

NUCLEAR SPIN-LATTICE RELAXATION IN GERMANIUM SINGLE CRYSTALS

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The temperature dependence of the spin-lattice relaxation time corresponding to the inelastic scattering of phonons by the ⁷³Ge quadrupole moment in Ge single crystals is calculated in the framework of the adiabatic bond charge model. The results obtained agree with the experimental data.

Introduction

Measurements of solid state magnetic resonance spectra and spin-lattice relaxation rates permit to obtain an information about the crystal lattice properties, namely, its local symmetry, structural and chemical imperfections, phonon spectrum. In the case of a nearly perfect crystal (both from the viewpoint of the chemical composition and the structure) the last barrier on the path to crystal further improvement is its naturally conditioned isotopic disorder. Recently it has been shown that the isotopic purification of germanium up to 99.99% in ⁷⁰Ge is capable to increase drastically (by 8 times) the thermal conductivity at the maximum as compared to germanium with the natural isotopic content (20.5% ⁷⁰Ge, 27.4% ⁷²Ge, 7.8% ⁷³Ge, 36.5% ⁷⁴Ge, 7.8% ⁷⁶Ge) [1].

Among numerous (although usually rather weak) isotope effects in the solid state physics, a slight dependence of the lattice constant on the isotopic composition should be noted [2]. The measured change of the lattice constant a per unit isotope mass of Ge at 78 K is equal to $(- 6.29 \pm 0.15) \cdot 10^{-5}$ E [3]. This geometric effect is caused by the anharmonic strains which are proportional to the mean square displacements of atoms from their equilibrium positions [4-6]. One of the germanium isotopes, ⁷³Ge, has the magnetic and quadrupole

moments (the nuclear spin $I = 9/2$, the nuclear gyromagnetic ratio $\gamma = -0.8795 \mu_N / \hbar$; the quadrupole moment $Q = -0.210 \cdot 10^{-24} \text{ cm}^2$ [7]). The isotopic disorder in Ge crystals causes random distribution of interion distances and the corresponding random distribution of the crystal electric fields affecting the ^{73}Ge quadrupole moments. Hence, fluctuations of the coupling length for a nuclear quadrupole moment in the non-monoisotopic lattice can be studied with the NMR techniques. In the perfect cubic germanium lattice, the electric field gradient (EFG) is zero, and the isolated ^{73}Ge nucleus has the equidistant Zeeman spectrum with a single resonance frequency independent on a direction of the applied magnetic field. Both the magnetic dipole-dipole interaction and the local lattice deformation affect the NMR spectrum of the multiisotopic lattice, and the shape of this spectrum depends on the magnetic field direction.

The propounded paper presents the first results of the thorough theoretical and experimental investigations of the lattice isotopic content effect on the NMR spectra and of the spin-lattice relaxation in the Ge single crystals with the composition corresponding to the natural isotope abundance and in the nearly monoisotopic crystal (96.3% of ^{74}Ge but with remanent 2.2% of NMR-active ^{73}Ge). To connect the peculiarities of the NMR spectrum with the isotopic content of a crystal we need to introduce the microscopic model of the crystal electric field with the explicit dependence on the atomic positions. For the case of ionic crystals the method of Sternheimer to calculate the EFG at the lattice sites is widely accepted [8,9]. In this method the crystal is conceived as an assembly of point charges surrounding the nucleus under consideration, and the polarization of the electronic core by the point charges is accounted for by a Sternheimer factor γ_∞ if the point charges are outside the considered core or by a Sternheimer function $\gamma(r)$ otherwise. In metals and semiconductors the actual electronic charge distribution can not be approximated by point charges in the lattice sites and has to be determined by accurate band-structure calculations [9]. However, the phenomenological

models based on the concept of the point bond charge have been derived to describe successfully the dielectric properties and the lattice dynamics of semiconductors [10]. In the present work the distribution of the quadrupole splittings in the spectra of ^{73}Ge nuclei due to the isotopic disorder in Ge crystals is considered in the framework of the Weber adiabatic bond charge model [11]. In the following, the temperature dependence of the spin-lattice relaxation time in Ge single crystals is calculated and compared with the experimental data to approve the model and to determine the model parameters. The detailed description of the measured NMR spectra and relaxation rates will be presented in the future publication [12].

General equations

The Hamiltonian of the ^{73}Ge nucleus can be presented as

$$\hat{H} = \hat{H}_0 + \hat{H}_1 + \hat{H}_2, \quad (1)$$

here the first term corresponds to the nuclear energy in the static lattice:

$$\hat{H}_0 = -\gamma\hbar\mathbf{H}\mathbf{I} - \frac{e^2Q}{4I(2I-1)} \sum_{m=-2}^2 O_2^m V_m, \quad (2)$$

\mathbf{H} is the applied magnetic field, $V_0 = V_{zz}$; $V_1 = V_{xz}$; $V_{-1} = V_{yz}$; $V_2 = \frac{1}{2}(V_{xx} - V_{yy})$; $V_{-2} = V_{xy}$ are components of the EFG (divided by the proton charge e) which are functionals of the electronic density in the lattice, and the nucleus quadrupole moment is expressed through the spin operators:

$$O_2^0 = 3I_z^2 - I(I+1), \quad O_2^1 = (I_+ + I_-)I_z + I_z(I_+ + I_-), \quad O_2^{-1} = -i[(I_+ - I_-)I_z + I_z(I_+ - I_-)],$$

$$O_2^2 = I_+^2 + I_-^2, \quad O_2^{-2} = -i(I_+^2 - I_-^2).$$

Two last terms in (1), H_1 and H_2 , correspond to the nuclear spin-phonon interaction, linear and quadratic in dynamic displacements of atoms from their equilibrium positions, respectively.

In the framework of a point charge model, $V_m = \sum_j V_m(j)$, and

$$V_{\alpha\beta}(j) = (1 - \gamma(r_j))Z(j) \frac{3x_{\alpha,j}x_{\beta,j} - r_j^2\delta_{\alpha\beta}}{r_j^5} \quad (3)$$

($Z(j)e$ is the value of a charge with the radius-vector \mathbf{r}_j relative to the fixed nucleus). The Sternheimer antishielding factors $\gamma(r_j)$ are different in a general case depending on the distance between the point charge and the nucleus [8]. Calculations of the EFG induced by the static or dynamic lattice deformations can be essentially simplified using the symmetrical properties of the lattice. It is convenient to consider the normalized linear combinations $u_{s\lambda}(\Gamma_n^p)$ of the charge displacements $u_\alpha(j)$ in the fixed coordination shell (labeled by s) of a given ^{73}Ge nucleus, corresponding to the irreducible representations Γ_n of the site symmetry group (indices n, p, λ denote the type, number and row of a representation, respectively). Expanding the EFG (3) in a series in the symmetrized dynamic displacements we can present the operator of the spin-lattice interaction in the following form:

$$\hat{H}_1 = -\frac{e^2 Q}{4I(2I-1)} \sum_m O_2^m \sum_{s\lambda np} V_{m,\lambda}(s, \Gamma_n^p) u_{s\lambda}(\Gamma_n^p), \quad (4)$$

$$\hat{H}_2 = -\frac{e^2 Q}{4I(2I-1)} \sum_{s,s'} \sum_{m=-2}^2 O_2^m \sum_{\substack{\lambda np \\ \lambda' n' p'}} \frac{1}{2} V_{m,\lambda\lambda'}(s\Gamma_n^p/s'\Gamma_{n'}^{p'}) u_{s\lambda}(\Gamma_n^p) u_{s'\lambda'}(\Gamma_{n'}^{p'}). \quad (5)$$

The charge displacements can be expressed by means of phonon creation and annihilation operators ($\omega_j(\mathbf{q})$ is a frequency of the j -th branch of the vibrational spectrum, \mathbf{q} is the phonon wave-vector, N is the number of crystal cells)

$$u_{s\lambda}(\Gamma_n^p) = \sum_{\mathbf{q}j} u_{s\lambda}(\Gamma_n^p | \mathbf{q}j) \left(\frac{\hbar}{2N\omega_j(\mathbf{q})} \right)^{1/2} (a_{\mathbf{q}j} + a_{-\mathbf{q}j}^+). \quad (6)$$

The spin-lattice relaxation rate, measured with the method of parallel fields (from the dynamical longitudinal susceptibility), is defined by the expression [13]

$$\frac{1}{T_1} = \frac{1}{2} \sum_{m,m'} \frac{W_{m \rightarrow m'} (E_m - E_{m'})^2}{Sp(\hat{H}_0^2)} \quad (7)$$

where E_m is the energy of a nucleus, and transition probabilities $W_{m \rightarrow m'}$ in the general case depend on a direction of the applied magnetic field. In the crystals with cubic symmetry, eqn.

(7) brings about the isotropic relaxation rate. Only the nonlinear terms in the spin-phonon interaction are relevant to calculations of the transition probabilities because contributions from the one-phonon relaxation processes can be neglected due to the low density of the lattice states at the NMR frequencies, and the contributions of the second order in \hat{H}_1 are negligible due to the Van-Vleck cancellation [14]. If the z-axis is parallel to the applied magnetic field, the transition probabilities, corresponding to the inelastic scattering of phonons by the nuclear quadrupole moment (Raman relaxation processes), are equal to

$$W_{m \rightarrow m \pm 1} = \frac{4}{\pi} \left[\frac{e^2 Q}{4I(2I-1)} \right]^2 | \langle m | O_2^1 | m \pm 1 \rangle |^2 \sum_{\substack{ss'tt'\lambda\lambda' \\ nn'pp'qq'}} V_{1,\lambda\lambda'}(s\Gamma_n^p/s'\Gamma_n^{p'}) V_{1,\lambda\lambda'}(t\Gamma_n^q/t'\Gamma_n^{q'}) \varphi(\Gamma_{n,st}\Gamma_{n',s't'}) \quad (8)$$

$$W_{m \rightarrow m \pm 2} = \frac{2}{\pi} \left[\frac{e^2 Q}{4I(2I-1)} \right]^2 | \langle m | O_2^2 | m \pm 2 \rangle |^2 \sum_{\substack{ss'tt'\lambda\lambda' \\ nn'pp'qq'}} \{ V_{2,\lambda\lambda'}(s\Gamma_n^p/s'\Gamma_n^{p'}) V_{2,\lambda\lambda'}(t\Gamma_n^q/t'\Gamma_n^{q'}) + V_{-2,\lambda\lambda'}(s\Gamma_n^p/s'\Gamma_n^{p'}) V_{-2,\lambda\lambda'}(t\Gamma_n^q/t'\Gamma_n^{q'}) \} \varphi(\Gamma_{n,st}\Gamma_{n',s't'}) \quad (9)$$

Here

$$\varphi(\Gamma_{n,st}\Gamma_{n',s't'}) = \int n(n+1) \text{Im}G(\Gamma_{n,st}^{pq}) \text{Im}G(\Gamma_{n',s't'}^{p'q'}) d\omega, \quad (10)$$

$n = (e^{\hbar\omega/kT} - 1)^{-1}$ is the phonon occupation number, and the imaginary parts of the lattice

Green functions are equal to

$$\text{Im}G(\Gamma_{n,ss'}^{pq}) = \frac{\pi}{N} \sum_{\mathbf{qj}} u_{s\lambda}(\Gamma_n^p | \mathbf{qj}) u_{s'\lambda'}(\Gamma_n^q | \mathbf{qj})^* \delta(\omega^2 - \omega_j^2(\mathbf{q})). \quad (11)$$

Model calculations

The adiabatic bond charge model of Weber [11] is used both to obtain the lattice Green functions and the EFG. A bond charge ($-Z_b e$) is placed on the each bond connecting two adjacent atoms. Each atom has four bonds (labeled by i), and the atom charge is equal to $Z_a = \frac{1}{2} \sum_i Z_b(i)$. In the equilibrium lattice, the bond charge is on the midway position

between the corresponding atoms, and has the value of $(-Z_b^0 e)$, the atom charge is equal to $2Z_b^0 e$. In the vibrating lattice the off mass bond charges move due to Coulombic and non-Coulombic forces, which appear when atoms and bond charges displace from their equilibrium positions. In the general case, the magnitude of the bond charge depends on the bond length r , which is equal to $r_0 = a\sqrt{3}/4$ at the equilibrium. Similarly to [15] we assume the exponential dependence in the form of

$$Z_b = Z_b^0 \left(\frac{r}{r_0} \right)^p. \quad (12)$$

The adiabatic bond charge model has been successfully used to describe the phonon spectrum of Ge [11] and anharmonic effects in the spectra of the Si/Ge heterostructures [15].

To estimate the spin-lattice relaxation rate, we considered explicitly interactions of the ^{73}Ge quadrupole moment with the three coordination shells, consisting of the nearest-neighbour four bond charges (the first shell), the four nearest atoms (the second tetrahedral shell) and the next twelve bond charges. Symmetrical coordinates of the atomic tetrahedron: $\Gamma_1, \Gamma_3, \Gamma_4$ (rotation), Γ_5^2, Γ_5^3 are given in Table 1 (translations Γ_5^1 are not included), and for the third coordination shell symmetrical coordinates are given in Table 2 (there were several errors in the previously published data [16], here only one row is given for the Γ_4^j, Γ_5^j representations, the other two can be obtained with the symmetry operations). Symmetrical coordinates of the bond charges in the first coordination shell (which are labeled similarly to atoms in the second shell) are defined by the same transformation, given in Table 1, but with one exception:

$$u_{1,1}(\Gamma_5^2) = (u_x(1) + u_x(2) + u_x(3) + u_x(4))/2 - 2 u_x(0),$$

and the $u_{1,2}(\Gamma_5^2)$ and $u_{1,3}(\Gamma_5^2)$ are defined similarly. The atom-atom, atom-bond charge, bond charge-bond charge Green functions have been calculated using the phonon frequencies and

the polarization vectors at the 5334 points in the 1/48 irreducible part of the Brillouin zone. The symmetrized Green functions are derived from the site-site Green functions, for example:

$$G(\Gamma_{1,22}) = G_{xx}(11) - 2G_{xx}(12) + G_{zz}(12) - 2G_{xy}(12) + 4G_{xz}(12);$$

$$G(\Gamma_{1,11}) = g_{xx}(11) - 2g_{xx}(12) + g_{zz}(12) - 2g_{xy}(12) + 4g_{xz}(12) + 2g_{xy}(11);$$

$$G(\Gamma_{5,11}^{22}) = g_{xx}(11) + g_{zz}(12) + 2g_{xx}(12) + 4G_{xz}(11) - 8G'_{xx}(01);$$

where $G_{\alpha\beta}(k,l)$ and $g_{\alpha\beta}(k,l)$ are the atomic and bond charge site (k)-site(l) Green functions, respectively, and $G'_{\alpha\beta}(k,l)$ is the atom-bond charge Green function.

Examples of the spectral densities for symmetrical coordinates of the first and second coordination shells are shown in Fig. 1. It is seen that there is some redistribution of the spectral densities of the bond charge displacements to higher frequencies. Due to strong correlations between the displacements of the adjacent bond charges (the distances between the bond charges in the first coordination shell are shorter than between the atoms in the second shell), the spectral densities for bond charges, as a rule, are much less than the corresponding densities for atoms, especially for coordinates of Γ_1 , Γ_3 and Γ_5^3 symmetry.

Analytical expressions of the parameters $V_{m,\lambda\lambda}(s\Gamma_n^p|s\Gamma_n^{p'}) = V_{m,\lambda\lambda}(s\Gamma_n^p|s\Gamma_n^p)$ in the Hamiltonian (5) of the spin-phonon interaction were obtained by expanding components of the EFG in a series in the atomic and bond charge dynamical displacements and then by transforming the displacement quadratic forms into the quadratic forms of the symmetrical coordinates. Two different values of the Sternheimer antishielding factor were introduced: γ_b for the electric field gradient due to the nearest bond charges, and γ_∞ for the electric field due to all other atoms and charges. From simple estimates with the physically justified model parameters, it follows that the contributions to the dynamic EFG at the ^{73}Ge nucleus from the four nearest atoms dominate. The explicit expressions of the corresponding independent non-zero coupling constants are given below:

$$V_{1,3}(2\Gamma_1|2\Gamma_5^2) = \sqrt{15}[V_{xyz} - 8W/3 - 2(\rho - 2)R/3];$$

$$V_{1,3}(2\Gamma_1|2\Gamma_5^3) = \sqrt{6}[V_{\text{xxx}y} + V_{\text{xy}y} + V_{\text{xy}z} - 5W - 2(\rho - 2)R]/3;$$

$$V_{1,12}(2\Gamma_3|2\Gamma_4) = -\sqrt{3}V_{1,22}(2\Gamma_3|2\Gamma_4) = \sqrt{3}(V_{\text{xxx}z} - V_{\text{xxx}z} - 4R)/2;$$

$$V_{1,13}(2\Gamma_3|2\Gamma_5^2) = V_{1,23}(2\Gamma_3|2\Gamma_5^2)/\sqrt{3} = \sqrt{30}(W + 4R)/6;$$

$$V_{1,13}(2\Gamma_3|2\Gamma_5^3) = V_{1,23}(2\Gamma_3|2\Gamma_5^3)/\sqrt{3} = \sqrt{3}(V_{\text{xxx}y} + V_{\text{xy}y} - 2V_{\text{xy}z} + 2W - 4R)/6;$$

$$V_{1,13}(2\Gamma_4|2\Gamma_4) = (V_{\text{xy}z} - V_{\text{xy}y} + 4R)/2;$$

$$V_{1,12}(2\Gamma_4|2\Gamma_5^2) = -V_{1,31}(2\Gamma_4|2\Gamma_5^2) = \sqrt{10}(V_{\text{xy}z} - V_{\text{xxx}y} - W + 4R)/2;$$

$$V_{1,12}(2\Gamma_4|2\Gamma_5^3) = -V_{1,31}(2\Gamma_4|2\Gamma_5^3) = (V_{\text{xy}z} - V_{\text{xy}y} - 2W - 4R)/2;$$

$$V_{1,12}(2\Gamma_5^2|2\Gamma_5^2) = 5[V_{\text{xy}y} - 4W/3 - 2(\rho - 4)R/3];$$

$$V_{1,12}(2\Gamma_5^3|2\Gamma_5^3) = (V_{\text{xy}y} + 3V_{\text{xy}z})/2 - 14W/3 - 2(2\rho - 5)R/3;$$

$$V_{1,12}(2\Gamma_5^2|2\Gamma_5^3) = V_{1,21}(2\Gamma_5^2|2\Gamma_5^3) = \sqrt{10}[V_{\text{xy}z} + V_{\text{xxx}z} - 11W/3 - 4(\rho - 1)R/3]/2;$$

$$V_{2,2}(2\Gamma_1|2\Gamma_3) = (V_{\text{xxx}x} - V_{\text{xy}y} + V_{\text{xxx}z} - V_{\text{xy}z} + 6W)/\sqrt{6};$$

$$V_{2,12}(2\Gamma_3|2\Gamma_3) = (V_{\text{xy}y} - V_{\text{xxx}x} + 2V_{\text{xxx}z} - 2V_{\text{xy}z})/2\sqrt{3};$$

$$V_{2,22}(2\Gamma_4|2\Gamma_4) = -V_{2,33}(2\Gamma_4|2\Gamma_4) = -(V_{\text{xy}y} - V_{\text{xxx}x} + 2V_{\text{xxx}z} - 2V_{\text{xy}z})/4;$$

$$V_{2,32}(2\Gamma_4|2\Gamma_5^2) = -V_{2,23}(2\Gamma_4|2\Gamma_5^2) = -V_{2,11}(2\Gamma_4|2\Gamma_5^2)/2 = V_{2,22}(2\Gamma_5^3|2\Gamma_5^2) =$$

$$= -V_{2,33}(2\Gamma_5^3|2\Gamma_5^2) = \sqrt{10}(V_{\text{xxx}z} - V_{\text{xy}z} + 2W)/4;$$

$$V_{2,32}(2\Gamma_4|2\Gamma_5^3) = -V_{2,23}(2\Gamma_4|2\Gamma_5^3) = -V_{2,11}(2\Gamma_4|2\Gamma_5^3)/2 = V_{2,22}(2\Gamma_5^2|2\Gamma_5^3)/10 =$$

$$= -V_{2,33}(2\Gamma_5^2|2\Gamma_5^3)/10 = (V_{\text{xxx}x} - V_{\text{xy}y} + 4W)/4;$$

$$V_{2,22}(2\Gamma_5^3|2\Gamma_5^3) = -V_{2,33}(2\Gamma_5^3|2\Gamma_5^3) = (V_{\text{xy}y} - V_{\text{xxx}x} - 2V_{\text{xxx}z} + 2V_{\text{xy}z} - 8W)/4.$$

Here $W = (1 - \gamma_\infty)\rho Z_b^0/4r_0^5$, $R = [(1 - \gamma_b) - (1 - \gamma_\infty)/16]\rho Z_b^0/r_0^5$,

and $V_{\alpha\beta\gamma\delta} = (3/r^5)(1 - \gamma_\infty)(2Z_b^0)[\delta_{\alpha\beta}\delta_{\gamma\delta} + \delta_{\alpha\gamma}\delta_{\beta\delta} + \delta_{\alpha\delta}\delta_{\beta\gamma} -$

$$(5/r^2)(\delta_{\alpha\beta}x_\gamma x_\delta + x_\alpha x_\beta \delta_{\gamma\delta} + \delta_{\alpha\gamma}x_\beta x_\delta + x_\alpha x_\gamma \delta_{\beta\delta} + \delta_{\alpha\delta}x_\beta x_\gamma + x_\alpha x_\delta \delta_{\beta\gamma}) + 35x_\alpha x_\beta x_\gamma x_\delta/r^4]$$

are the second derivatives of the EFG due to the atom in the site $(a a a)/4$ with the constant charge $2Z_b^0$. The coupling constants, which determine the dynamic EFG due to the bond

charges in the first coordination shell, are similar to those given above, but with $W = 0$, $R = 0$.

The cross terms in the operator (5), containing symmetrical coordinates of the first two shells,

appear due to changes of the atomic and bond charges with the change of the bond length, the corresponding coupling constants are proportional to $(1 - \gamma_b)\rho Z_b^0/r_0^5$. Because of neglecting the EFG modulation due to vibrations of the atoms and bond charges which are outward the selected complex, we have not considered changes of the bond charges in the third coordination shell. Thus, in the framework of this model we have four unknown parameters: the antishielding factors γ_∞ and γ_b , the magnitude of the bond charge Z_b^0 and the exponent ρ (see eqn. (12)) which determines the dependence of the bond charge on the bond length. As the preliminary values of these parameters we can use results obtained from the analysis of the lattice dynamics: $(Z_b^0)^2/\epsilon = 0.162$ ($\epsilon = 16$ is the static dielectric constant of germanium) [11], $\rho = 12.55$ [15], and $1 - \gamma_\infty > 100$ [17]. From the physical reasons it follows that $1 - \gamma_\infty > 1 - \gamma_b$ [8].

We calculated the spin-lattice relaxation time (see eqns. (7-9))

$$\frac{1}{T_1} = \frac{2}{5} \left[W_{(3/2) \rightarrow (1/2)} + \left(\frac{4}{5}\right)^2 W_{(3/2) \rightarrow (-1/2)} \right] = \frac{2}{5} \left[2W_{(3/2) \rightarrow (1/2)}^{\mathbf{H} \parallel [100]} + 3W_{(3/2) \rightarrow (1/2)}^{\mathbf{H} \parallel [111]} \right] \quad (13)$$

as a function of temperature using different sets of the model parameters. While the values of the bond charge, the exponent ρ and the Sternheimer factor γ_∞ (or γ_b) were fixed, the value of γ_b (or γ_∞) was determined by fitting the calculated time T_1 to the measured relaxation time at the temperature 120 K. It should be noted that the screened Coulombic fields of the atomic and bond charges were considered in the analysis of the lattice dynamics [11, 15], and not the magnitude of the bond charge Z_b^0 but the ratio $(Z_b^0)^2/\epsilon$ was determined in [11]. The EFG is proportional to Z_b^0/ϵ , and we have considered both possibilities with $\epsilon = 16$ and $\epsilon = 1$ using the fixed value of $(Z_b^0)^2/\epsilon$. The positive value of the exponent ρ , obtained in [15], corresponds to a metall-like zero approximation for the electronic density in a crystal, and we have examined the case of the negative exponent ρ , corresponding to the atom-like zero approximation, as well.

The temperature dependence of the spin-lattice relaxation time obtained with the values of the model parameters $1-\gamma_\infty = 100$, $\rho = 12.55$, $Z_b^0 = 1.61$, $\epsilon = 16$, given in the literature, and with $1-\gamma_b = 32.85$ from the fitting procedure is compared with the experimental data in Fig. 2. It is seen that the agreement is satisfactory in the temperature range over 50 - 300 K. When substituting $\rho = 0$ (the case of the constant bond and atom charges) or $\rho = -10$ (the magnitude of the bond charge diminishes with the increasing bond length) for the value of the exponent, we have to diminish the bond charge antishielding to match the calculated and measured values of T_1 at the point $T = 120$ K; with the values of $1-\gamma_b = 30.86$ and $1-\gamma_b = 17.74$, respectively, we obtain almost the same temperature dependence as that presented in Fig. 2. For the case of the zero bond charge antishielding ($\gamma_b = 0$) and $\rho = 12.55$, we have found it possible to describe the experimental data assuming $\epsilon = 1$ ($Z_b^0 = 0.402$) and with the value of $1-\gamma_\infty = 84.13$. The most preferable set of the model parameters will be selected in our future work from the more detailed analysis of the experimental findings [12].

Conclusion

From the comparison of the measured and calculated temperature dependencies of the relaxation time (see Fig. 2), it follows that the inelastic phonon scattering is the most essential mechanism of the spin-lattice relaxation for Ge. It is evident that only at low temperatures ($T < 30$ K) some other mechanisms (the most probable one is the relaxation due to a small amount of paramagnetic impurities) become dominant. At temperatures $T > 300$ K some additional mechanism of relaxation may exist as well. The interaction of the nuclear quadrupole moment with vibrations of the nearest four Ge atoms brings about the main contribution to the spin-lattice relaxation rate. The effective modulation of the EFG by the nearest bond charges is greatly reduced because of strong correlations between their displacements.

As the main result of the present investigation of the spin-lattice relaxation, we have derived the simple model which can be used to estimate for the ^{73}Ge nuclei the quadrupole

splittings induced by local deformations of the crystal lattice. This fact makes it possible to develop investigations of the NMR spectra as a method of the isotopic composition control in Ge crystals.

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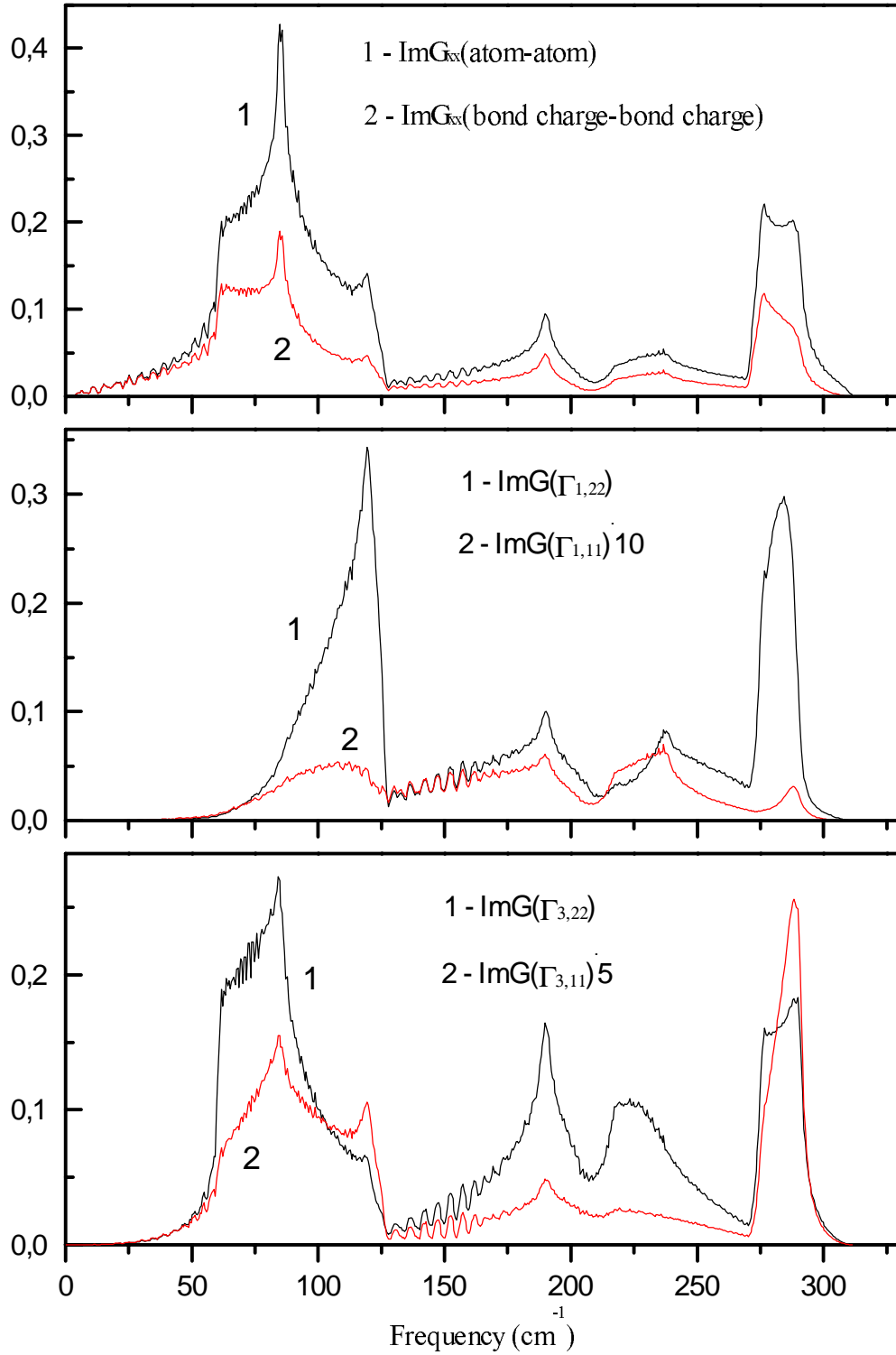
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Fig. 1. The spectral densities of the symmetrical coordinates, defined in the Table 1, for Ge atoms (1) and the bond charges (2). The curve a)1 corresponds to the phonon density of states.



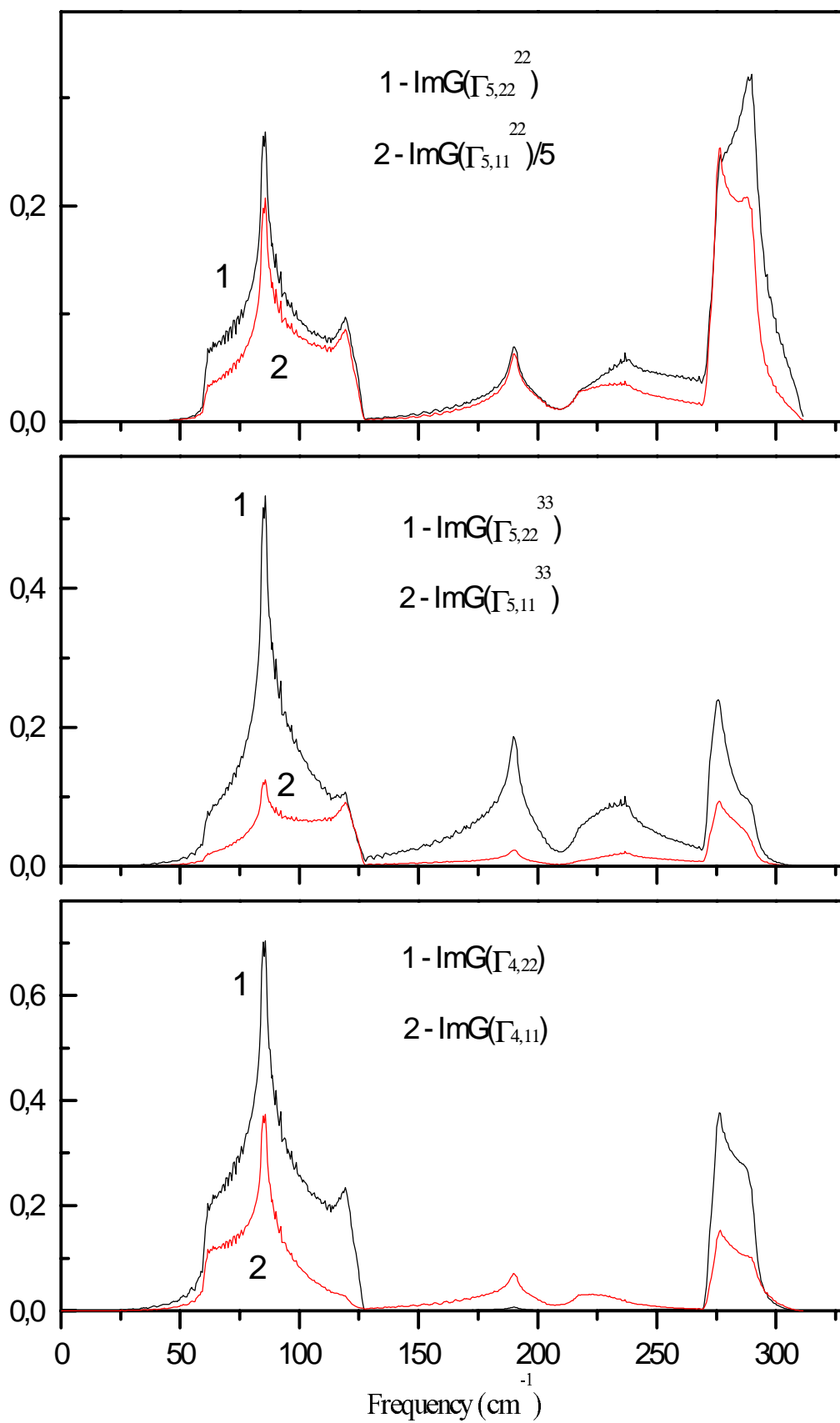


Fig. 2. The spin-lattice relaxation time for $^{73}\text{Ge}_{0.022}^{74}\text{Ge}_{0.97}$ sample. Results of calculations are presented by a solid line, open circles correspond to the measured data ($\mathbf{H} \parallel [001]$, $H = 5 \text{ T}$) [12].

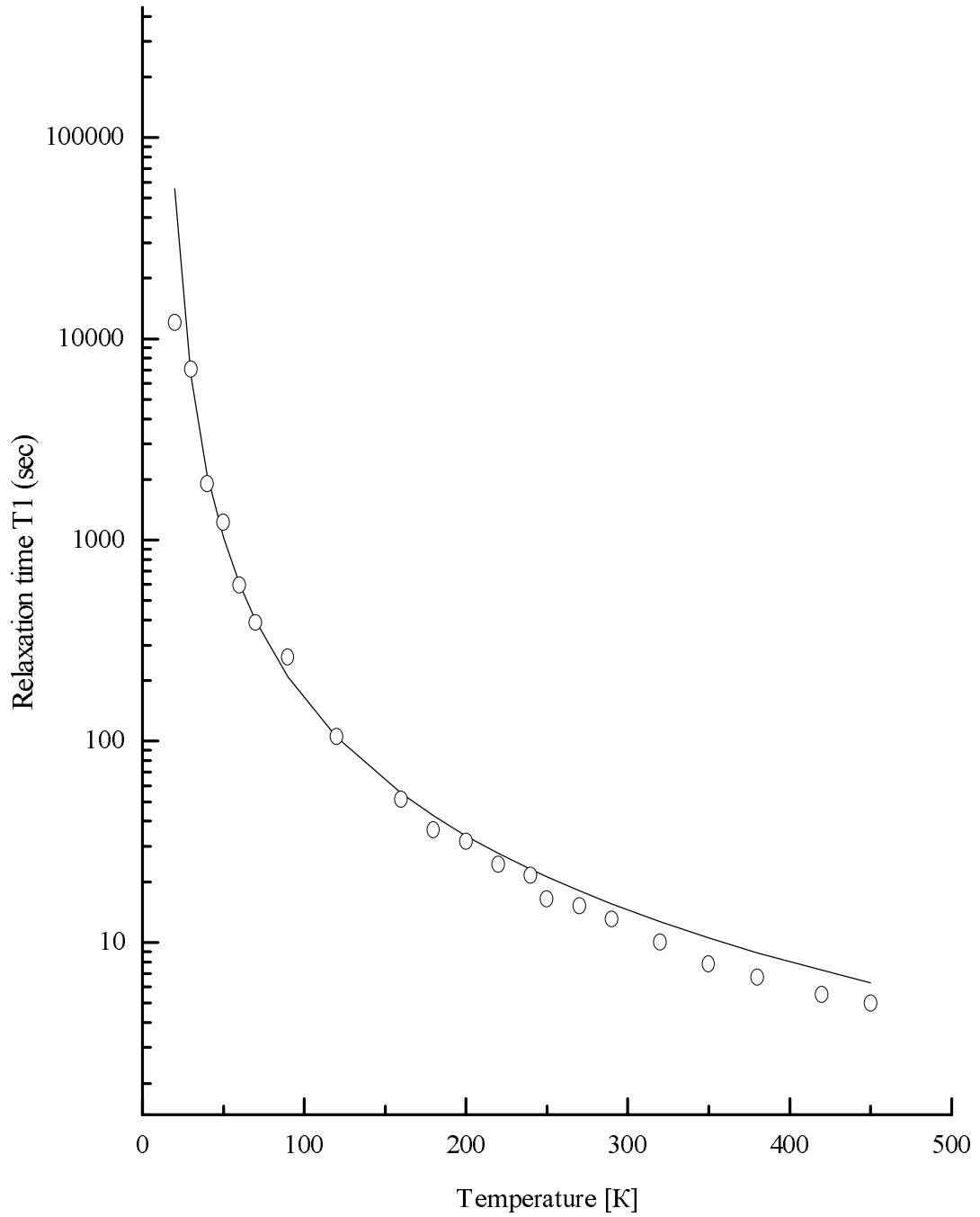


Table 1. Symmetrical coordinates of a crystal tetrahedral unit.

Atom (bond) number and coordinates in units of $a/4$	Γ_1	${}_1\Gamma_3$	${}_2\Gamma_3$	${}_1\Gamma_4$	${}_2\Gamma_4$	${}_3\Gamma_4$	${}_1\Gamma_5^2$	${}_2\Gamma_5^2$	${}_3\Gamma_5^2$	${}_1\Gamma_5^3$	${}_2\Gamma_5^3$	${}_3\Gamma_5^3$
x0 0	0	0	0	0	0	0	0	-4	0	0	0	0
y0 0	0	0	0	0	0	0	0	0	-4	0	0	0
z0 0	0	0	0	0	0	0	-4	0	0	0	0	0
x1 1	1	-1	1	-1	-1	0	0	1	0	1	0	1
y1 1	1	-1	-1	1	0	-1	0	0	1	1	1	0
z1 1	1	2	0	0	1	1	1	0	0	0	1	1
x2 -1	-1	1	-1	1	-1	0	0	1	0	-1	0	1
y2 -1	-1	1	1	-1	0	-1	0	0	1	-1	1	0
z2 1	1	2	0	0	-1	-1	1	0	0	0	-1	-1
x3 1	1	-1	1	1	1	0	0	1	0	-1	0	-1
y3 -1	-1	1	1	1	0	1	0	0	1	1	-1	0
z3 -1	-1	-2	0	0	1	-1	1	0	0	0	-1	1
x4 -1	-1	1	-1	-1	1	0	0	1	0	1	0	-1
y4 1	1	-1	-1	-1	0	1	0	0	1	-1	-1	0
z4 -1	-1	-2	0	0	-1	1	1	0	0	0	1	-1
normalization constant	$\frac{1}{\sqrt{12}}$	$\frac{1}{\sqrt{24}}$	$\frac{1}{\sqrt{8}}$	$\frac{1}{\sqrt{8}}$	$\frac{1}{\sqrt{8}}$	$\frac{1}{\sqrt{8}}$	$\frac{1}{\sqrt{20}}$	$\frac{1}{\sqrt{20}}$	$\frac{1}{\sqrt{20}}$	$\frac{1}{\sqrt{8}}$	$\frac{1}{\sqrt{8}}$	$\frac{1}{\sqrt{8}}$