The Possibility of the “Giant” Isotope Effect for Ultrasound Absorption in Crystals

I. G. Kuleevab,*, I. I. Kuleevb, A. V. Inyushkina, and V. I. Ozhoginb

aInstitute of Metal Physics, Ural Division, Russian Academy of Sciences, Yekaterinburg, 620219 Russia
bInstitute of Molecular Physics, Russian Research Centre Kurchatov Institute, pl. Kurchatova 1, Moscow, 123182 Russia
*e-mail: kuleev@imp.uran.ru

Received December 21, 2004

Abstract—The effect of isotopic disorder on ultrasound absorption in germanium, silicon, and diamond crystals is considered. The “giant” isotope effect is predicted in the ultrasound absorption coefficient (in contrast to the isotope effect in the thermal conductivity and thermopower) of these crystals. The parameters determining the ultrasound absorption coefficients for the crystals under study with different degrees of isotopic disorder are determined from the known values of elastic moduli of the second and third order. The ultrasound absorption coefficients are analyzed as functions of temperature and wavevector for isotopically modified crystals. The possibility of experimental observation of this effect is considered. © 2005 Pleiades Publishing, Inc.

1. INTRODUCTION

The properties of isotopically enriched germanium, silicon, and diamond crystals have been attracting the attention of researchers over the last decade [1–11]. Like many other crystals, these crystals consist of two or more stable isotopes distributed at random over crystal lattice sites. The parameters of the force interactions between atoms in isotopically disordered crystals are virtually independent of the isotopic composition since the electron shell configuration of an atom weakly depends on the nuclear mass (the scale of the effect is on the order of the ratio of the electron and nuclear masses, i.e., ~ 10^{-4}–10^{-5}). The dependence of atomic vibrational modes on the isotopic composition emerges due to the fact that the motion of an atom in the potential produced by neighboring atoms is determined, among other things, by its mass $M_i$. Atomic vibrations at lattice sites can be treated as motion in a harmonic potential whose parameters depend on the unit cell volume of the crystal; in the quasi-harmonic approximation, the frequency and the squared amplitude of atomic vibrations are proportional to $M_i^{-1/2}$. As a rule, the effect of isotopic disorder on the vibrational spectra of the crystals is weak in view of the smallness of the ratio $|\Delta M/\bar{M}|$, where $\Delta M = M_i - \bar{M}$, $M_i$ is the mass of the $i$th isotope, $\bar{M} = \sum_i C_i M_i$ is the average mass of the isotopic composition, and $C_i$ is the concentration of the $i$th isotope. However, some kinetic coefficients may change significantly depending on the isotopic composition since the isotopic disorder violates translational invariance of the lattice and leads to phonon scattering. The effect of isotopic disorder on phonons can be described in terms of temperature-independent isotope scattering of phonons. The relaxation rate of this process is proportional to the fourth power of the phonon wavevector and the isotopic disorder factor

$$g = \sum_i C_i \left( \frac{\Delta M_i}{\bar{M}} \right)^2. \quad (1)$$

Upon a transition from germanium crystals with the natural isotopic composition $^{70}$Ge ($g = 5.87 \times 10^{-4}$) to crystals enriched to 99.99% in the $^{70}$Ge isotope ($g = 0.816 \times 10^{-4}$), the value of $g$ decreases almost by four orders of magnitude. However, according to [2, 3], the maximal (in $\bar{T}$) values of thermal conductivity increase by an order of magnitude, while the absolute values of thermopower [7] increase by more than two times. For silicon crystals enriched to 99.983% in the $^{28}$Si isotope ($g = 3.2 \times 10^{-7}$), the maximal values of thermal conductivity increase, according to [4, 5], by a factor of 7.5 as compared to its value for $^{28}$Si with the natural isotopic composition ($g = 2.01 \times 10^{-4}$). It follows from these results that the observed increase in the maximal values of thermal conductivity and thermopower is much smaller than the ratio of the isotopic disorder factors for natural and enriched compositions of both germanium and silicon. This is due to the fact that thermal conductivity is determined by the total time of phonon relaxation averaged over all thermally excited phonon states. The contribution to this quantity comes not only from isotope scattering, but also from the phonon–phonon scattering processes associated with anharmonism in lattice vibrations, phonon scattering at the sample surface, etc.

Here, we pay attention to another kinetic effect, namely, ultrasound absorption which can be affected by
scattering of acoustic waves from isotopic disorder more strongly than thermal conductivity and thermpower.

The absorption coefficient \( \alpha(\mathbf{q}, T) \) of ultrasound waves with wavevector \( \mathbf{q} \) and polarization \( \lambda \) is proportional to the phonon relaxation rate \( \nu_{\lambda}^{\mathbf{ph}}(\mathbf{q}, T) \) (see, for example, [12, 13]),

\[
\alpha(\mathbf{q}, T) = \frac{\nu_{\lambda}^{\mathbf{ph}}(\mathbf{q}, T)}{2s_{\lambda}},
\]

where \( s_{\lambda} \) is the velocity of phonons and \( T \) is the temperature. This expression implies that the procedure of averaging over phonon modes is not involved in determination of absorption coefficient \( \alpha(\mathbf{q}, T) \). Expression (2) is valid when the inequality \( \omega_{\mathbf{q}} \tau_{\lambda}^{\mathbf{ph}}(\mathbf{q}, T) \gg 1 \) holds, where \( \tau_{\lambda}^{\mathbf{ph}}(\mathbf{q}, T) = 1/\nu_{\lambda}^{\mathbf{ph}}(\mathbf{q}, T) \). For this reason, we will confine subsequent analysis to the range of temperatures and wavevectors \( \mathbf{q} \), in which this inequality holds. The dominant contribution to attenuation of acoustic waves comes from the boundary scattering of phonons, scattering from defects (including isotope scattering) and normal phonon–phonon scattering processes (see, for example, [12]). We will confine our analysis to these relaxation processes. Experimental studies of ultrasound absorption [12, 13] show that the inequality \( \omega_{\mathbf{q}} \tau_{\lambda}^{\mathbf{ph}}(\mathbf{q}, T) \gg 1 \) holds at low temperatures (namely, at temperatures lower than 50, 100, and 400 K for germanium, silicon, and diamond crystals, respectively). Obviously, phonon–phonon Umklapp scattering processes are ineffective in these temperature regions. In the opposite limiting case \( \omega_{\mathbf{q}} \tau_{\lambda}^{\mathbf{ph}}(\mathbf{q}, T) \ll 1 \) corresponding to the Akhiezer absorption mechanism, it is more appropriate to treat phonons as quasiparticles moving in a smoothly varying field of an acoustic wave. In this case, we must take into account the phonon–phonon Umklapp processes [12].

It should be noted that isotope scattering is independent of temperature and makes a constant contribution to the ultrasound absorption coefficient. The relaxation rates of phonon modes in anharmonic scattering processes rapidly decrease upon cooling; consequently isotope scattering at low temperatures can make the main contribution to attenuation of acoustic waves. In this case, for perfect crystal we have

\[
\alpha_{\lambda}(g_{l}) = \alpha_{0\lambda}g_{l}.
\]

Since the isotopic disorder factor decreases by more than 7000 (!) times upon a transition from \(^{60}\text{Ge}\) to \(^{70}\text{Ge}\) (99.99%), the magnitude of the isotope effect for the ultrasound absorption coefficient may reach giant values in contrast to isotope effects for thermal conductivity and thermopower.

One more circumstance that considerably increases the value of experimental study of ultrasound absorption in isotopically modified crystals is also worth noting. Namely, such structurally perfect and chemically pure crystals may exhibit fine features of phonon mode relaxation associated with anharmonicity of lattice vibrations.

2. ISOPOE EFFECT IN TRANSVERSE ULTRASOUND ABSORPTION IN GERMANIUM, SILICON, AND DIAMOND CRYSTALS

Let us consider by way of example the transverse sound absorption coefficient. In accordance with the prevailing idea [1–11, 14–16], the main mechanism of transverse ultrasound relaxation in normal three-phonon scattering processes is the Landau–Rumer mechanism, in which a longitudinal phonon is generated as a result of merging of a transverse and a longitudinal phonon \((T + L \rightarrow L)\) [17]. This relaxation mechanism is regarded as the main mechanism for transverse thermal phonons as well [1–11, 14–16]. It should be noted that the matrix element of three-phonon scattering processes for the relaxation mechanism of transverse phonons \((T \rightarrow T + T)\) is shown to vanish in the model [18] of an isotropic medium.

For this reason, we will henceforth assume that isotope scattering and normal phonon–phonon scattering processes in the Landau–Rumer mechanism make the dominant contribution to relaxation of transverse sound. In this case, in accordance with the Matthesien rule, we have

\[
\nu_{\phi}^{T} = \nu_{\phi}^{TLL} + \nu_{\phi}^{T},
\]

where the transverse phonon relaxation rate for scattering upon isotopic disorder is given by

\[
\nu_{\phi}^{T} = B_{i}T^{4}z_{1}^{4}, \quad z_{1} = z_{q}T = \frac{\hbar \omega_{q}}{k_{B}T},
\]

\[
B_{i} = g\frac{\hbar}{k_{B}}V_{0}\frac{V_{0}}{4\pi s_{T}^{3}}.
\]

Here, \( V_{0} \) is the volume per atom and \( s_{T} \) is the velocity of transverse photons.

The transverse phonon relaxation rate in the Landau–Rumer mechanism for cubic crystals has the form

\[
\nu_{\phi}^{TLL} = T^{5}z_{1}B_{i}^{T}(z_{1}, T),
\]

\[
B_{i}^{T}(z_{1}, T) = \frac{k_{B}^{3}z_{0}^{2}(1 - S^{2})}{16\pi\hbar^{4}\rho s_{T}^{3}L^{8}}(z_{1}/2)J_{1}(z_{1}, T).
\]

Coefficient \( B_{i}^{T}(z_{1}, T) \) was defined in [19]. In the long-wave limit \((z_{1}/z \ll 1)\), at temperatures much lower than the Debye temperature, expression (6) is transformed into the Landau–Rumer formula [17] with coefficient