Quadrupole Effects on ^{73}Ge NMR Spectra in Isotopically Controlled Ge Single Crystals

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NMR spectra of ^{73}Ge (nuclear spin $I = 9/2$) in perfect single crystals of germanium with different isotopic content were measured at 80, 300, and 450 K. The observed specific line shapes gave evidence of the isotopic disorder, in particular, abnormal broadening of the spectrum was found for the magnetic field directed along the [111] axis. Local lattice deformations in the germanium crystal lattice due to "isotopic disorder" were calculated in the framework of the adiabatic bond charge model. The results were applied to study random non-cubic crystal field interactions with the nuclear quadrupole moments and corresponding effects on NMR spectra. The simulated second moment of the resonance frequency distributions caused by the magnetic dipole-dipole and electric quadrupole interactions are used to analyze the lineshapes, theoretical predictions being in a qualitative agreement with the experimental data.

Key words: Isotopic Disorder; Ge Single Crystal; NMR Spectra; Quadrupole Effects.

1. Introduction

In perfect crystals the electric field gradient (EFG) at a nuclear site with cubic symmetry of the charge surrounding is zero. Magnetic energy levels of quadrupole nuclei ($I > 1/2$), characterized by spin projections m_I on the magnetic field, are spaced equidistantly. A single NMR line is expected in this case with a width determined only by magnetic spin-spin interactions. Crystal lattice defects should destroy the cubic symmetry of the charge distribution around the atom positioned near a defect. As a result an additional broadening of the NMR line should occur due to the nonvanishing quadrupolar shifts ($\Delta E_Q \sim m_I^2 V_{zz}$) of Zeeman energy levels. The change of the EFG (V_{zz}) around the lattice defect depends on its origin. The influence of traditional crystal defects (dislocations, vacancies and impurity atoms) on the NMR line patterns was discussed in excellent reviews [1 - 3].

In this paper we deal with another kind of structural disorder in crystals: the isotopic disorder among atoms of the same chemical element. In the framework of the harmonic approximation, a change in isotopic composition results only in changes of the vibration spectrum of the crystal lattice. As is known, the dynamical effect of isotopic disorder on the phonon scattering process manifests itself as one of the main contributions to the thermal conductivity at low temperatures [4, 5]. Spacings between atoms, as in the thermal expansion, experience variations only as a consequence of the anharmonicity of vibrations. In static phenomena, the isotopic disorder in real crystals causes a random distribution of the interion distances and a corresponding random distribution of the crystal electric fields affecting the quadrupole interaction of nuclei. The most pronounced effect of the isotopic disorder should be expected in cubic crystals. Single crystals of germanium represent the acme

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among them. Advanced technology of synthesis permits to grow Ge single crystals with a minimal concentration of traditional defects mentioned above. The rather large quadrupole moment of the ^{73}Ge isotope (I = 9/2, eQ = –0.19 barn) provides high sensitivity of the NMR experiments to detect small deviations from cubic symmetry around the resonant nuclei. A further increase of experimental performance can be achieved in single crystals predominantly enriched by an isotope without nuclear paramagnetic moment, like ^{70}Ge or ^{74}Ge. Magnetic spin-spin interactions in these crystals are reduced greatly, and the NMR of the isotope ^{73}Ge as an impurity atom will represent extremely high sensitivity to detect small lattice deformations due to isotopic disorder.

In Sect. 2 of this paper, the specific peculiarities of the quadrupole nuclei NMR signals due to isotopic disorder in the cubic crystal lattice are studied. Local static lattice deformations induced by the impurity isotopes are calculated in the framework of the adiabatic bond charge model (ABCM) of Weber [6]. NMR lineshape and linewidth dependences on the isotopic composition and temperature are considered. In Sect. 3, ^{73}Ge NMR spectra measured in crystals with different isotopic content for different orientations of a magnetic field with respect to crystallographic axes are compared with simulated spectral envelopes.

2. Quadrupole Line Broadening Induced by the Isotopic Disorder in the NMR Spectra

In the present work, ABCM [6, 7] is used both to obtain the lattice deformations, induced by the isotopic disorder, and the electric field gradient at the magnetic ^{73}Ge nuclei. In the frame of this model a bond charge (BC) $-Z_b e$ is placed on each bond connecting two adjacent Ge atoms. Each atom has four bonds of $a\sqrt{3/4}$ length (a is the lattice constant), and the atom charge is $eZ_a = 2eZ_b$. In the vibrating lattice the mass-less BC move due to Coulomb and non-Coulomb forces which appear when atoms and BC displace from their equilibrium positions. In the general case, the magnitude of the BC depends on the bond length [7]. In this work, we consider the constant value of the BC $Z_b = 0.1$ which has been used already in lattice dynamics calculations [6] and in the analysis of the nuclear spin-lattice relaxation (NSLR) of ^{73}Ge induced by the thermal fluctuations of the EFG in a perfect Ge crystal without isotopic disorder [8].

The Hamiltonian of the ^{73}Ge nucleus subsystem in the strong applied magnetic field $\boldsymbol{H}$ = (0;0;H) can be presented as

$$\hat{H} = \sum_j \Big(-\gamma\hbar I H_{zj} + \frac{e^2 Q}{4I(2I-1)} - \big[3I_{zj}^2 - I(I+1)\big] V_{zz}(j) \Big) + \frac{1}{2}\sum_{jk} \hat{H}_{jk}. \quad (1)$$

The first two terms in (1) corresponds to the magnetic and quadrupole energies of a single nucleus and the last term represents the secular part of the magnetic dipole-dipole interaction [2].

In the framework of the ABCM, the EFG (in units of the proton charge e) equals

$$V_{zz}(j) = -\sum_k (1-\gamma_\infty) Z_a c_{ij} + \sum_p \big(1-\gamma(r_{jp}L)\big) Z_b c_{b,jp}, \quad (2)$$

where $c_{jk} = [r(jk)^2 - 3z(jk)^2]/r(jk)^5$; $c_{b,jp} = [r_{jp}^2 - 3z_{jp}^2]/r_{jp}^5$; $\boldsymbol{r}(jk)$ and $\boldsymbol{r}_{jp}$ are vectors connecting j, k lattice sites, and j site with the bond charge p, respectively. We use two different values of the Sternheimer antishielding factor $\gamma(\boldsymbol{r})$, which have been approved in [8] by calculations of the NSLR rate: $1 - \gamma_b = 30.86$ for the EFG due to the nearest BC, and $1 - \gamma_\infty = 100$ for the EFG due to all other atoms and BC. An isolated ^{73}Ge atom has equidistantly spaced energy levels which lead to a single resonance frequency independent on a direction of $\boldsymbol{H}$. Both the magnetic dipole-dipole interaction and the EFG induced by local lattice deformations change the Zeeman splittings, and the shape of the spectrum in a real crystal depends on the magnetic field direction.

The crystal lattice consists of two (λ = 1,2) Bravais sublattices with sites defined by vectors $\boldsymbol{r}_{N\lambda} = an_1(1/2,0,1/2) + an_2(0,1/2,1/2) + an_3(1/2,1/2,0) + \boldsymbol{R}_\lambda$, where $\boldsymbol{R}_1 = 0$, $\boldsymbol{R}_2 = (a/4)(1,1,1)$. The isolated mass defect in the lattice site $N\lambda$ induces a totally symmetric local lattice deformation. Let us denote radial displacements of the nearest BC as $\sqrt{3}\delta_b$, radial displacements of Ge atoms in the first coordination shell of a single impurity isotope as $\sqrt{3}\delta_a$, the displacement vector of the BC with coordinates $(a/8)(1\,3\,3)$ as $(\delta_{b1}\ \delta_{b2}\ \delta_{b2})$; displacements of other BC in this coordination shell can be obtained by the

Table 1. Parameters of local lattice deformations due to impurity isotope centers in Ge in units of 10^{-6} nm per unit mass difference.

	— Temperature (K) —			
	0	80	300	450
Single impurity atom				
δ_b	–0.18	–0.13	–0.08	–0.05
δ_a	–0.85	–0.61	–0.21	–0.11
δ_{b1}	–0.020	–0.018	–0.004	0
δ_{b2}	–0.047	–0.042	–0.010	0
First nn pair				
$\delta^f{}_a$	–1.18	–0.79	–0.29	–0.13
$\delta^f{}_{a1}$	–0.92	–0.68	–0.25	–0.11
$\delta^f{}_{b1}$	0.11	0.08	0.04	0.01
$\delta^f{}_{b2}$	–0.72	–0.59	–0.16	–0.08
Second nn pair				
$\delta^s{}_a$	–1.80	–1.36	–0.50	–0.22
$\delta^s{}_{b1}$	–0.21	–0.15	–0.08	–0.05
$\delta^s{}_{b2}$	–0.14	–0.09	–0.08	–0.05

symmetry operations. It follows from results of calculations [9] that displacements of more distant atoms and BC do not exceed 1/10 of the nearest neighbor displacements. Calculated parameters of the local lattice deformation at different temperatures are given in Table 1.

Anisotropic properties of the cubic lattice reveal themselves by different responses on external perturbations along tetragonal and trigonal symmetry axes. Redistribution of the crystal charge density in the vicinity of the distant mass defect at the site $N\lambda$ leads to the following induced EFG at the site 01 ($\Delta = 8\delta_a - \delta_b - 9\delta_{a1} - 6\delta_{b2}$):

$$\delta V_{zz}(N\lambda) = \mp 90(1-\gamma_\infty)\frac{a^2\Delta}{64} Z_b V_5^{-2}(N\lambda)/r_{N\lambda}^6, \quad (3)$$

$$\boldsymbol{H}\|[001],$$

$$\delta V_{zz}(N\lambda) = \mp 50\sqrt{3}(1-\gamma_\infty)\frac{a^2\Delta}{64} Z_b \cdot \left(V_5^0(N\lambda) + \frac{\sqrt{2}}{5} V_5^{-2}(N\lambda)\right)/r_{N\lambda}^6, \quad (4)$$

$$\boldsymbol{H}\|[111].$$

The upper and lower signs in (3,4) correspond to $\lambda = 1$ and 2, respectively; $V_{pk}(N\lambda)$ are the spherical polynomials of order p, dependent on spherical angular coordinates of the vector $rN\lambda$ [10]. Due to the strong dependence of $\delta V_{zz}(N\lambda)$ on the distance $r_{N\lambda}$, the most essential changes of the equidistant Zeeman spectrum may be expected when the magnetic nucleus ^{73}Ge and the mass defect are close to one another. In real samples, we have to consider the ^{73}Ge atom itself as an impurity center, and correlations between this atom and mass defects in its surroundings are to be taken into account. As it can be seen in Table 1, these correlations enhance relative atomic displacements in the first nearest-neighbor pair by up to 40%.

This pair center (for example, two identical mass defects in the sites (000) and $(a/4)(111)$) has trigonal symmetry, its local structure can be characterized by four parameters: $\sqrt{3}\delta_f$ = displacements of the impurity atoms in opposite directions along the pair axis, $\sqrt{3}\delta^f_{a1}$ = radial displacements of the six nearest-neighbor Ge atoms, displacement vectors of the BC on the corresponding six bonds are determined by two parameters, in particular, $\delta\boldsymbol{r} = (\delta^f_{b1}, \delta^f_{b2}, \delta^f_{b2})$ for the BC at the site $(a/8)(1,-1,-1)$. Taking into account interactions of the nuclear quadrupole moment with the four nearest BC's and four nearest atoms, we obtain

$$\delta V_{zz}(02) = \frac{2^{10}\sqrt{3}Z_b}{27a^4}\left[8(1-\gamma_b)(200\delta^f_a + \delta^f_{b1} + \delta^f_{b2}) - (1-\gamma_\infty)(29\delta^f_a - 9\delta^f_{a1})\right] \quad (5)$$

If the symmetry axis of the nearest-neighbor pair is declined from the magnetic field direction [111], the variation of the EFG equals $(-\delta V_{zz}(02)/3)$. With the local deformation parameters given in Table 1, we obtain quadrupole shifts of the resonance frequencies in the nearest neighbor ^{73}Ge pairs in perfect ^{74}Ge or ^{72}Ge crystals as large as 90 Hz at room temperature. These shifts exceed the contributions of the magnetic dipole-dipole interaction into the nuclear energies, namely these shifts determine the total spread of the NMR spectra in the magnetic field directed along the trigonal symmetry axis of a crystal.

The next nearest-neighbor impurity pair center (in particular, two identical mass defects in sites (000) and $(a/2)(101)$) has rhombic symmetry. In this case, we can neglect the displacements of the impurity atoms. The displacement $(0, \delta^s_a, 0)$ of the Ge atom, which is the common nearest neighbor of both impurity atoms, is perpendicular to the pair axis. Displacements of the corresponding bond charges in sites $(a/8)(111)$ and $(a/8)(313)$ can be denoted as $(\delta^s_{b2}, \delta^s_{b1}, \delta^s_{b2})$ and $(-\delta^s_{a2}, \delta^s_{b1}, -\delta^s_{b2})$. Calculated values of displacements are given in Table 1. Quadrupole shifts of

Table 2. Isotopic composition of Ge samples.

Sample	$\langle m \rangle$	— Isotope (atomic weight) — ^{70}Ge (69.924)	^{72}Ge (71.922)	^{73}Ge (72.923)	^{74}Ge (73.921)	^{76}Ge (75.921)	$c(^{73}\mathrm{Ge})^{1/2}$	$100\, g_2$
		— Isotope content, % —						
Ge-0.1	70.03	96.3	2.1	0.1	1.2	0.3	0.0316	0.880
Ge-3	73.91	0.1	0.9	3.8	92.6	2.6	0.1949	0.596
Ge-n	72.61	20.84	27.54	7.73	36.28	7.61	0.2780	2.424

the resonance frequencies in these twelve second nn-pairs are nonzero when the magnetic field is directed along the [001] or [111] axes, but their values are by an order of magnitude less than the shifts in the first nn-pairs in the field $\boldsymbol{H} \parallel [111]$.

It is well known that point defects induce lattice strains [11]. From crude estimates based on the elastic continuum approximation it follows that the long-range interactions between a nuclear quadrupole moment and lattice strains already prevail over the short-range interaction with local electric multipoles at the defect sites at distances between the ^{73}Ge atom and the impurity isotope larger than 2-3 lattice constants. However, this coupling with lattice strains would cause a more pronounced isotopic disorder effect in the magnetic field $\boldsymbol{H} \parallel [001]$ than in the field $\boldsymbol{H} \parallel [111]$ [9], but this is not the case experimentally (see the next section).

Due to the random distribution of atoms with different masses over lattice sites, we can consider quadrupole moments of ^{73}Ge in random crystal fields with the EFG represented by a sum of expressions (3-5) weighted by site occupation numbers (which are random variables and equal to 0 or 1). Note that contributions due to the second nn-pairs are not presented above in the explicit form [9]. As the simplest approximation for the NMR line shape, we can use the sum of weighted Gaussians corresponding to individual magnetic dipole transitions $m_I \leftrightarrow m_{I\pm1}$ (the detailed discussion of other distribution functions is presented in [9]):

$$f(\nu) = \Big(\int f(\nu)\mathrm{d}\nu \Big) \sum_{m=-7/2}^{m=9/2} \frac{|\langle m|I_x|m-1\rangle|^2}{[(2m-1)^2 b^2/4 + d^2]^{1/2}} \cdot \exp\Big(\frac{-\nu^2}{[(2m-1)^2 b^2/4 + d^2]} \Big) \Big/ \pi^{1/2} \sum_{m=-7/2}^{m=9/2} |\langle m|I_x|m-1\rangle|^2, \quad (6)$$

here $b^2 = 2M_2$ ($m_I = 1/2$).

The second moment $M_2(m_I)$ of the line corresponding to the transitions $\pm m_I \leftrightarrow \pm(m_I{+}1)$ (m_I = 1/2, 3/2, 5/2, 7/2) equals (we neglect correlations between mass defects)

$$M_2(m_1) = \langle (\nu_{m_1} - \nu_0)^2 \rangle = \big(\langle m \rangle g_2 \big)^2 \sum_{N\lambda} \Big[\frac{3e^2Q(2M_1+1)}{8\pi\hbar I(2I-1)} \delta V_{zz}(N\lambda) \Big]^2, \quad (7)$$

where $\nu_0 = (\gamma/2\pi)H$, $\langle m \rangle$ is the average atomic mass in the crystal, and the parameter $g_2 = \langle \Delta m^2 \rangle^{0.5} / \langle m \rangle$ characterizes the isotopic disorder in the real crystal. The parameter d determines the magnetic contribution to the total linewidth. Long tails of the NMR signal are to be expected: in particular, for the Gaussian line shape, the ratio of the full width at half height to the full width at 1/20 of line height equals 2.078, for (6) this ratio increases up to 3.445. The nearest-neighbor pairs do not contribute to the linewidth in the field $\boldsymbol{H} \parallel [001]$, thus the minimum and maximum values of linewidths may be expected for $\boldsymbol{H} \parallel [001]$ and $\boldsymbol{H} \parallel [111]$, respectively. We neglected in numerical simulations the interactions of magnetic nuclei with lattice strains at large distances from the impurity isotopes because it is difficult to estimate errors caused by the elastic continuum approximation.

3. Experimental Results and Discussion

The NMR spectra of ^{73}Ge have been measured for 3 single crystals ($6\times6\times6$ mm^3) of the purest germanium (carrier concentration $n_{\mathrm{carr}} \sim 10^{12}$ cm^{-3} at 290 K) with different isotopic content (see Table 2). The crystal "Ge-n" was synthesized from the material with natural isotope abundance. A much lower level of the isotopic disorder was reached in the two other crystals. The isotopic content of the sample "Ge-0.1" presented the possibility to suppress the magnetic

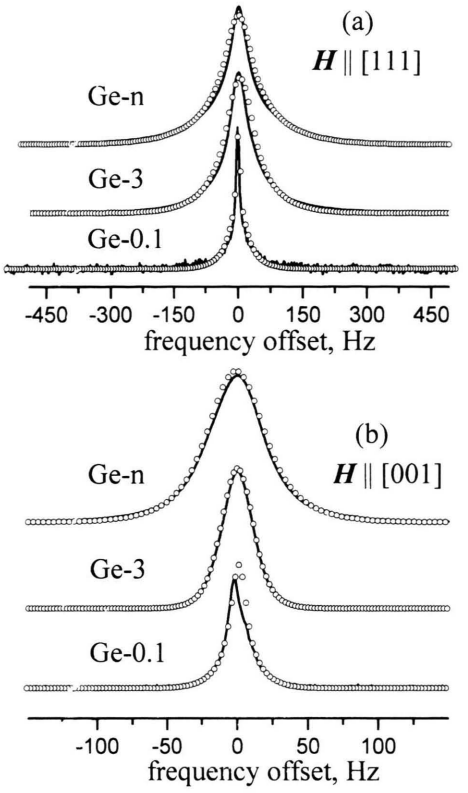

Fig. 1. ^{73}Ge NMR spectra of Ge-n, Ge-3 and Ge-0.1 samples at room temperature in a magnetic field of 12 T oriented along the crystal trigonal [111] (a) and tetragonal [001] (b) symmetry axis. Solid lines: – experimental, dotted lines correspond to (6).

dipole mechanism of the line broadening. The samples were prepared following the procedure described in [12]. The electron microscopy studies confirmed a presence of an extremely low total density of dislocations ($n_{\mathrm{disl}} < 10^3$ cm^{-2}). This permits us to exclude from the following consideration a possible influence of traditional defects [1 - 3] as a source of quadrupole broadening of the NMR line measured for all three single crystals.

The NMR measurements were performed at room temperature with pulse NMR spectrometers operating at the frequencies ν = 3.6 and 17.44 MHz. The inhomogeneity of the static magnetic field was minimized to 0.2 ppm within the sample. In experiments with a single exciting pulse ($t_{\mathrm{pulse}} = \pi(6 \leq 5\mu\mathrm{s})$, the ^{73}Ge free induction decay signal (FID) was measured. After Fourier-transformation of the FID the NMR line was found to be shifted at $\Delta\nu/\nu$ = –108(2) ppm with respect to the peak of the ^{73}Ge line in a water solution of $GeCl_4$, used as a reference.

Table 3. The quadrupole ($\Delta\nu_{\mathrm{Q}}$) and magnetic ($\Delta\nu_{(\mathrm{M})}$) width parameters (in Hz) obtained from the fit with expression (6) for ^{73}Ge NMR spectra measured at T = 300 K. Corresponding calculated values are given in brackets.

	— $\boldsymbol{H}$ || [001] —		— $\boldsymbol{H}$ || [111] —	
	$\Delta\nu_{\mathrm{Q}}$	$\Delta\nu_{\mathrm{M}}$	$\Delta\nu_{\mathrm{Q}}$	$\Delta\nu_{\mathrm{M}}$
Ge-n	27.8 (25.4)	32.5 (25.1)	93.2 (113.8)	50 (71.0)
Ge-3	11.4 (6.1)	20.2 (17.6)	66.6 (28.5)	48 (49.8)
Ge-0.1	13.0 (9.1)	8.5 (2.9)	34.1 (39.8)	9.6 (8.1)

The ^{73}Ge NMR spectra measured at the room temperature are shown in Figure 1. We studied with care the influence on the spectra of the magnetic interactions linear in nuclear spin (MILNS), such as inhomogeneity of magnetic field within a sample, demagnetizing effects, distribution of the chemical shift. As a result of measurements at different frequencies we found that the width of the central peak (–1/2 ↔ 1/2) was independent on ν for Ge-n and Ge-3 samples. Independently the ^{73}Ge spectra in these crystals were obtained at ν = 13.55 MHz as Fourier transformation of the spin-echo amplitude envelope $A(2t_i)$ in the following sequence: $\{(\pi/2)_x - t_i - \pi_x - t_i$-echo-$5T_1$-$(\pi/2)_{-x} - t_i - \pi_x - t_i$-echo-$5T_1\}_n$ with t as a variable parameter. Here T_1 is the NSLR time of ^{73}Ge. As it is well known, the MILNS do not contribute to the $A(2t)$ envelope in these spin-echo experiments [2]. From these additional experiments, we found that the main peculiarities of the line shapes were due to the combined effect of the quadrupole and magnetic interactions bilinear in nuclear spins.

The measured spectra were analyzed using the assumed line shape (6). The quadrupole $\Delta\nu_{\mathrm{Q}} = 2(\ln 2)^{1/2}b$ and magnetic $\Delta\nu_{\mathrm{M}} = 2(\ln 2)^{1/2}d$ parameters were obtained (see Table 3), and the calculated envelopes were compared with the experimental data in Figure 1. The temperature variation of the fitting parameters for the NMR lines in the Ge-3 sample (measured in spin-echo experiments with $\boldsymbol{H}$ || [111]) are the following: $\Delta\nu_{\mathrm{Q}}$ = 98 Hz (80 K), 79 Hz (300 K), 75 Hz (450 K); $\Delta\nu_{\mathrm{M}}$ = 28 Hz. Comparison of these preliminary data with the calculated relative changes of $\Delta\nu_{\mathrm{Q}}(\boldsymbol{H} \parallel [111])$ vs. T: 1(0 K): 0.72(80 K): 0.325(300 K): 0.160 (450 K), shows that the theory predicts much stronger effects (temperature narrowing) than observed, diminishing of the quadrupole contributions to the line broadening.

A comparison of the calculated and measured lineshapes, given in Fig. 1, shows that there are specific peculiarities of the NMR signals in crystals with isotopic disorder which cannot be described by the assumed superposition of the Gaussians. These peculiarities (the "logarithmic type" line shape) are very expressive in the spectra taken in the magnetic field parallel to the trigonal symmetry axis of the Ge crystal. The quadrupole contributions $\Delta\nu_Q$ to the linewidths were calculated in accordance with (7), supposing the Gaussian lineshape. In this case $\Delta\nu_Q$ should be a linear function of the parameter g_2. In all cases the considered calculated and experimental values of $\Delta\nu_Q$ agree satisfactorily (see Table 3), except the spectrum of the Ge-3 sample measured at $\boldsymbol{H}$ || [111]. The expected ratios of the quadrupole widths in the samples Ge-0.1-Ge-3-Ge-*n* (1:0.68:2.75) are qualitatively consistent with the experimental data for $\boldsymbol{H}$ || [001] - (1:0.87:2.14), but there is a remarkable contradiction with the data for $\boldsymbol{H}$ || [111] - (1:1.95:2.73). The Ge-3 sample has the minimum g_2 value, about 1.5 times less than in the Ge-0.1 sample (see Table 2). But the width of the plateau in the NMR signal in this sample in the case of $\boldsymbol{H}$ || [111] is more than 2.5 times larger than in the corresponding NMR line in the Ge-0.1 sample. It seems that in this case the approximation of the line shapes by Gaussians is not approved [9].

Now we consider the magnetic contribution to the linewidth ($\Delta\nu_M$) as a function of the concentration c of the magnetic nuclei. An examination of $\Delta\nu_M$ values obtained by the fitting procedure (see Table 3) gives evidence that the approximation $\Delta\nu_M(c) \sim c^{1/2}$, which is consistent with the assumed Gaussian line shape, describes variations of $\Delta\nu_M$ through the set of Ge-0.1, Ge-3, Ge-*n* samples with remarkable errors – ratios of $c(^{73}\mathrm{Ge})^{1/2}$ equal 1:6.17:8.80 (see Table 2), and the relative changes of the magnetic linewidths are 2.60:6.17:9.93 for $\boldsymbol{H}$ || [001] and 1.23:6.17:6.43 for $\boldsymbol{H}$ || [111].

The calculated linewidths $\Delta\nu_M = 2(2M_{m,2}\ln 2)^{1/2}$ [2], presented in Table 3, agree qualitatively with the data obtained by the fitting procedure. If we restrict ourselves only to magnetic dipole-dipole interactions we find that the calculated contribution is smaller than $\Delta\nu_M$ obtained by the fitting with expression (6). We suggest that an additional, bilinear in nuclear spins, interaction like the pseudo dipolar indirect coupling might be involved to explain quantitatively the observed $\Delta($$_M$ in the crystal with low concentration of the ^{73}Ge magnetic isotope.

4. Conclusion

We have unambiguously shown that the isotopic disorder causes some specific features of the NMR spectra of nuclei with nonzero quadrupole moment. In particular, we would like to emphasize the unusual (in solid state NMR spectra) line narrowing with increasing temperature and the specific lineshape with a wide plateau.

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[1] U. H. Cohen and F. Reif, Solid State Phys. ed by Seitz and Turnbull, **5**, 321 (1957).
[2] A. Abragam, The Principles of Nuclear Magnetism, Clarendon Press, Oxford 1961.
[3] M. Mehring and O.Kanert, in Progr. Magn. Res. Spectroscopy, ed. by Freude, v.3 (1972).
[4] M. Omini and A. Sparavigna, Physica B **233**, 230 (1997).
[5] V. Ozhogin, A. Inyushkin, and A. Taldenkov, JETP Lett. **63**, 490 (1996).
[6] W. Weber, Phys. Rev. B **15,** 4789 (1997).
[7] R. Eryigit and I. P. Herman, Phys. Rev. B **53**, 7775 (1996).
[8] B. Z. Malkin, S. K. Saikin, and V. I. Ozhogin, Appl. Magn. Res. **14**, 513 (1998).
[9] A. Trokiner, B. Z. Malkin, and S. V. Verkhovskii, Appl. Magn. Res. (1999).
[10] L. K.Aminov, B. Z. Malkin, and M. A. Teplov, Handbook on the Physics and Chemistry of the Rare-Earths, v. **22**, ed. K. A. Gschneidner and LeRoy Eyring, North-Holland, Amsterdam 1996, pp. 295-506.
[11] M. Ivanov, V. Ya. Mitrofanov, L. D. Falkovskaya, and A. Ya. Fishman, JMMM, **36**, 26 (1983).
[12] Kohei Itoh, Ph. D. Thesis, University of California at Berkeley. Dec. 1994.