\mathbf{p},\varepsilon)\,\mathrm{Im}\,G^A(\varepsilon,\mathbf{p}),\tag{2.4}$$

where $\mathbf{v}$ is the electron velocity and $s(\mathbf{p},\varepsilon)$ is a distribution function [at equilibrium $s(\mathbf{p},\varepsilon)=s_0(\varepsilon)=-\tanh(\varepsilon/2T)$].

The advanced and retarded Green's functions are connected with the temperature Green's function by a well-known relation,[38] so that we have from Eq. (2.2)

$$[G_0{}^A(\varepsilon,\,\mathbf{p})]^*=G_0{}^R(\varepsilon,\,\mathbf{p})=[\varepsilon-\xi(\mathbf{p})+i/2\tau]^{-1}.\tag{2.5}$$

The Green's function G^c, to first order in inhomogeneity, has the form[26]

$$G^c(\varepsilon,\,\mathbf{p})=s(\varepsilon,\,\mathbf{p})[G^A-G^R]+{}^1\!/_2 i\{s_0(\varepsilon),\,G^A+G^R\}.\tag{2.6}$$

The Poisson brackets in the presence of a temperature gradient is expanded as follows[26]

$$\{A,B\}=\nabla T\left[\frac{\partial A}{\partial T}\frac{\partial B}{\partial\mathbf{p}}-\frac{\partial A}{\partial\mathbf{p}}\frac{\partial B}{\partial T}\right].$$

The kinetic equation for the electron distribution function, linearized with respect to ∇T, is

$$\mathbf{v}\nabla T\frac{\partial s_0(\varepsilon)}{\partial\varepsilon}\frac{\varepsilon}{T}=I_{e\text{-}e}+I_{\text{e-DCE}},\tag{2.7}$$

where I are collision integrals, which are related, respectively, with the electron–electron interaction and the scattering of electrons by DCE.

The kinetic equation (2.7) is solved by the method of iterations with $s=s_0+\varphi_0+\varphi_1$, as done in Ref. 26, since we assume that the main electron momentum relaxation mechanism is scattering of electrons by DCE. In the absence of electron–electron interaction the nonequilibrium correction to the distribution function is[26]

$$\varphi_0(\varepsilon,\mathbf{p})=\tau\mathbf{v}\nabla T\frac{\partial s_0(\varepsilon)}{\partial\varepsilon}\frac{\varepsilon}{T}.\tag{2.8}$$

To first order in the perturbation theory in the interaction

$$\varphi_1(\varepsilon,\ p)=\tau\{I_{e\text{-}e}(s_0+\varphi_0)+\delta_{int}I_{\text{e-DCE}}(s_0+\varphi_0)\},\tag{2.9}$$

where $\delta_{\text{int}}I_{\text{e-DCE}}$ is the correction to the collision integral, determined by the scattering of electrons by DCE, due to the renormalization of the electron density of states

$$\delta_{int}I_{\text{e-DCE}}(\varepsilon,\mathbf{p})=\frac{2}{\pi\nu_0\tau}\int\frac{d^3\mathbf{k}}{(2\pi)^3}[s(\varepsilon,\mathbf{k})-s(\varepsilon,\mathbf{p})]$$

$$\times\,\mathrm{Im}[\delta_{int}G^A(\varepsilon,\mathbf{k})].\tag{2.10}$$

Here $\delta_{\text{int}}G^A=(G_0^A)^2\Sigma_{e-e}^A(s_0+\varphi_0)$ is the correction introduced by the electron–electron interaction into the electronic Green's function. According to Eqs. (2.4), (2.7), and (2.8), the correction $\delta\kappa$ to the thermal conductivity is made up of the correction to the distribution function $\varphi_0(\varepsilon,\mathbf{p})$ and the correction to the electron density of states:

$$\delta k=-\frac{2}{\nabla T}\int\frac{d^3\mathbf{p}\,d\varepsilon}{(2\pi)^4}\varepsilon(\mathbf{v},\mathbf{n})\{\varphi_1\,\mathrm{Im}\,G_0{}^A+\varphi_0\,\mathrm{Im}[\delta_{int}G^A(s_0)]$$

$$+s_0\,\mathrm{Im}[\delta_{int}G^A(\varphi_0)]+s_0\,\mathrm{Im}[\delta G^A(s_0)]\},\tag{2.11}$$

where $\mathbf{n}$ is a unit vector directed along ∇T.

In Refs. 31 and 32 we calculated in terms of temperature Green's functions the correction introduced by the electron–electron interaction and multiple scattering of electrons by DCEs into the electronic Green's function. The calculation was performed in the ladder approximation in the interaction of the electrons with DCE and to first order in the Coulomb electron–electron interaction.

The screened Coulomb potential, calculated in the indicated approximation, has the form[31,32]

$$V(\omega_m,\mathbf{q})=\frac{4\pi e^2}{q^2\varepsilon(\omega_m,\mathbf{q})},$$

$$\varepsilon(\omega_m,\mathbf{q})=1+\frac{D\varkappa^2\theta(\omega_m)}{\omega_m+Dq^2-i(T/T_0)\omega_m}$$

$$+\frac{D\varkappa^2\theta(-\omega_m)}{-\omega_m+Dq^2+i(T/T_0)\omega_m}\tag{2.12a}$$

where $\theta(x)$ is a step function, $\varkappa^2\equiv4\pi e^2\nu_0$, $D=\frac{1}{3}v_F^2\tau$, and v_{F} is the Fermi velocity. The quantity

$$T_0=\frac{16(2\pi)^3[\mu_0{}^2+{}^1\!/_4\tau^2]^{\overline{3/4}}\omega_0{}^2}{15{,}45\mu_0\varkappa\Delta^2(0)(2m)^{3/2}c(1-c)|\mu|},$$

where ω_0 is the DCE limiting frequency, has the dimension of temperature. According to the estimates given for this quantity in Refs. 31–34, $T_0\sim1$–5 K. From the definition of T_0 one can see that it is a function of the concentration c of the components of the alloy, of the formation energy ΔE ($\sim\Delta(0)$), and of other physical characteristics of amorphous alloys.

It follows from Eq. (2.12a) that

$$V^R(\omega,\mathbf{q})=[V^A(\omega,\mathbf{q})]^*$$

$$=\frac{4\pi e^2}{q^2}\left\{1+\frac{D\varkappa^2}{-i\omega+Dq^2-(T/T_0)\omega}\right\}^{-1}.\tag{2.12b}$$

The interaction vertex function $\gamma(\omega_m,\mathbf{q},\varepsilon_n)$, obtained in Refs. 31 and 32 in a calculation of the correction to the electron Green's function, has the form

$$\gamma(\omega_m,\mathbf{q},\varepsilon_n)=\theta(\varepsilon_n)\theta(\varepsilon_n+\omega_m)+\theta(-\varepsilon_n)\theta(-\varepsilon_n-\omega_m)$$

$$+\frac{\theta(\varepsilon_n)\theta(-\varepsilon_n-\omega_m)}{-\omega_m+Dq^2+i(T/T_0)\omega_m}+\frac{\theta(-\varepsilon_n)\theta(\varepsilon_n+\omega_m)}{\omega_m+Dq^2-i(T/T_0)\omega_m}.\tag{2.13}$$

Thus we have determined all quantities appearing in Eqs. (2.4), (2.7), (2.8), and (2.11). The equation (2.1) is completely analogous to the corresponding equation derived for the case of impurity scattering.[26] Its solution is presented in detail in Ref. 26 together with an analysis of the contribution of all diagrams to δk. For this reason, we give directly the final expression for the Coulomb correction to the thermal conductivity of amorphous metals and alloys:

$$\delta k(T)=\frac{2\pi^2\nu_0\tau}{3}\int\frac{d\varepsilon\,d\omega}{(2\pi)^2}s_0(\varepsilon)\frac{\partial s_0(\varepsilon+\omega)}{\partial\varepsilon}\frac{\varepsilon(\varepsilon+\omega)}{T}$$
$$\times\int\frac{d^3\mathbf{q}}{(2\pi)^3}\left\{2v_F{}^2\tau^2\,\mathrm{Im}\frac{V^R(\mathbf{q},\omega)}{[-i\omega+Dq^2-(T/T_0)\omega]^2}\right.$$
$$\left.-4D^2\tau^2q^2\,\mathrm{Im}\frac{V^R(\mathbf{q},\omega)}{[-i\omega+Dq^2-(T/T_0)\omega]^3}\right\}. \qquad (2.14)$$

At low temperatures ($T\ll T_0$) we obtain from Eq. (2.14)

$$\delta k(T)\approx\frac{5\cdot 2^{1/2}T^{3/2}}{36\pi^2D^{1/2}}\left\{1+\left(\frac{T}{T_0}\right)^2\right\}^{-1/4}\Gamma\left(\frac{5}{2}\right)\xi\left(\frac{5}{2}\right). \qquad (2.15a)$$

At intermediate temperatures $T_0\lesssim T\lesssim 10T_0$ we have, in the long-wavelength limit ($\omega\sim Dq^2$), when $(T/T_0)\omega\sim D\varkappa^2$,

$$\delta k(T)\approx\frac{1{,}83\cdot 2^{5/2}\pi\varkappa^2D^{1/2}}{24}\Gamma\left(\frac{1}{2}\right)T^{1/2} \qquad (2.15b)$$

and, finally, for $T\gg T_0$ we easily obtain from Eq. (2.14)

$$\delta k(T)\approx\frac{1{,}83\pi\varkappa^2D^{1/2}}{12}\Gamma\left(\frac{1}{2}\right)\frac{T_0{}^{3/2}}{T}. \qquad (2.15c)$$

We have thus described three types of temperature dependence of the electronic contribution to the thermal conductivity: $\delta k(T)\propto T^{3/2}$ for $T\ll T_0$; $\delta k(T)\propto T^{1/2}$ for $T_0\lesssim T\lesssim 10T_0$; and, $\delta k(T)\propto T^{-1}$ for $T\gg T_0$. Estimates of the contributions (2.15a)–(2.15c) are made according to the formula

$$\delta k(T)\sim\begin{cases}\left(\dfrac{k_B}{\hbar}\right)^{3/2}k_B\dfrac{T^{3/2}}{D^{1/2}}, & T\ll T_0\,,\\ \left(\dfrac{k_B}{\hbar}\right)\varkappa^2(\hbar D)^{1/2}(k_BT)^{1/2}, & T_0\lesssim T\leqslant 10T_0\,,\\ \left(\dfrac{k_B}{\hbar}\right)\varkappa^2(\hbar D)^{1/2}\dfrac{T_0{}^{3/2}}{T}, & T\gg T_0\,.\end{cases}$$

Figure 5 shows the computed contribution $\delta k(T)$ for an amorphous alloy with $T_0\sim 1$ K, $D\sim 3$ cm^2/s, and $\varkappa\sim 10^8$ cm^{-1}. According to the figure, in the region $T\ll T_0$ the thermal conductivity increases sharply—by several orders of magnitude—with increasing temperature. Next, at intermediate temperatures $T_0\lesssim T\leqslant 10T_0$, $\delta k(T)$ changes by an amount within one order of magnitude. On the logarithmic scale usually employed in measurements of $\delta k(T)$ in amorphous systems, such a change is virtually unnoticeable. Further, for $T\gg T_0$ the electron component of $k(T)$ drops with increasing temperature, but in this temperature range the phonon mechanism of scattering becomes significant. Therefore, a phonon contribution, which increases with temperature, should appear in the temperature dependence $k(T)$.

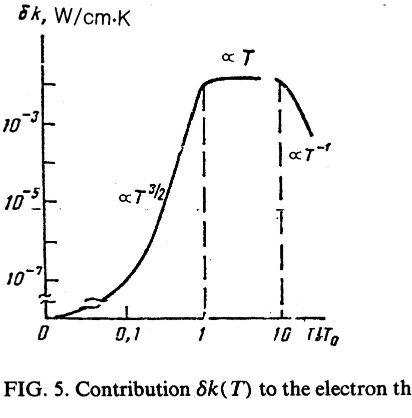

FIG. 5. Contribution $\delta k(T)$ to the electron thermal conductivity.

The changes, described here, in the temperature dependence of the thermal conductivity were observed experimentally. For example, Figs. 6a and 6b display data on the low-temperature thermal conductivity $k(T)$ for the amorphous alloy $Zr_{0.7}Pd_{0.3}$ (Ref. 8) and the metastable alloy $Zr_{0.8}Nb_{0.2}$ (Ref. 11). Comparison of our calculations with the experiments of Refs. 8 and 11 shows that not only is the agreement qualitative, but the quantitative description of the low-temperature thermal conductivity of the systems studied is also good.

It is curious to note that the result obtained here is not unexpected. As one can see from Eqs. (2.10) and (2.11), the temperature behavior of $\delta\kappa$ is largely determined by the corresponding dependence of the self-energy part $\Sigma_{ee}(\varepsilon,T)$ of the electronic Green's function. In Refs. 33 and 34 we calculated the temperature dependence of the effective relaxation time $\tilde{\tau}(T)$ of the Fermi electrons, which, as is well known, is inversely proportional to the imaginary part of $\Sigma_{ee}(\varepsilon,T)$ (Fig. 7). According to Fig. 7, $\tilde{\tau}(T)$ at low temperatures drops rapidly and, therefore, $\Sigma_{ee}(T)$ increases just as rapidly with increasing T—immediately by several orders of magnitude. $\Sigma_{ee}(T)$ increases next more gradually and saturates for $T\gg T_0$. However, $\delta k(T)$ repeats the change in $\Sigma_{ee}(T)$ at order-of-magnitude lower temperatures. As one can see from Eq. (2.11), δk is an integral function not only of $\Sigma_{ee}(\varepsilon,T)$ but also of the temperature Green's functions and the distribution functions. This is what causes the temperature interval where $\delta k(T)$ at first increases and then exhibits a plateau to shift.

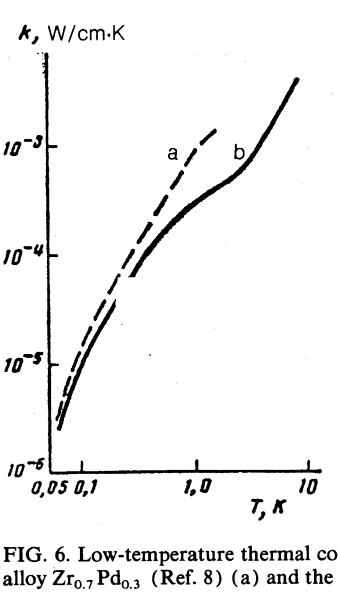

FIG. 6. Low-temperature thermal conductivity $k(T)$ of the amorphous alloy $Zr_{0.7}Pd_{0.3}$ (Ref. 8) (a) and the metastable alloy $Zr_{0.8}Nb_{0.2}$ (b).[11]

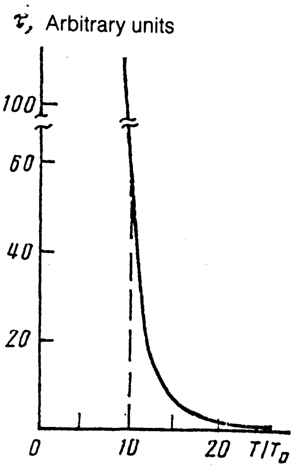

FIG. 7. Temperature dependence of the effective conduction electron relaxation time $\tilde{\tau}(T)$.[33,34]

We also note that the contribution, computed in this work, to the thermal conductivity (2.15) and the contribution made by individual scattering processes to the electric conductivity[31,32] and presented at the end of Sec. 1 are not related to one another by the Wiedeman-Franz law. This is obvious, for besides the multiple elastic scattering by DCE, we took into account inelastic electron–electron scattering.

Our final remark is that in the limit as $c \to 0$ Eq. (2.15a) is identical to the corresponding results of Ref. 26, which were obtained for impure metals. Thus as in the case of electric conductivity and electron heat capacity, the limiting transition from an amorphous binary alloy to a disordered impure metal is satisfied.

In conclusion it should be stated that in spite of the good qualitative and quantitative description of the low-temperature electron transport in metallic amorphous systems, the proposed mechanism for the formation of anomalies in the transport properties cannot be regarded as finally established. The results must be carefully checked experimentally. In particular, it is possible to determine experimentally whether or not $k(T)$ saturates in the intermediate temperature range $T_0 \lesssim T \lesssim 10T_0$ or the thermal conductivity changes with temperature within one order of magnitude. Further, if the proposed mechanism does exist, the temperature at which the phonon mechanism of scattering ($T_{\rm ph}$) comes into play can be determined from the length of the temperature "plateau" in $k(T)$. If $T_{\rm ph}$ is equal to T_0, then the curve $k(T)$ will not have a plateau, and $k(T)$ will be characterized only by an inflection point. It could also happen that $T_{\rm ph} > 10T_0$. Then $k(T)$ can be expected to exhibit four types of behavior: increasing ($T \ll T_0$), "plateau" ($T_0 \lesssim T \lesssim 10T_0$), decreasing ($10T_0 < T < T_{\rm ph}$), and once again increasing ($T > T_{\rm ph}$). Quite possibly this is what explains the temperature dependences $k(T)$, obtained in Refs. 39–41, characterized either by a point of inflection or decreasing $k(T)$ with increasing T at intermediate temperatures.

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