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NMR study of the isotopically engineered Ge single crystals

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Abstract We report on the first NMR study of ^{73}Ge nuclear spin decoherence in germanium single crystals with different abundance of the ^{73}Ge isotope. Hahn echo decays are well fit by a superposition of two exponentials. The deviation from the single exponential is more pronounced in the more spin-diluted sample, causing long-lived echoes. We show that the decay of these echoes becomes slower with the reduction of ^{73}Ge abundance and is therefore caused by dipole–dipole interaction, reflecting the fundamental decoherence process in the spin system. The fast decay at the beginning of the relaxation process is shown to be mainly caused by the quadrupole interaction. Our experimental findings are supported by the calculations of Hahn echo decays in the germanium crystals under study. Quite good agreement between the theory and experiment is demonstrated.

Keywords Germanium · NMR · Quantum computing

Semiconductor-based architectures are quite realistic in constructing the large-scale quantum computer (QC) owing to the well-developed semiconductor technology. Among different QC concepts, nuclear spins are considered to be ideal quantum bits

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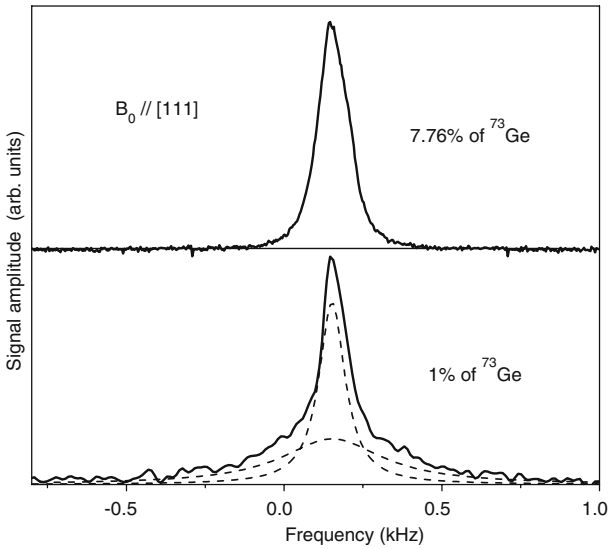

Fig. 1 Room temperature ^{73}Ge NMR spectra of the Ge-7.76% (*top*) and Ge-1% (*bottom*) single crystals. External magnetic field B_0 is applied along the [111] direction. *Dashed lines* show bi-Lorentzian fit

(qubits) embodied in semiconductor crystals, particularly in Si and Ge, since in both these crystals only one isotope (^{29}Si and ^{73}Ge) has nuclear spin and therefore isotopic engineering of Si and Ge permits to control the density of spins and vary the spin coherence time, a crucial parameter in spintronics and quantum computing. In the present paper, we report on the first NMR study of ^{73}Ge nuclear spin decoherence in germanium crystals with different abundance of the^{73}Ge isotope ($I = 9 / 2$).

NMR measurements were carried out on Ge single crystals with ^{73}Ge abundance of $f = 1\%$ and 7.76% (natural abundance), later denoted as Ge-1% and Ge-7.76%, respectively. ^{73}Ge spectra (Fig. 1) are symmetric resonances. In Ge-7.76% crystal the contributions to the NMR line width coming from the dipole–dipole and quadrupole interactions and magnetic field inhomogeneity are of the same order of magnitude. In the isotopically diluted Ge-1% crystal, the dipole–dipole coupling is significantly reduced. In spite of this fact, the NMR spectra of this crystal (Fig. 1) show broader wings in comparison with the Ge-7.76% crystal, which is evidently caused by the worse crystal perfection and larger contribution coming from the first order quadrupole coupling.

To measure the spin-spin relaxation (decoherence) time T_2, Hahn echo pulse sequence was applied. Ge-7.76% crystal shows a non-exponential Hahn echo decay (Fig. 2), which is well fit by a superposition of two exponentials with the decay times T_{21} and T_{22}. The observed effect is much more pronounced in Ge-1% crystal, in which the Hahn echo decay envelope exhibits strikingly non-exponential behavior (Fig. 3). Decay parameters T_{21} and T_{22}, obtained by fitting the $M(t)$ plots, are given in Table 1.

Statistical theory of NMR lineshape [1, 2] in magnetically diluted spin system with dipolar coupling predicts Lorentzian line shape with the line width proportional to the nuclear spin concentration ($\Delta v \sim f$). In such a case, T_2 should be inversely proportional to the isotope content. One can find out from Table 1 that the echo decay at the long-time domain, described by the time T_{22}, becomes slower with the reduced ^{73}Ge abundance, and the ratio T_{22}(Ge-1%)/T_{22}(Ge-7.76%) is more or less close to the reciprocal ratio of the isotope contents. Therefore we conclude that the

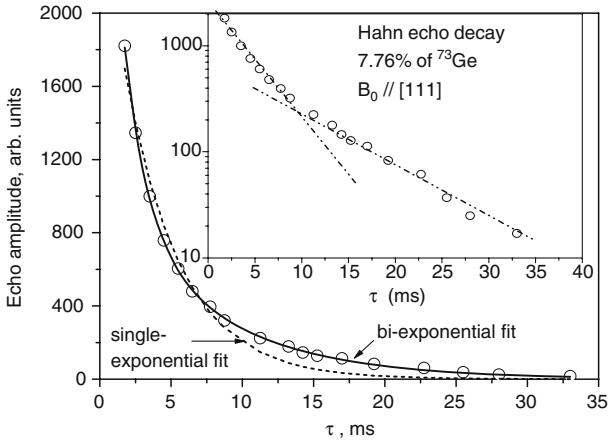

Fig. 2 ^{73}Ge Hahn echo decay envelope in the linear and semi-logarithmic (*insert*) scales in Ge single crystal with natural abundance of ^{73}Ge isotope. External magnetic field is applied along the [111] axis. *Dash* and *solid lines* show single and bi-exponential fits, respectively

slow part of the decay is caused by the dipole–dipole interaction among nuclear spins and reflects the primary decoherence process in the spin system. For the fast echo decay at the beginning of the relaxation process, the decay time T_{21} is shorter in the Ge-1% crystal in comparison with the Ge-7.76% crystal, and thus is not caused by the dipolar coupling. The reduction in T_{21} in the crystal with 1% of ^{73}Ge correlates with the broader line wings observed in this crystal, which are caused by the quadrupole interaction considered above. Therefore we conclude that the initial part of the echo decay is caused by the quadrupole interaction. Qualitatively, this effect may be described by a following manner. For nuclear spin $I = 9/2$, the frequency of the central transition $(-1/2 \leftrightarrow 1/2)$ is not shifted in the first order by the quadrupole interaction, while the frequencies of the other transitions are, and the satellite lines corresponding to transitions $m \leftrightarrow (m-1)$ with $m \neq 1/2$ appear on each side of the central line, yielding a broad envelope. The central transition is broadened by dipole–dipole interactions only and therefore shows a narrow line. In the time domain, these two components would cause two different decays.

To support the above conclusions, we carried out calculations of the Hahn echo decay. The interaction Hamiltonian of the ^{73}Ge nuclei ($I = 9/2$) is the sum of the interaction of nuclear spin with inhomogeneous magnetic field, quadrupolar and dipolar couplings, respectively:

$$H = -\sum_k \delta_k I_{Zk} + \sum_k \omega_{Qk}\left(I_{Zk}^2 - \frac{1}{3}I(I+1)\right) + \sum_{i>j} b_{ij}\left[I_{Zi}I_{Zj} - \frac{1}{4}\left(I_{+i}I_{-j} + I_{-i}I_{+j}\right)\right]. \quad (1)$$

Since the dipolar term does not commute with two other terms, the time evolution of the ensemble of the ^{73}Ge nuclei caused by different terms of the Hamiltonian can not be separated, making the calculation of the echo decays in analytic form impossible. Therefore, in the present work we use a simplified model neglecting the flip-flop terms in the dipolar Hamiltonian, which is a quite good approach for magnetically diluted systems and allows to simplify calculation and to receive physically reasonable data.

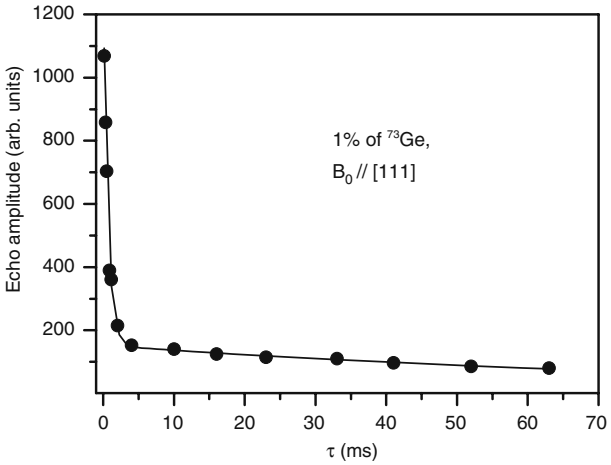

Fig. 3 ^{73}Ge Hahn echo decay envelope in Ge single crystal with 1% of ^{73}Ge isotope. External magnetic field is applied along the [111] axis. *Solid line* shows bi-exponential fit

Table 1 Experimental nuclear spin decoherence times T_{21} and T_{22} in Ge single crystals

	T_{21}, ms	T_{22}, ms
Ge-7.76%, B_0 // [111]	5.8	15.8
Ge-7.76%, B_0 // [110]	7.1	17
Ge-1%, B_0 // [111]	1.8	185
Ge-1%, B_0 // [110]	1.75	127

Accuracy is around 10–15%

The echo signal following a $\left(\frac{\pi}{2}\right)_0 - \tau - R - \tau^{/} - echo$ pulse sequence is written [3, 4] as

$$E\left(\tau^{/}, \tau\right) = \sum_{k,n} a_{nk} G_k\left(\tau^{/}\right) G_n\left(\tau\right), \tag{2}$$

where $a_{nk} = \frac{\left\langle k \mid RnR^{-1}\right\rangle}{\langle 0 \mid 0\rangle}$, the operator R describes the action of the second *rf* pulse and functions $G_k(t)$ form a system of the connected equations

$$i\dot{G}_k\left(t\right) = G_{k-1}\left(t\right) + v_k^2 G_{k+1}\left(t\right), \quad k = 0, 1, 2, \ldots, G_{-1}\left(t\right) = 0, \tag{3}$$

where $G_0(t)$ is the free induction decay that appears after the 90° pulse and the coefficients v_k^2 depend on the moments of NMR line. ^{73}Ge nuclei are randomly distributed over the sites of the magnetically diluted lattice, yielding the random distributions of the coupling constants δ_j, ω_{Qj} and b_{ij}. Suggesting these distributions to be Lorentzian,

$$f\left(\delta_j\right) = \frac{\Delta}{\pi}\frac{1}{\delta_j^2 + \Delta^2}, \quad f_Q\left(\omega_{Qj}\right) = \frac{\Delta_Q}{\pi}\frac{1}{\omega_{Qj}^2 + \Delta_Q^2}, \quad f_d\left(d_j\right) = \frac{\Delta_d}{\pi}\frac{1}{d_j^2 + \Delta_d^2} \tag{4}$$

where Δ, Δ_Q and Δ_d are the half widths at the half height of the Lorentzian distributions of the magnetic field inhomogeneity (δ_j), quadrupole (ω_{Qj}) and dipole–dipole (b_{ij}) interaction constants respectively, and $d_j = \sum_i b_{ij} I_{Zi}$, we calculated the

Hahn echo decay envelope as

$$E(2\tau) \approx 0.85 \exp(-2\tau/T_{21}) + 0.15 \exp(-2\tau/T_{22}), \quad (5)$$

where

$$T_{21} = \frac{1}{5\Delta_Q}, \quad T_{22} = \frac{1}{\Delta_d} \quad (6)$$

The amplitudes of two exponentials in Eq. 5 were fixed to 0.85 and 0.15 since the quadrupolar-perturbed spectrum of ^{73}Ge ($I = 9/2$) comprises four satellites with the total intensity 0.85 and the central line with the intensity 0.15.

The above expressions for the T_{12}'s and T_{22}'s reflect realistic physical picture of the spin-spin relaxation. They show that Hahn echo decay in Ge single crystals is caused by two different decoherence processes. The fast decay at the beginning of the relaxation process is caused by the quadrupole interaction. Then this process proceeds to slowly decaying, long-lived spin echoes that are caused by dipole–dipole interaction among nuclear spins. This slow decay may be elongated by means of spin dilution. Nowadays, semiconductor technology allows successful isotopic engineering and growing the pure monoisotopic ^{70}Ge crystal enriched up to the level of 99.99%, which means that the content of ^{73}Ge isotope is less than 0.01% [5]. Our findings show that in such a crystal decoherence time T_2 would be elongated up to ~20 s, which is quite an encouraging result for application of this material in nuclear spin-based quantum computers.

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