Journal of Experimental and Theoretical Physics, Vol. 96, No. 6, 2003, pp. 1078–1088.
Translated from Zhurnal Éksperimental'noĭ i Teoreticheskoĭ Fiziki, Vol. 123, No. 6, 2003, pp. 1227–1238.
Original Russian Text Copyright © 2003 by I. G. Kuleev, I. I. Kuleev, Taldenkov, Inyushkin, Ozhogin, Itoh, Haller.

SOLIDS
Structure

Normal Processes of Phonon–Phonon Scattering and the Drag Thermopower in Germanium Crystals with Isotopic Disorder

I. G. Kuleev[a],*, I. I. Kuleev[a], A. N. Taldenkov[b], A. V. Inyushkin[b], V. I. Ozhogin[b], K. M. Itoh[c], and E. E. Haller[d]

[a] *Institute of Physics of Metals, Ural Division, Russian Academy of Sciences,*
Yekaterinburg, 620219 Russia

**e-mail: kuleev@imp.uran.ru*

[b] *Institute of Molecular Physics, Russian Research Center Kurchatov Institute, Moscow, 123182 Russia*

[c] *Department of Applied Physics and Physico-Informatics, Keio University,*
Yokohama 223-8522, Japan

[d] *University of California and Lawrence Berkeley National Laboratory,*
Berkeley, CA, 94720, USA

Received December 10, 2002

Abstract—A strong dependence of the thermopower of germanium crystals on the isotopic composition is experimentally found. The theory of phonon drag of electrons in semiconductors with nondegenerate statistics of current carriers is developed, which takes into account the special features of the relaxation of phonon momentum in the normal processes of phonon–phonon scattering. The effect of the drift motion of phonons on the drag thermopower in germanium crystals of different isotopic compositions is analyzed for two options of relaxation of phonon momentum in the normal processes of phonon scattering. The phonon relaxation times determined from the data on the thermal conductivity of germanium are used in calculating the thermopower. The importance of the inelasticity of electron–phonon scattering in the drag thermopower in semiconductors is analyzed. A qualitative explanation of the isotope effect in the drag thermopower is provided. It is demonstrated that this effect is associated with the drift motion of phonons, which turns out to be very sensitive to isotopic disorder in germanium crystals.

1. INTRODUCTION

Recently, high-quality single crystals of germanium of different chemical compositions have been successfully grown, including a uniquely pure (both chemically and isotopically) crystal with 99.99% ^{70}Ge isotopic enrichment [1], hereinafter referred to as ^{70}Ge (99.99%). Experimental investigations of the thermal conductivity [2, 3] of these crystals have revealed that, for monoisotopic samples of ^{70}Ge (99.99%), the maximal values of thermal conductivity are an order of magnitude higher than those for crystals with natural isotopic composition. It is evident that this effect is associated with the increase in the free path of thermal phonons because of the decrease in the scattering by "impurity" isotopes; the normal processes of phonon–phonon scattering play an important part in the case of isotopically pure crystals at temperatures in the vicinity of the maximum of thermal conductivity [3–5]. A variation in the isotopic composition must also affect the thermoelectric phenomenon of the phonon drag thermopower $\alpha_{\mathrm{ph}}(T)$, which explicitly depends on phonon lifetime. Therefore, a decrease in the degree of isotopic disorder must further result in an increase in the absolute values of the phonon drag thermopower. However, the Herring theory [6] predicts a very weak dependence of $\alpha_{\mathrm{ph}}(T)$ on the impurity concentration in the case of a fairly pure semiconductor (see also [7–9]). Within a standard one-parameter approximation, the phonon relaxation rate in the normal processes (N processes) of phonon–phonon scattering was included in the total phonon relaxation rate, which was the only parameter defining the nonequilibrium phonon distribution function. This approach is justified in the case of "impure" semiconductors, when the phonon relaxation rate in the N processes, $\nu_{\mathrm{ph}N}(q)$, is much lower than the phonon relaxation rate in the resistive processes of scattering, $\nu_{\mathrm{ph}R}(q)$, caused by the phonon–phonon scattering in the umklapp processes, and from the defects and boundaries of the sample. In the opposite extreme case of fairly pure semiconductors, one must take into account the phonon system drift caused by the N processes of phonon–phonon scattering [10, 11].

In nondegenerate conductors, the electrons interact only with long-wavelength phonons whose wave vector is much less than the wave vector of thermal phonons making the main contribution to thermal conductivity. Because the probability of isotopic scattering of a phonon is proportional to the fourth power of its wave vector q, the thermopower calculated within a one-parameter approximation turns out to be insensitive to the degree of isotopic disorder. Kozlov and Nagaev [12] called attention to the anomalies of thermopower arising in such a situation as long as 30 years ago. They have demonstrated that, in the case of very perfect crystals, the thermal phonon drag of long-wavelength phonons may cause anomalously high values of thermopower. In contrast to the Herring thermopower, this thermopower (two-stage drag thermopower) is inversely proportional to the impurity concentration [13] and is closely associated with the mechanism of relaxation of long-wavelength phonons from thermal phonons in the normal processes of phonon–phonon scattering.

The first attempt at detecting the effect of isotopic phonon scattering on the thermopower was made by Oskotskii *et al.* [14], who investigated the thermal conductivity and thermopower of Te crystals with two different isotopic compositions, of which one was subjected to 92% ^{128}Te isotopic enrichment. The isotopic enrichment resulted in a threefold increase in the maximal values of thermal conductivity; however, Oskotskii *et al.* [14] observed no effect of isotopic disorder on the phonon drag thermopower at low temperatures. This negative result is possibly due either to the different concentrations of charged impurities in the investigated samples or to the relatively weak contribution of the N processes to the overall phonon relaxation rate.

In recent measurements of the thermopower in germanium crystals of different isotopic compositions, we found an almost twofold increase in the thermopower at low temperatures in a monoisotopic sample of ^{70}Ge (99.99%) compared to Ge of natural isotopic composition [15]. This result is indicative of the important part played by the N processes in the relaxation of the phonon system for isotopically enriched germanium crystals. The importance of these processes in the lattice thermal conductivity without the separation of the contributions made by longitudinal and transverse phonons is studied quite well [16–18]. In the N processes of scattering, the phonon momentum is conserved. These processes make no direct contribution to the thermal resistance; they provide for the relaxation of the phonon subsystem to the drift locally equilibrium distribution. Therefore, the N processes redistribute the energy and momentum between different phonon modes to form the nonequilibrium phonon distribution function and prevent a strong deviation of each phonon mode from the equilibrium distribution. This is accompanied by a variation of the relative contribution by various resistive processes of scattering (scattering from defects and boundaries of the sample and in the phonon–phonon umklapp processes) to thermal resistance. The drift motion of phonons must be taken into account under conditions in which the phonon relaxation rate in the N processes, $\nu_{\mathrm{ph}N}(q)$, is higher than or comparable to the rate of relaxation in resistive processes of scattering, $\nu_{\mathrm{ph}R}(q)$. It is evident that, in isotopically pure Ge samples at low temperatures, when the phonon–phonon umklapp processes are largely frozen, the rate of relaxation of longitudinal phonons in the N processes significantly exceeds the resistive rate of relaxation $\nu_{\mathrm{ph}R}(q)$ which is mainly due to isotopic disorder. In this paper, we will demonstrate that the inclusion of the phonon drift caused by the N processes enables one to qualitatively explain the significant effect of isotopic disorder on the drag thermopower in Ge crystals.

In describing the drag thermopower, in contrast to the previous investigations, we will separate the contributions by longitudinal and transverse phonons and take into account the redistribution of the phonon momentum in the N processes of scattering both within each vibrational branch (Simons mechanism [19]) and between different vibrational branches of phonons (Herring mechanism [20]). In this approximation, the nonequilibrium of the phonon system is described by six parameters, namely, by the rates of phonon relaxation in the resistive and normal processes of scattering and by the average drift velocities for each branch of the phonon spectrum. This description of phonon nonequilibrium enables one to reveal new features of relaxation of the momentum of quasi-particles and their effect on the thermopower and thermal conductivity of semiconductors. We will demonstrate below that the drift velocity of phonons (as well as the thermal conductivity) is largely defined by thermal phonons for which the scattering from defects plays a significant part. Therefore, when the drift of the phonon system is taken into account, the thermopower becomes sensitive to the degree of isotopic disorder. We further give the results of measurements and quantitative analysis of the isotope effect in the drag thermopower.

2. EXPERIMENTAL RESULTS

In this paper, we analyze the experimental data on the thermopower $\alpha(T)$ of single crystals of germanium with three different isotopic compositions, namely, the natural composition and compositions subjected to ^{70}Ge isotopic enrichment of 96.3% and up to 99.99%. Ge crystals of the n and p types with the concentration of charged impurities of $|N_d - N_a| < 2 \times 10^{13}$ cm^{-3} were used. Note that Geballe and Hull [21] found that, in the case of highly pure samples of Ge of the n and p types, the phonon drag thermopower very weakly depends on the concentration of electrically active impurities at a doping level below 10^{15} cm^{-3} and decreases in magnitude at higher concentrations. Our samples were shaped as parallelepipeds of square cross section. The samples

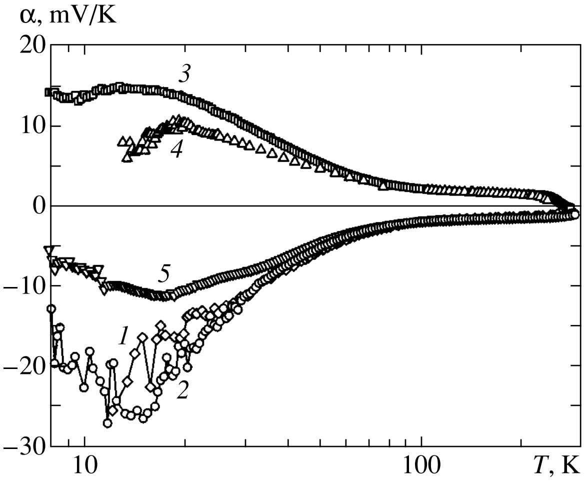

Fig. 1. The magnitude of differential thermopower as a function of temperature for samples of germanium crystals with different isotopic compositions: (*1*) sample no. G2, (*2*) G7, (*3*) G70, (*4*) Gn21, (*5*) S1.

had a total length of approximately 40 mm, with the square side in cross section of approximately 2.5 mm. The thermopower was measured using the method of steady longitudinal heat flux in vacuum in the temperature range from 8 to 300 K. The heat flux was directed along the longer axis of the sample; the temperature difference along the sample did not exceed 1% of its average temperature. The parameters of five investigated samples are given in the table.

The experimental data on the temperature dependence of the thermopower are given in Fig. 1. One can see in the figure that, at temperatures above 70 K, the thermopower is almost independent of temperature. The diffusion component of thermopower $\alpha_e(T)$ predominates in this temperature range; this component is defined by the degree of doping and by the band parameters of the semiconductor and is independent of the phonon lifetime. At low temperatures, where the phonon drag thermopower $\alpha_{ph}(T)$ predominates, $\alpha(T)$ increases with decreasing isotopic disorder; in so doing, the thermopower at the maximum for isotopically pure ^{70}Ge (99.99%) is approximately twice as high as that for germanium with the natural isotopic composition (^{nat}Ge). Compared to the thermal conductivity, the thermopower of germanium turned out to be approximately five times less sensitive to the variation of the degree of isotopic disorder. Note that, in the case of samples with the same isotopic composition, the thermopower is independent of the degree of doping within the experimental error. This is in good agreement with the well-known fact of the weak sensitivity of the magnitude of the drag thermopower to the dopant concentration in fairly pure germanium crystals [6, 21]. These special features of thermopower call for detailed theoretical treatment.

Given below are the results of quantitative analysis of the isotope effect in the thermopower of germanium. Attention is focused on the investigation of the effect of the drift motion of the phonon system, due to the normal processes of phonon scattering, and of the inelasticity of electron–phonon scattering on the drag thermopower. The effect of the normal processes of phonon scattering on the mutual drag of electrons and phonons in metals and in degenerate semiconductors is treated in [10, 11]. In our paper, this theory is generalized to the case of semiconductors with nondegenerate statistics of current carriers. We have treated the redistribution of the momentum of longitudinal and transverse phonons in the *N* processes of scattering both within each vibrational branch and between different vibrational branches. Previously, this approach made it possible to successfully explain the effect of the isotopic composition on the thermal conductivity of germanium and silicon crystals [22, 23]. Here, this method is used to investigate the effect of isotopic disorder on the drag thermopower. In calculating the emf, we used the times of phonon relaxation determined from the data on the thermal conductivity for the same samples of germanium [3, 22]. It is demonstrated that, in fairly pure semiconductors, both the thermopower and the lattice thermal conductivity [22] (with the separation of the contributions by longitudinal and transverse phonons)

Parameters of investigated samples of Ge crystals

Sample no.	Isotopic composition, % ^{70}Ge	g, 10^{-5}	Axis	$\|N_d - N_a\|$, 10^{12} cm^{-3}
G2	99.99	0.008	[100]	2.7
G7	99.99	0.008	[111]	20
G70	96.6	7.75	[100]	2
Gn21	natural	58.9	[100]	0.5
S1	natural	58.9	[111]	4

Note: $g = \sum_i f_i\left(\frac{M_i - \bar{M}}{\bar{M}}\right)^2$ is the factor characterizing the isotopic disorder of the crystal [3].

largely depend on the mechanism of relaxation of the phonon momentum in the N processes of scattering.

3. THE EFFECT OF THE N PROCESSES OF PHONON–PHONON SCATTERING ON THE RELAXATION OF MOMENTUM OF ELECTRONS AND PHONONS IN A NONEQUILIBRIUM ELECTRON–PHONON SYSTEM

For simplicity, we will treat a semiconductor with the isotropic law of dispersion of current carriers. We will calculate a charge flow caused by the effect of electric field $\mathbf{E} = \{E_x, 0, 0\}$ and the temperature gradient $\nabla T = (\nabla_x T, 0, 0)$. The set of kinetic equations for the nonequilibrium electron $f(\mathbf{k}, \mathbf{r})$ and phonon $N^\lambda(\mathbf{q}, \mathbf{r})$ distribution functions in view of the N processes of scattering has the form [11]

$$\begin{gathered}\frac{e}{\hbar}\mathbf{E}_0\cdot\frac{\partial f_\mathbf{k}}{\partial\mathbf{k}} + (\mathbf{v}_k\cdot\nabla_r)f_\mathbf{k} = I_{ei}(f_\mathbf{k}) + I_{e\,\mathrm{ph}}(f_\mathbf{k}, N_q^\lambda),\\ \mathbf{v}_q^\lambda\cdot\nabla_r N_q^\lambda = -(N_q^\lambda - N_{q\lambda}^{(0)})\nu_{\mathrm{ph}}^{(1)\lambda}\\ -(N_q^\lambda - N(\mathbf{q}\cdot\mathbf{u}_\lambda))\nu_{\mathrm{ph}N}^\lambda + I_{\mathrm{ph}\,e}(N_q^\lambda, f_\mathbf{k}).\end{gathered} \tag{1}$$

Here, $\mathbf{v}_q^\lambda = s_\lambda\mathbf{q}/q$ is the group velocity of acoustic phonons with polarization λ; $N_{q\lambda}^0$ is the Planck function; $\nu_{\mathrm{ph}N}^\lambda(q)$ is the phonon relaxation rate in the N processes of scattering; and the rate

$$\nu_{\mathrm{ph}}^{(1)\lambda}(q) = \nu_{\mathrm{ph}i}^\lambda(q) + \nu_{\mathrm{ph}B}^\lambda(q) + \nu_{\mathrm{ph}U}^\lambda(q)$$

includes all of the nonelectron resistive rates of phonon relaxation, due to the phonon scattering from phonons in the umklapp processes, $\nu_{\mathrm{ph}U}^\lambda(q)$, from defects and isotopic disorder, $\nu_{\mathrm{ph}i}^\lambda(q)$, and from the boundaries of the sample, $\nu_{\mathrm{ph}B}^\lambda(q)$. The collision integrals of electrons with impurities, I_{ei}, and with phonons, $I_{e\,\mathrm{ph}}$, and of phonons with electrons, $I_{\mathrm{ph}\,e}$, were determined in [7–9, 24–27]. In Eq. (1), it is taken into account that the N processes of scattering bring the phonon subsystem to the locally equilibrium Planck distribution with the drift velocity $\mathbf{u}_\lambda$ which may be different for phonons of different polarizations [16–18],

$$\begin{gathered}N(\mathbf{q}, \mathbf{u}_\lambda) = \left[\exp\left(\frac{\hbar\omega_{q\lambda} - \hbar\mathbf{q}\cdot\mathbf{u}_\lambda}{k_BT}\right) - 1\right]^{-1}\\ \approx N_{q\lambda}^0 + \frac{\hbar\mathbf{q}\cdot\mathbf{u}_\lambda}{k_BT}N_{q\lambda}^0(N_{q\lambda}^0 + 1).\end{gathered} \tag{2}$$

We will represent the electron and phonon distribution functions in the form [3–5]

$$\begin{gathered}f_\mathbf{k} = f_0(\varepsilon_k) + \delta f_\mathbf{k}, \quad \delta f_\mathbf{k} = -\frac{\partial f_0}{\partial\varepsilon_k}\mathbf{v}_k\cdot\boldsymbol{\chi}(\varepsilon),\\ N_\mathbf{q}^\lambda = N_{q\lambda}^0 + g_\lambda(\mathbf{q}),\end{gathered} \tag{3}$$

where $f_0(\varepsilon_k)$ is the equilibrium electron distribution function, and $\delta f_\mathbf{k}$ and $g_\lambda(\mathbf{q})$ are nonequilibrium additions to the distribution functions, which are linear as regards the external effects. We will linearize the collision integrals with respect to these additions. We will represent the collision integrals $I_{ie}(\delta f_\mathbf{k})$ and $I_{\mathrm{ph}\,e}(f_0, g_\lambda(\mathbf{q}))$, as well as $I_{e\,\mathrm{ph}}(\delta f_\mathbf{k}, N_{q\lambda}^0)$ in the approximation of elastic scattering, in terms of relaxation rates [27]. In calculating the collision integral $I_{\mathrm{ph}\,e}(f_0, g_\lambda(\mathbf{q}))$, we will not restrict ourselves to the linear approximation with respect to the inelasticity parameter [7–9, 24–28] and will take into account the inelasticity of collisions between nonequilibrium phonons and equilibrium electrons.

We will substitute expressions (2) and (3) into (1) to derive, similar to [11], the expression for the phonon distribution function $g_\lambda(\mathbf{q})$,

$$\begin{gathered}g_\lambda(\mathbf{q}) = -\frac{N_{q\lambda}^0(N_{q\lambda}^0 + 1)}{\nu_{\mathrm{ph}}^\lambda(q)}\frac{\hbar\omega_{q\lambda}}{k_BT^2}\mathbf{v}_q^\lambda\cdot\nabla T\\ + \frac{\hbar\mathbf{q}\cdot\mathbf{u}_\lambda}{k_BT}N_{q\lambda}^0(N_{q\lambda}^0 + 1)\frac{\nu_{\mathrm{ph}N}^\lambda(q)}{\nu_{\mathrm{ph}}^\lambda(q)}.\end{gathered} \tag{4}$$

Here,

$$\begin{aligned}\nu_{\mathrm{ph}}^\lambda(q) &= \nu_{\mathrm{ph}}^{(1)\lambda}(q) + \nu_{\mathrm{ph}\,e}^\lambda(q) + \nu_{\mathrm{ph}N}^\lambda(q)\\ &= \nu_{\mathrm{ph}N}^\lambda(q) + \nu_{\mathrm{ph}R}^\lambda(q)\end{aligned}$$

is the total rate of relaxation of phonons with the wave vector q and polarization λ, and $\nu_{\mathrm{ph}\,e}^\lambda(q)$ is the rate of relaxation of momentum of phonons from electrons [24–27]. The first term in expression (4) is defined by the diffusion motion of phonons, and the second term takes into account the drift phonon motion and is associated with the normal processes of phonon–phonon scattering. The phonon drift velocity $\mathbf{u}_\lambda$ is found from the balance equation for phonon momentum, which follows from the law of conservation of momentum in the normal processes of phonon–phonon scattering,

$$\begin{gathered}\frac{1}{V}\sum_{q,\lambda}\hbar\mathbf{q}\nu_{\mathrm{ph}N}^\lambda(q)(N_q^\lambda - N(\mathbf{q}, \mathbf{u}_\lambda)) = \frac{1}{V}\sum_{q,\lambda}\hbar\mathbf{q}\nu_{\mathrm{ph}N}^\lambda(q)\\ \times\left[g_\lambda(\mathbf{q}) - \frac{\hbar\mathbf{q}\cdot\mathbf{u}_\lambda}{k_BT}N_{q\lambda}^0(N_{q\lambda}^0 + 1)\right] = 0.\end{gathered} \tag{5}$$

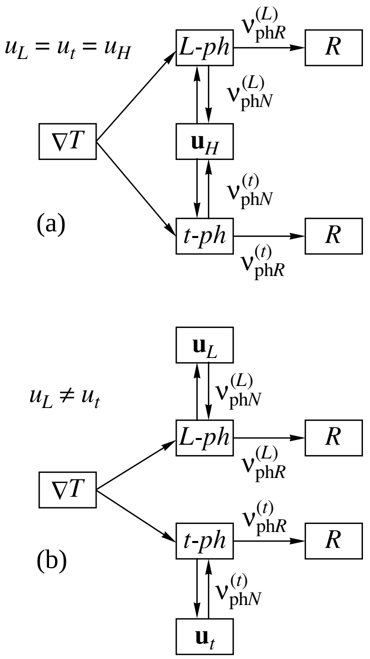

Fig. 2. A scheme illustrating the relaxation of momentum in a phonon system for two mechanisms of phonon scattering in the normal processes: (a) for the Herring mechanism, (b) for the Simons mechanism.

A scheme illustrating the redistribution and relaxation of the momentum received by a phonon system from the temperature gradient is given in Fig. 2. The phonon scattering in the resistive processes of scattering (R) (from isotopic disorder, electrons, and sample boundaries, and the phonon–phonon scattering in the umklapp processes) brings about the relaxation of the momentum of the phonon system. The N processes redistribute the momentum between different phonon modes (*L-ph* and *t-ph*) and bring about the phonon drift with an average velocity $\mathbf{u}_\lambda$. As in [10, 11, 16–18], we assume that the drift velocity is independent of the wave vector of phonons. Two mechanisms of normal three-phonon processes of scattering are usually examined, namely, the Herring [20] and Simons [19] mechanisms. In the Herring mechanism of N processes, phonons of different polarizations are involved: the rate of relaxation of transverse phonons in the Herring mechanism is defined by the scattering processes ($t + L \longleftrightarrow L$) in which one transverse and two longitudinal phonons are involved; in this case, the main contribution to the rate of relaxation of longitudinal phonons is made either by the processes of decay of a longitudinal phonon into two transverse phonons belonging to different branches or by the fusion of two transverse phonons to form a longitudinal phonon ($L \longleftrightarrow t_1 + t_2$). This relaxation mechanism provides for redistribution of the drift momentum between longitudinal and transverse phonons (see Fig. 2) and tends to establish a locally equilibrium distribution with a drift velocity that is the same for phonons of both polarizations, $u_L = u_t = u_H$. The Simons mechanism of relaxation [19] involves phonons of one polarization. In the case of this mechanism of scattering, the law of conservation of momentum in the N processes is valid for each branch of the phonon spectrum, and the drift velocity of longitudinal phonons differs from that of transverse phonons. Therefore, we will treat below two options for the relaxation of the phonon momentum in the N processes.

We use expressions (4) and the balance equation for phonon momentum (5) to find the phonon drift velocity $\mathbf{u}_\lambda$ for the Herring (H) and Simons (S) mechanisms of relaxation, as was done in [11]. After this, we derive, for the phonon distribution function $g_\lambda(q)$,

$$g_\lambda(\mathbf{q}) = -\frac{N_{q\lambda}^0(N_{q\lambda}^0+1)}{\tilde{\nu}_{ph}^\lambda(q)}\frac{\hbar\omega_{q\lambda}}{k_BT^2}\mathbf{v}_q^\lambda\cdot\nabla T,$$
$$\tilde{\nu}_{ph}^{\lambda(S,\,H)}(q) = \tilde{\nu}_{ph}^\lambda(q)(1+\tilde{\nu}_{phN}^\lambda(q)\beta_{(S,\,H)})^{-1}, \qquad (6)$$

$$\beta_S = \frac{\Psi_N^\lambda}{\Psi_{NR}^\lambda}, \quad \beta_H = \left(\frac{s_L}{s_\lambda}\right)^2\frac{\Psi_N^L+2S_*^2\Psi_N^t}{\Psi_{NR}^L+2S_*^5\Psi_{NR}^t}. \qquad (7)$$

Here, $S_* = s_L/s_t$, and the remaining functions are defined by the expressions

$$\Psi_N^\lambda = \left\langle\frac{\nu_{phN}^\lambda(q)}{\nu_{ph}^\lambda(q)}\right\rangle_{z_{d\lambda}}$$
$$\equiv\int_0^{z_{d\lambda}}dz_q^\lambda(z_q^\lambda)^4\frac{\nu_{phN}^\lambda(q)}{\nu_{ph}^\lambda(q)}N_{q\lambda}^0(N_{q\lambda}^0+1), \qquad (8)$$
$$\Psi_{NR}^\lambda = \left\langle\frac{\nu_{phR}^\lambda(q)\nu_{phN}^\lambda(q)}{\nu_{ph}^\lambda(q)}\right\rangle_{z_{d\lambda}},$$

where

$$z_q^\lambda = \frac{\hbar\omega_{q\lambda}}{k_BT} = \frac{q}{q_{T\lambda}}, \quad q_{T\lambda} = \frac{k_BT}{\hbar s_\lambda},$$

$$z_{2k}^\lambda = \frac{2k}{q_{T\lambda}}, \quad z_{d\lambda} = \frac{\hbar\omega_{d\lambda}}{k_BT}$$

($\omega_{d\lambda}$ is the Debye frequency of phonons with polarization λ). One can see in expression (6) that the inclusion of the normal processes of phonon–phonon scattering reduces to the renormalization of the rate of relaxation of phonon momentum. The phonon drift motion caused by the N processes leads to a decrease in the effective phonon relaxation rate; this decrease is different for the Herring [20] and Simons [19] relaxation mechanisms.

We will now consider the electron subsystem. The electron scattering from impurities, ν_{ei}, results in the relaxation of electron momentum, and the mechanisms of electron–phonon relaxation characterized by the rates $\nu_{e\,\mathrm{ph}}$ and $\nu_{\mathrm{ph}\,e}$ bring about the redistribution of momentum within an electron–phonon system; in so doing, the electrons interact only with long-wavelength phonons. We do not treat the renormalization of the thermopower due to the mutual drag of electrons and phonons. Note that the quantities such as the thermopower and thermal conductivity are found from the condition that the total current through the sample is zero. In this case, the average velocity of ordered electron motion in any physically small sample volume is zero. Therefore, the transfer of momentum of ordered electron motion to the phonon subsystem is low, and the effect of electron nonequilibrium on the electrons via the phonon subsystem may be ignored [11]. On the other hand, a steady phonon flow from the hot end of the sample to the cold end exists in the presence of a temperature gradient, and the magnitude of thermopower is largely defined by the transfer of momentum of ordered phonon motion to electrons. Note that, for longitudinal phonons in Ge crystals at low temperatures, when the electron–phonon drag makes a marked contribution to the thermopower, the relaxation rate $\nu_{\mathrm{ph}N}(q) \gg \nu_{\mathrm{ph}R}(q)$ [22]. It follows from the foregoing that the relaxation of phonon momentum in a nonequilibrium electron–phonon system must be taken into account more rigorously than was done in the case of one-parameter approximation [7–9, 24–28].

The purpose of this theoretical analysis is to investigate the effect of the phonon drift caused by the N processes on the drag thermopower. In this case, one can ignore the mutual drag of electrons and phonons and obtain, as was done in [27], the following solution for the function $\boldsymbol{\chi}(\varepsilon)$:

$$\boldsymbol{\chi}(\varepsilon) = -e\tau(\varepsilon)\left[\mathbf{E} + \frac{k_B}{e}\left(\tilde{A}_{\mathrm{ph}}(\varepsilon) + \frac{\varepsilon-\zeta}{k_BT}\right)\nabla T\right], \qquad (9)$$

$$\tilde{A}_{\mathrm{ph}}(\varepsilon) = \sum_\lambda \frac{ms_\lambda^2}{k_BT}\int_0^{z_{2k}^\lambda} dz_q^\lambda \frac{\nu_{e\,\mathrm{ph}}^\lambda(k,q)}{\tilde{\nu}_{\mathrm{ph}}^\lambda(q)}. \qquad (10)$$

Here, $\tau(\varepsilon)$ is the total relaxation time of electrons;

$$\tau^{-1}(\varepsilon_k) = \nu_e(k) = \nu_{ei}(k) + \nu_{e0}(k) + \nu_{e\,\mathrm{ph}}(k);$$

the rates of electron relaxation from neutral, $\nu_{e0}(k)$, and charged, $\nu_{ei}(k)$, impurities have the known form (see, for example, [7], formulas (10.29) and (10.50)); and the electron–phonon collision rate is defined by the expression

$$\nu_{e\,\mathrm{ph}}^\lambda(k) = \sum_\lambda \langle \nu_{e\,\mathrm{ph}}^\lambda(k,q)\rangle_{z_{2k}^\lambda},$$

$$\nu_{e\,\mathrm{ph}}^\lambda(k,q) = \frac{m_e|C_{0\lambda}|^2}{2\pi\hbar^2k^3}q^5N_{q\lambda}^0(N_{q\lambda}^0+1)$$
$$\times\left\{\left(1-\frac{2ms_\lambda}{\hbar q}\right)F^+(\varepsilon_k) + \left(1+\frac{2ms_\lambda}{\hbar q}\right)F^-(\varepsilon_k)\right\}, \qquad (11)$$

where

$$F^\pm(\varepsilon_k) = \pm\frac{f_0(\varepsilon_k)-f_0(\varepsilon_k\pm\hbar\omega_q)}{f_0(\varepsilon_k)(1-f_0(\varepsilon_k))}, \quad |C_{0\lambda}|^2 = \frac{E_{0\lambda}^2\hbar}{s_\lambda\rho},$$

$E_{0\lambda}$ is the deformation potential constant, and ρ is the density. For semiconductors with nondegenerate statistics of current carriers,

$$f_0(\varepsilon) \approx \exp\left(-\frac{\varepsilon-\zeta}{k_BT}\right),$$

the $F^\pm$ functions may be represented in the form

$$F^+ \approx 1-e^{-z}, \quad F^- \approx e^z - 1;$$

then, in view of the inelasticity of electron–phonon scattering, we find

$$(\tau_{e\,\mathrm{ph}}(\varepsilon))^{-1} = \sum_\lambda (\tau_{0e\,\mathrm{ph}}^\lambda)^{-1}x^{-3/2}J_{1\lambda}(x),$$

$$J_{1\lambda}(x) = \frac{1}{2}(J_{1\lambda}^+(x) + J_{1\lambda}^-(x)),$$

$$J_{1\lambda}^\pm(x) = \int_0^{z_{\mathrm{max},\lambda}^\pm} J_{1\lambda}^\pm(z)dz, \qquad (12)$$

$$J_{1\lambda}^+(z) = \frac{z^3(z-\delta_\lambda)}{e^z-1},$$

$$J_{1\lambda}^-(z) = \frac{z^3(z+\delta_\lambda)}{e^z-1}e^{-z},$$

$$(\tau_{0e\,\mathrm{ph}}^\lambda)^{-1} = \frac{m_e|C_{0\lambda}|^2}{2\pi\hbar^3}q_{T\lambda}^3\delta_\lambda^{-3/2}.$$

The upper limits of integration in Eqs. (12) are defined by the expressions

$$z^{\pm}_{\max,\lambda} = \frac{2\hbar s_\lambda k}{k_B T} = 2\delta_\lambda^{1/2}x^{1/2} \pm \delta_\lambda,$$
$$\delta_\lambda = \frac{2m_e s_\lambda^2}{k_B T} = \frac{T_{s\lambda}}{T}, \qquad (13)$$
$$T_{s\lambda} = \frac{2m_e s_\lambda^2}{k_B}, \quad x = \frac{\varepsilon}{k_0 T},$$

where δ_λ is the inelasticity parameter. In the case of semiconductors, the effective temperature $T_{s\lambda}$ defining the inelasticity of electron–phonon scattering is, as a rule, less than 1 K; for example, in the case of Ge, $T_{sL} \approx 0.8$ K at $s_L \approx 5.21 \times 10^5$ cm/s, $m_e \approx 0.22m_0$. Therefore, even at $T > 10$ K, $\delta_\lambda \ll 1$ and $z_{\max} \ll 1$; therefore, expressions (12) may be expanded in powers of z. In a zero approximation with respect to the inelasticity parameter δ_λ, we will derive from Eqs. (12) the known expression for the electron–phonon relaxation time,

$$\tau^\lambda_{e\,\mathrm{ph}}(\varepsilon) = \tau^\lambda_{0e\,\mathrm{ph}}(2\delta_\lambda)^{-2}x^{-1/2}. \qquad (14)$$

In the same approximation, the expression for the inverse time of phonon–electron relaxation has the form

$$\nu^\lambda_{\mathrm{ph}\,e}(k, q) = \frac{m_e^2|C_{0\lambda}|^2 k_B T}{\pi\hbar^5}e^{\eta}(1-e^{-z})e^{-x_m},$$
$$x_m = \frac{(z-\delta_\lambda)^2}{4\delta_\lambda}. \qquad (15)$$

The concentrations of electrons, n_e, and of charged donors, N_{d+}, and the reduced Fermi level $\eta = \zeta/k_BT$ are found from the condition of electroneutrality for germanium (see [7], formula (6.9)): $N_d \approx 10^{12}$–10^{13} cm^{-3}, $\varepsilon_d \approx 0.01$ eV, and $m_e \approx 0.22m_0$. For these values of the parameters, the criterion of nondegenerate statistics is well valid.

In the approximation we adopted, the $\boldsymbol{\chi}(\varepsilon)$ function allows for the direct effect of the electric field and temperature gradient on the electron subsystem, as well as for the effect of the phonon drag of electrons.

4. THE DRAG THERMOPOWER IN SEMICONDUCTORS WITH NONDEGENERATE STATISTICS OF CURRENT CARRIERS

We will examine the effect of the normal processes of phonon–phonon scattering on the thermal electromotive force of semiconductors with nondegenerate statistics of current carriers. We will calculate the conduction current **j** by dividing it into three parts proportional to nonequilibrium additions to the electron distribution function $\boldsymbol{\chi}(\varepsilon)$,

$$\mathbf{j} = -\frac{e}{3\pi^2}\int_0^\infty d\varepsilon\left(-\frac{\partial f_0}{\partial \varepsilon}\right)\frac{k^3}{m(\varepsilon)}\boldsymbol{\chi}(\varepsilon)$$
$$\approx \mathbf{j}_{dr} + \mathbf{j}_{\mathrm{drag}} + \mathbf{j}_{\mathrm{dif}} = 0. \qquad (16)$$

From the condition **j** = 0, we find

$$\alpha = \alpha_{\mathrm{ph}} + \alpha_{\mathrm{dif}}. \qquad (17)$$

We will not consider below the diffusion component of thermopower: for germanium crystals at $T < 100$ K, this contribution is small. In the case of nondegenerate statistics, the expression for the phonon drag thermopower may be represented in the form

$$\alpha_{\mathrm{ph}} = -\frac{k_B}{e}\frac{\langle\langle \tau(\varepsilon)\tilde{A}_{\mathrm{ph}}(\varepsilon)\rangle\rangle}{\langle\langle \tau(\varepsilon)\rangle\rangle},$$
$$\langle\langle f(\varepsilon)\rangle\rangle = \frac{\int_0^\infty dx e^{-x}x^{3/2}f(\varepsilon)}{\int_0^\infty dx e^{-x}x^{3/2}}, \quad x = \frac{\varepsilon}{k_0 T}, \qquad (18)$$

$$\tilde{A}_{\mathrm{ph}}(\varepsilon) = \sum_\lambda \frac{m_e|C_{0\lambda}|^2}{4\pi\hbar^3}q_{T\lambda}^2\delta_\lambda^{-1/2}x^{-3/2}(J^+_{2\lambda}(x) + J^-_{2\lambda}(x)), \qquad (19)$$

$$J^{\pm}_{2\lambda}(x) = \int_0^{z^{\pm}_{\max,\lambda}} \frac{J^{\pm}_{1\lambda}(z)dz}{\tilde{\nu}^\lambda_{\mathrm{ph}}(q)},$$
$$J^+_{1\lambda}(z) = \frac{z^3(z-\delta_\lambda)}{e^z - 1}, \qquad (20)$$
$$J^-_{1\lambda}(z) = \frac{z^3(z+\delta_\lambda)}{e^z - 1}e^{-z}.$$

The upper integration limits in Eqs. (20) are defined by expressions (13). First of all, note that the drag thermopower includes, as does the lattice thermal conductivity [11, 22], the phonon momentum relaxation rate renormalized by the *N* processes. Unlike standard one-parameter approximations for the drag thermopower [7–9, 24–28], expressions (18)–(20) include the inelasticity of electron–phonon scattering, as well as the contribution made by the phonon drift motion. This contribution has different forms for the Herring and Simons mechanisms of relaxation. Because the phonon drift velocity is defined by all thermally excited phonons, the thermopower becomes sensitive to the degree of isotopic disorder. It follows from formulas (6), (19), and (20) that the inclusion of the drift of the phonon subsystem, which is associated with the normal processes of phonon scattering, brings about a decrease in the effective relaxation rate of phonons and, accordingly, an increase in the fraction of momentum transferred to electrons by phonons. This result is of practical importance as regards the interpretation of experimental data on the thermopower of germanium crystals with isotopic disorder.

In the extreme case of $\nu_{\mathrm{ph}N}(q) \ll \nu_{\mathrm{ph}R}(q)$, one can ignore the contribution of the phonon drift motion and use the expression for the drag thermopower that was previously derived with a one-parameter approximation [7–9, 24–28]. With $\nu_{\mathrm{ph}N}(q) \gg \nu_{\mathrm{ph}R}(q)$, the normal processes of phonon–phonon scattering and the drift of the phonon system associated with this scattering lead to a significant increase in the absolute values of thermopower. Note that, in interpreting the experimental data on the drag thermopower in previous studies involving the use of a one-parameter approximation (see [7–9, 24–28]), the relaxation rate in the normal processes $\nu_{\mathrm{ph}N}(q)$ was included in the total phonon relaxation rate $\nu_{\mathrm{ph}}^{\lambda}(q)$ as the resistive mechanism of phonon scattering, and, at $\nu_{\mathrm{ph}N}(q) \gg \nu_{\mathrm{ph}R}(q)$, it was treated as the only mechanism of relaxation of momentum of long-wavelength phonons [7, 8]. However, it follows from expressions (18)–(20) that, in this extreme case, the relaxation rate $\nu_{\mathrm{ph}N}(q)$ is eliminated from the drag thermopower, and α_{ph} is fully defined by the averaged relaxation rate of phonons in the resistive processes of scattering,

$$\alpha_{\mathrm{ph}}^{(S,\,H)} \approx -\frac{k_B}{e}\sum_{\lambda}\frac{m_F s_{\lambda}^2}{k_B T}\frac{\langle\langle\tau(\varepsilon)\nu_{e\,\mathrm{ph}}^{\lambda}(\varepsilon)\rangle\rangle}{\langle\nu_{\mathrm{ph}\,R}^{\lambda}(q)\rangle_{z_{d\lambda}}^{(S,\,H)}\langle\langle\tau(\varepsilon)\rangle\rangle},$$

$$\langle\nu_{\mathrm{ph}R}^{\lambda}(q)\rangle^{(S)} = \frac{\langle\nu_{\mathrm{ph}R}^{\lambda}\rangle}{J_{\lambda}^{(4)}},$$

$$\langle\nu_{\mathrm{ph}R}^{\lambda}(q)\rangle^{(H)} = \frac{\langle\nu_{\mathrm{ph}R}^{L}\rangle + 2S_*^5\langle\nu_{\mathrm{ph}R}^{t}\rangle}{J_L^{(4)} + 2S_*^3 J_t^{(4)}}, \tag{21}$$

$$J_{\lambda}^{(4)} = \int_0^{z_{d\lambda}}\frac{dz z^4 e^z}{(e^z-1)^2}.$$

A one-parameter approximation yields in this case an entirely different result,

$$\alpha_{\mathrm{ph}}^{(S,\,H)} \approx -\frac{k_B}{e}$$

$$\times\sum_{\lambda}\frac{m_F s_{\lambda}^2}{k_B T}\frac{1}{\langle\langle\tau(\varepsilon)\rangle\rangle}\left\langle\left\langle\tau(\varepsilon)\int_0^{z_{2k}^{\lambda}}dz_q^{\lambda}\frac{\nu_{e\,\mathrm{ph}}^{\lambda}(k,q)}{\nu_{\mathrm{ph}\,N}^{\lambda}(q)}\right\rangle\right\rangle. \tag{22}$$

Therefore, the inferences previously made with respect to the temperature and field dependences of the drag thermopower [7–9, 24–28] need to be refined. It is evident that, in the case of one-parameter approximation (22), the drag thermopower α_{ph} is insensitive to the degree of isotopic disorder. Note that a different approach to the calculation of the drag thermopower was suggested in [12, 13]. This approach is based on dividing the entire system into two subsystems, namely, the subsystem of long-wavelength phonons ($q < 2k$) with which electrons interact and the subsystem of thermal phonons ($q > q_{T\lambda}$). The authors of [12, 13] suggested a mechanism of two-stage drag: the phonon drift motion is defined by the thermal phonons, which, in turn, drag the long-wavelength phonons. By their physical content, our method and the approach developed in [12, 13] coincide, because, in our theory, it is the thermal phonons that define the phonon drift motion, as well as the thermal conductivity. However, our method is more general: we treat correctly the *N* processes of scattering of thermal phonons with regard for their drift and diffusion motion, identify the contributions by phonons of different polarizations, and treat both the intrabranch and interbranch redistribution of the phonon momentum in the *N* processes of scattering.

5. THE RESULTS OF CALCULATION OF THE DRAG THERMOPOWER OF GERMANIUM CRYSTALS OF DIFFERENT ISOTOPIC COMPOSITIONS

Given below are the results of numerical analysis of the drag thermopower in germanium crystals of different isotopic compositions, which, in view of the assumptions made, may only pretend to be a qualitative explanation of the effect. The main results include the isotropic band approximation and the assumption that the phonon drift velocity is independent of the wave vector, i.e., the drift velocities of thermal and long-wavelength phonons are the same. The calculation of the drag thermopower with a real band structure of germanium within the suggested method of inclusion of the normal processes of phonon scattering, with the long-wavelength and thermal phonons treated separately, is of interest *per se*. In this analysis, we restrict ourselves to examining the effect of the phonon drift motion and of the inelasticity of electron–phonon scattering on the drag thermopower in germanium crystals. The values of the parameters defining the phonon relaxation rate were borrowed from the results of analysis of the data on the thermal conductivity of Ge crystals of different isotopic compositions, obtained in [3, 22]. The use of these parameters made it possible to fit the results of calculations of thermal conductivity for the Herring mechanism of relaxation [22] to the experimental data of [3] in a wide temperature range in the entire investigated range of isotopic enrichment. In our calculation of the drag thermopower, these parameters are not varied. The fitting parameter of the theory is the

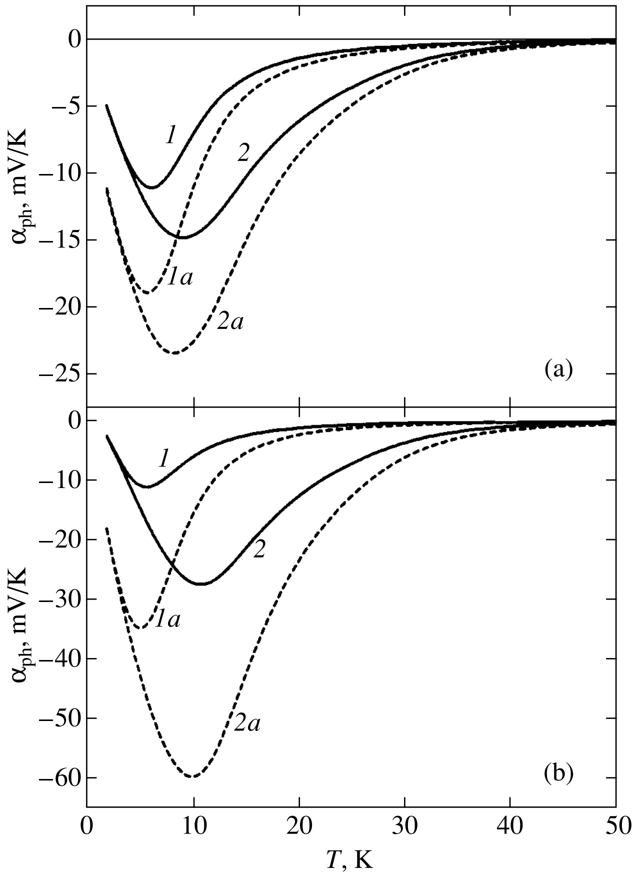

Fig. 3. The temperature dependence of the drag thermopower for the following values of parameters: (a) $m_e \approx 0.22m_0$, $E_{0L} = 16$ eV; (b) $m_e = m_0$, $E_{0L} = 4$ eV. Curves *1* and *1a* are for germanium of natural isotopic composition ($N_d = 4 \times 10^{12}$ cm^{-3}), and curves *2* and *2a* are for germanium with 99.99% ^{70}Ge ($N_d = 2 \times 10^{13}$ cm^{-3}). Curves *1* and *2* allow for the inelasticity of electron–phonon scattering, and curves *1a* and *2a* are plotted in a linear approximation with respect to the parameter of inelasticity of electron–phonon scattering.

deformation potential constant. Given a fixed effective mass of electrons, this constant is selected on the basis of the condition of agreement between the calculated value of absolute thermopower at the point of maximum and the experimentally obtained values for germanium of natural isotopic composition and is then used to calculate the thermopower of ^{70}Ge (99.99%). Because the effective mass of one of four ellipsoids in the crystallographic direction [111] is $m_e \approx 1.68m_0$, its average magnitude was varied from the value of the effective mass of the density of states $m_e \approx 0.22m_0$ to the value of $m_e \approx m_0$.

We will first examine the part played by the inelasticity during the transfer of momentum from nonequilibrium phonons to equilibrium electrons. Figures 3a and 3b give the results of calculations of the drag thermopower for $^{\text{nat}}$Ge and ^{70}Ge (99.99%) at $m_e \approx 0.22m_0$ and $m_e \approx m_0$. One can see in the figures that the exact inclusion of the inelasticity of electron–phonon scattering brings about a marked suppression of the contribution of phonon drag in the thermopower. The maximal values of thermopower $|\alpha_{\max}|$ decrease by a factor of 1.6–1.7 for the value of $m_e \approx 0.22m_0$. However, the importance of inelasticity increases with the effective mass of electrons: at $m_e \approx m_0$, the value of $|\alpha_{\max}|$ decreases by a factor of 3 for $^{\text{nat}}$Ge and by a factor of 2.2 for ^{70}Ge (99.99%). This result came as a surprise to us. The thing is that analysis of the time of relaxation of electrons from phonons [7–9] revealed that, for temperatures $T \gg T_{s\lambda}$ ($T_{sL} \approx 0.8$ K for Ge at $m_e \approx 0.22m_0$), the importance of inelasticity is minor and, at temperatures above 5 K, it may be ignored. Therefore, in the previously published papers dealing with the drag thermopower in semiconductors [6–9, 24–30], the inelasticity of electron–phonon scattering was taken into account in a linear approximation with respect to the inelasticity parameter $\hbar\omega_{q\lambda}/k_BT$.

Note that the inclusion of scattering from charged and neutral donor impurities at concentrations of the order of 10^{12}–10^{13} cm^{-3} has little effect on the magnitude of the thermopower (this scattering introduces a contribution of less than 3%), while the magnitude of the electron mobility varies more significantly in the low-temperature region.

Figure 4 gives the contributions of the phonon drift and diffusion motions into the drag thermopower for $^{\text{nat}}$Ge and ^{70}Ge (99.99%). One can see in the figure that, in the case of $^{\text{nat}}$Ge, the predominant contribution to the thermopower is made by the phonon diffusion motion. The contribution by the drift motion is small and amounts to 21% of the diffusion contribution at the maximum of $|\alpha|$. In contrast, in the case of ^{70}Ge (99.99%), the drift contribution to the drag thermopower predominates. It is six times the diffusion contribution. In view of the foregoing, indeed, the isotope effect in the thermopower for Ge is associated with the drift motion of thermal phonons. As was already observed in analyzing the thermal conductivity of Ge and Si crystals of different isotopic compositions [22, 23], a decrease in the degree of isotopic disorder brings about an abrupt increase in the contribution made by the drift motion of longitudinal phonons to the thermal conductivity. The same effect shows up in the drag thermopower.

Figure 5 gives the theoretically and experimentally obtained temperature dependence of the drag thermopower for $^{\text{nat}}$Ge and ^{70}Ge (99.99%). One can see in the figure that the theory provides a qualitative explanation of the isotope effect in the thermopower: the maximal values of $|\alpha_{\max}|$ in the case of transition from $^{\text{nat}}$Ge to ^{70}Ge (99.99%) increase by a factor of 1.3 for the value of $m_e \approx 0.22m_0$ and by a factor of 2.25 for $m_e \approx m_0$, which actually agrees with the experimentally observed increase in the direction [111]. This may point to the

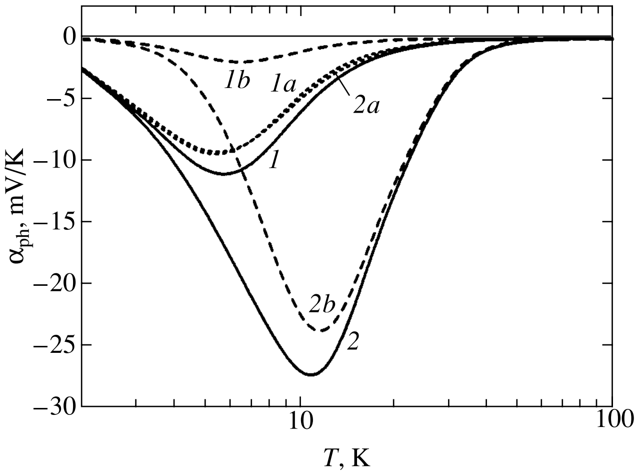

Fig. 4. The temperature dependence of (*1*, *2*) the drag thermopower, as well as of the contributions by (*1a*, *2a*) the diffusion and (*1b*, *2b*) drift of phonons for germanium of different isotopic compositions ($m_e = m_0$, E_{0L} = 4 eV) allowing for the inelasticity of electron–phonon scattering. Curves *1*, *1a*, and *1b* are for germanium of natural isotopic composition ($N_d = 4 \times 10^{12}$ cm^{-3}), and curves *2*, *2a*, and *2b* are for germanium with 99.99% ^{70}Ge ($N_d = 2 \times 10^{13}$ cm^{-3}).

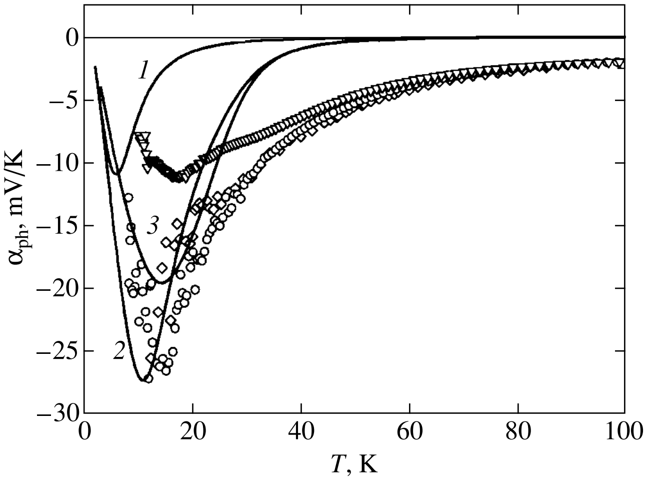

Fig. 5. The temperature dependence of the drag thermopower (E_{0L} = 4 eV). Curve *1* is for germanium of natural isotopic composition ($m_e = m_0$, $N_d = 4 \times 10^{12}$ cm^{-3}), curve *2* is for germanium with 99.99% ^{70}Ge ($m_e = m_0$, $N_d = 2 \times 10^{13}$ cm^{-3}) in the direction [111], and curve *3* is for germanium with 99.99% ^{70}Ge ($m_e \approx 0.9m_0$, $N_d = 2 \times 10^{12}$ cm^{-3}) in the direction [100]; the symbols indicate the experimental data.

predominant part played by one of four ellipsoids with the maximal effective mass along the direction [111]. However, the position of maxima for $^{\text{nat}}$Ge (see Figs. 3, 4, and 5) turns out to be shifted to the low-temperature region, $T_{\max} \approx 6$ K, while experiment gives $T_{\max} \approx 17$ K. For ^{70}Ge (99.99%), calculation gives $T_{\max} \approx 10$ K, while experiment produces $T_{\max} \approx 15$ K. In calculating the thermopower in the direction [100] (see Fig. 5, curve *3*), the deformation potential constant was not varied, and the velocities of sound were taken to be $s_L = 4.92 \times 10^5$ cm/s and $s_t = 3.55 \times 10^5$ cm/s, in accordance with [31]. In this case, the isotope effect in the thermopower with the same constant of deformation potential turned out to be 35% lower, which may be indicative of some anisotropy of the drag thermopower.

Note that the contribution by longitudinal phonons alone was taken into account in the calculation of the drag thermopower. Analysis revealed that, within the assumptions made, the isotope effect for transverse phonons was low and, upon transition from $^{\text{nat}}$Ge to highly enriched germanium, this contribution increased by approximately 10%. This is associated with the predominant part played by the diffusion motion of transverse phonons (for more detail, see [22]). Therefore, in this analysis, we ignored the contribution of transverse phonons, although the position of the maximum of $|\alpha_{ph}^t|$ is found at approximately 20–22 K. The inclusion of this contribution could have markedly improved the agreement between the calculated curves and the experimental data at temperatures above the maximum. However, the introduction of an additional fitting parameter into the theory hardly added anything to the physical content of this paper.

6. CONCLUSIONS

In this paper, we have interpreted the experimentally found strong dependence of the thermopower of germanium crystals on the isotopic composition. We have developed a theory of phonon drag of electrons in semiconductors with nondegenerate statistics of current carriers, which takes into account the effect of the phonon drift motion associated with the normal processes of phonon scattering. A qualitative explanation has been given of the isotope effect in the drag thermopower. It has been demonstrated that the rigorous inclusion of inelastic electron scattering brings about a significant (by factor of more than two) reduction of the absolute values of the drag thermopower. In our opinion, the isotropic band approximation for conduction electrons, as well as the assumption of the equality of the drift velocities of long-wavelength and thermal phonons, failed to provide for quantitative agreement with the experimental data on the drag thermopower, in contrast to calculations of thermal conductivity [22].

The inclusion of both of the above-identified factors requires significant mathematical effort, namely, a separate study of the relaxation of thermal and long-wavelength phonons and analysis of the Simons mechanism of normal processes of scattering, which leads to the redistribution of momentum between the thermal and long-wavelength phonons of different vibrational branches.

ACKNOWLEDGMENTS

We are grateful to A.P. Tankeev for the discussion of our results and critical comments.

This study was supported by the Russian Foundation for Basic Research (project nos. 00-02-16299 and 01-02-17469), by the CRDF (grant no. RP2-2274), and by the Dinastiya and MTsFFM Foundations.

REFERENCES

1. K. Itoh, W. L. Hansen, E. E. Haller, *et al.*, J. Mater. Res. **8**, 1341 (1993).
2. V. I. Ozhogin, A. V. Inyushkin, A. N. Taldenkov, *et al.*, Pis'ma Zh. Éksp. Teor. Fiz. **63**, 463 (1996) [JETP Lett. **63**, 490 (1996)].
3. M. Asen-Palmer, K. Bartkowski, E. Gmelin, *et al.*, Phys. Rev. B **56**, 9431 (1997).
4. A. P. Zhernov and D. A. Zhernov, Zh. Éksp. Teor. Fiz. **114**, 1757 (1998) [JETP **87**, 952 (1998)].
5. A. P. Zhernov, Fiz. Tverd. Tela (St. Petersburg) **41**, 1185 (1999) [Phys. Solid State **41**, 1079 (1999)].
6. C. Herring, Phys. Rev. **96**, 1163 (1954).
7. V. M. Askerov, *Electronic Transport Phenomena in Semiconductors* (Nauka, Moscow, 1985), p. 318.
8. I. M. Tsidil'kovskiĭ, *Thermomagnetic Effects in Semiconductors* (Nauka, Moscow, 1960; Academic, New York, 1962).
9. P. S. Zyryanov and M. I. Klinger, *Quantum Theory of Electron Transport Phenomena in Crystalline Semiconductors* (Nauka, Moscow, 1976), p. 480.
10. I. G. Kuleev, Fiz. Met. Metalloved. **90** (1), 14 (2000).
11. I. G. Kuleev, Fiz. Tverd. Tela (St. Petersburg) **42**, 649 (2000) [Phys. Solid State **42**, 423 (2000)]; Fiz. Tverd. Tela (St. Petersburg) **44**, 215 (2002) [Phys. Solid State **44**, 223 (2002)].
12. V. A. Kozlov and É. L. Nagaev, Pis'ma Zh. Éksp. Teor. Fiz. **13**, 639 (1971) [JETP Lett. **13**, 455 (1971)].
13. A. A. Bel'chik and V. A. Kozlov, Fiz. Tekh. Poluprovodn. (Leningrad) **20**, 53 (1986) [Sov. Phys. Semicond. **20**, 31 (1986)].
14. V. S. Oskotskiĭ, A. M. Pogarskiĭ, I. N. Timchenko, and S. S. Shalyt, Fiz. Tverd. Tela (Leningrad) **10**, 3247 (1968) [Sov. Phys. Solid State **10**, 2569 (1968)].
15. A. N. Taldenkov, A. V. Inyushkin, V. I. Ozhogin, *et al.*, in *Proceedings of IV Conference on Physicochemical Processes under Atom and Molecule Selection, Zvenigorod, 1999* (TRINITI, Troitsk, 1999), p. 243.
16. J. Callaway, Phys. Rev. **113**, 1046 (1959).
17. R. Berman, *Thermal Conduction in Solids* (Clarendon Press, Oxford, 1976; Mir, Moscow, 1979).
18. B. M. Mogilevskiĭ and A. F. Chudnovskiĭ, *Thermal Conductivity of Semiconductors* (Nauka, Moscow, 1972), p. 536.
19. S. Simons, Proc. Phys. Soc. London **82**, 401 (1963); Proc. Phys. Soc. London **83**, 799 (1963).
20. C. Herring, Phys. Rev. **95**, 954 (1954).
21. T. H. Geballe and G. W. Hull, Phys. Rev. **94**, 1134 (1954).
22. I. G. Kuleev and I. I. Kuleev, Zh. Éksp. Teor. Fiz. **120**, 1952 (2001) [JETP **93**, 568 (2001)].
23. I. G. Kuleev and I. I. Kuleev, Zh. Éksp. Teor. Fiz. **122**, 558 (2002) [JETP **95**, 480 (2002)].
24. L. É. Gurevich and I. Ya. Korenblit, Fiz. Tverd. Tela (Leningrad) **6**, 856 (1964) [Sov. Phys. Solid State **6**, 661 (1964)].
25. I. G. Lang and S. T. Pavlov, Zh. Éksp. Teor. Fiz. **63**, 1495 (1972) [Sov. Phys. JETP **36**, 793 (1973)].
26. I. G. Kuleev, Fiz. Met. Metalloved. **87**, 5 (1999).
27. I. G. Kuleev, Fiz. Tverd. Tela (St. Petersburg) **42**, 979 (2000) [Phys. Solid State **42**, 1009 (2000)].
28. E. Kaden and H.-L. Günter, Phys. Status Solidi B **126**, 733 (1984).
29. A. G. Samoilovich, I. S. Buda, and I. V. Dakhovski, Phys. Status Solidi **23**, 229 (1967).
30. A. G. Samoĭlovich and I. S. Buda, Fiz. Tekh. Poluprovodn. (Leningrad) **3**, 400 (1969) [Sov. Phys. Semicond. **3**, 340 (1969)].
31. R. Truel, C. Elbaum, and B. B. Chick, *Ultrasonic Methods in Solid State Physics* (Academic, New York, 1969; Mir, Moscow, 1972).

Translated by H. Bronstein