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Dynamic nuclear polarization of ^{29}Si nuclei in the isotope enriched n-type silicon

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Dynamic nuclear polarization of the ^{29}Si nuclei due to the "solid-effect" was observed clearly after saturation of the phosphorus electron paramagnetic resonance lines in three kinds of silicon crystals containing different amount of the ^{29}Si isotope (1 %, 4.7 %, and 99.3 %). Maximal enhancement (E) of the room-temperature ^{29}Si nuclear magnetic resonance signals $E \approx 605$ was obtained at the temperatures of 10–14 K in silicon containing 1 % of the ^{29}Si isotope.

1 Introduction The first experimentson the dynamic nuclear polarization (DNP) in silicon were performed by A. Abragam et al. [1]. It has been shown that in silicon containing phosphorus atoms at the concentrations lower than 5×10^{16} cm^{-3} the dominant mechanism of DNP is so called "solid-effect" [1–3]. This mechanism is realized under saturation of the forbidden "flip-flop" and "flip-flip" transitions in the dipole-dipole coupled electron nuclear system [2, 3]. The enhancement of the nuclear polarization $E = P_N/P_{N0} = 30$, where P_N is the DNP degree and P_{N0} is the equilibrium nuclear polarization, was obtained at 4.2 K under saturation of the forbidden transitions by microwave field at the frequency of 9 GHz [1]. The observed enhancement E is significantly lower than the maximal theoretical value $E_m = (\gamma_e/\gamma_N) = 3310$, where γ_e and γ_N are electron and ^{29}Si nuclear gyromagnetic ratios, respectively.

The low value of E can be explained by two factors. First, the long electron spin relaxation time $T_e \approx$ 1 – 10 s [4] of electrons localized at phosphorus atoms at 4.2 K reduces the value of E by factor $1/(1+f)$ [3] where $f = NT_e/nT^p_1$ is the leakage factor. Here N and n are the numbers of nuclei and paramagnetic centers, respectively, and T^p_1 is the nuclear spin lattice relaxation time under saturation of the forbidden transitions. The value of $T^p_1 = 3$ h determined in Ref. [1] for silicon doped with phosphorus at concentration of $n = 5\times10^{16}$ cm^{-3} allows us to estimate $f \approx 50$. The second factor reducing E is the broadening of the phosphorus electron paramagnetic resonance (EPR) lines due to the dipole-dipole interactions with the distant ^{29}Si nuclear moments. The forbidden "flip-flip" and "flip-flop" transitions, when the electron (m_S) and nuclear (m_N) spin projections are changed in opposite ($\Delta m_S + \Delta m_N = 0$) or in the same ($\Delta m_S + \Delta m_N = \pm 2$) directions, are shifted from the center of EPR line (B_0). This shift is equal to $\Delta B_\pm = B - B_0 = \pm B_0 (\gamma_N/\gamma_e)$. For ^{29}Si nuclei $\Delta B_\pm \approx \pm 0.1$ mT at $B_0 \approx 320$ mT. The width of the phosphorus EPR line in the naturally abounded (4.7 %) silicon is about 0.2 mT. In this case of the inhomogeneously broadened EPR line the DNP arises under saturation of the forbidden transitions corresponding to different spin packets

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at the same time and the "differential solid-effects" which orient ^{29}Si in opposite directions take place and reduces the DNP enhancement E [3].

In the present paper we report the results of the experiments on DNP in silicon crystals containing different amount of ^{29}Si isotopes. The effects of the temperature and illumination on DNP are discussed.

2 Experimental The experiments were performed with two types of isotopically controlled phosphorus-doped [$n(P) \approx 10^{15}$ cm^{-3}] silicon crystals having the ^{29}Si isotope concentrations 1 % and 99.3 %. The naturally abounded (4.7 % ^{29}Si) phosphorus doped silicon with $n(P) \approx 10^{15}$ cm^{-3} and 5×10^{16} cm^{-3} were also measured. The X-band EPR spectrometer was used for detection and saturation of the phosphorus EPR lines. The temperature of samples was controlled by an Oxford Instruments He gas flow cryostat in the range of 4 - 50 K. The DNP of the ^{29}Si nuclei were performed using the EPR spectrometer at the microwave power of 1 - 200 mW. The time of saturation (microwave irradiation), t, was changed between 10 min and 15 h. The 100 W halogen lamp was used for illumination of the samples. The long nuclear spin lattice relaxation time $T_1 > 1$ hr for all investigated samples at room temperature allows us to transfer the sample from EPR to the pulse nuclear magnetic resonance (NMR) spectrometers to detect the NMR signal at room-temperature.

3 Results and discussions Well known EPR spectrum of phosphorus donor atoms [4] was observed in all investigated silicon samples. This spectrum with the isotropic g-factor of 1.9985 consists of two hyperfine structure lines separated by 4.2 mT due to contact interaction between electron and phosphorus nuclear magnetic moments. The linewidth of the phosphorus EPR lines, ΔB_{pp} measured between maxima of the first derivative absorption line, is determined by the hyperfine interaction between electron localized on the phosphorus and ^{29}Si nuclear magnetic moments. The linewidths were $\Delta B_{pp} \approx 0.07$ mT, 0.2 mT, and 1.0 mT for the samples containing 1 %, 4.7 %, and 99.3 % of ^{29}Si isotopes, respectively.

The temperature dependence of the phosphorus EPR absorption line amplitudes observed in the temperature range of 4.2 - 30 K was similar for all the samples. The maximal signals were observed at the temperature range 10 - 14 K. The decrease of the EPR absorption signals by the two orders of magnitude at lower temperature ($T < 10$ K) is a result of the saturation of EPR signals related to the increase of the electron relaxation time T_e up to 1 - 10 s at 4.2 K for naturally abounded silicon [4]. The decrease of the EPR spectrum at higher temperatures $T > 14$ K is caused by thermal excitation of the electrons from the phosphorus donor level (E_c - 0.044 eV) to the conduction band. The decrease of T_e by a few orders of magnitude under the band gap illumination [4] increases the phosphorus EPR signals.

DNP of the ^{29}Si nuclei after saturation of the phosphorus EPR lines was observed in all the investigated samples. The dependences of the NMR signal amplitude on the magnetic field B near one of the phosphorus EPR lines in silicon containing the minimal (1 %) and maximal (99.3 %) amounts of ^{29}Si isotopes are shown in Fig. 1. The dependence of DNP on the deviation of the magnetic field from the center of EPR line in the 99.3 % abounded sample (see Fig. 1(b)) resembles the first derivative of EPR absorption on B. A similar dependence was observed in the naturally abounded silicon. These experiments show that DNP is a result of the "differential solid-effect" [1–3] when the EPR linewidth ΔB_{pp} exceeds the splitting $\Delta B_{\pm}$ between the forbidden "flip-flop" and "flip-flip" transitions. In the 1 % abounded silicon $\Delta B_{\pm} > \Delta B_{pp}$ and the maximal value of DNP were observed at the magnetic fields broader than the linewidth of the first derivative EPR line (see Fig. 1(a)) This result clearly confirms that the DNP in the silicon is due to the "solid-effect."

The typical temperature dependence of the degree of DNP P_N and of the nuclear polarization enhancement E obtained by extrapolation to the infinite time of saturation are shown in Fig. 2. The maximal values of P_N without illumination are achieved around $T \sim 10$ - 14 K. The temperature dependence of P_N correlates with the temperature dependence of the phosphorus EPR absorption line intensity. It was found that the band gap illumination at $T = 4.2$ K during the saturation of the EPR line increases P_N and E up to the values observed at 12 K without illumination. This is explained by shortening of the electron spin relaxation time T_e under illumination leading to the decrease of the leakage factor f [4].

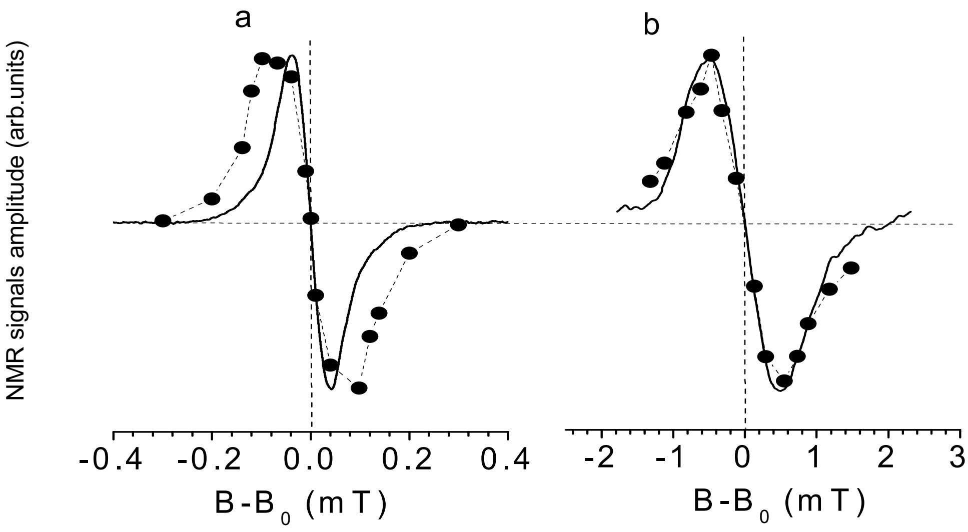

Fig. 1 Dependences of the amplitude of the ^{29}Si NMR signals (points) on the deviation of magnetic field B from the center of the phosphorus EPR line at B_0 = 326.1 mT (solid lines) in silicon crystals containing (a) 1 % and (b) 99.3% of the ^{29}Si isotopes after DNP at T = 12 K and the microwave power of 100 mW for the duration of 20 min. The positive sign of the DNP signal corresponds to the nuclear polarization opposite to the equilibrium ^{29}Si polarization.

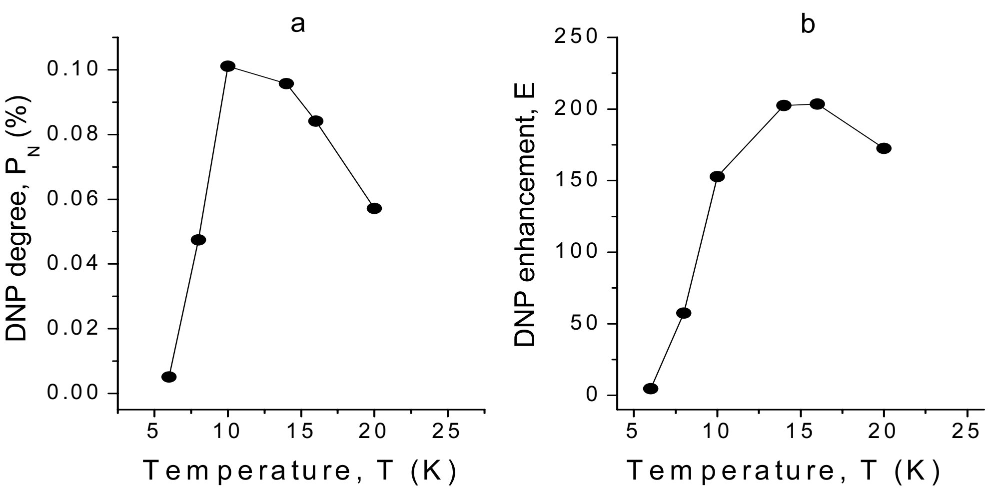

Fig. 2 Dependences of the (a) degree of ^{29}Si DNP, P_N, and (b) enhancement E on the temperature T in the naturally abounded silicon crystals.

The DNP degree $P_N \sim 0.1 - 0.35$ % corresponding to the enhancement of nuclear polarization $E \sim 200 - 600$ was observed in all investigated samples at $T = 12$ K The strongest DNP polarization $P_N \approx 0.35$ % and the enhancement $E \approx 605$ was obtained in silicon containing 1 % of ^{29}Si isotopes. In this sample the "solid-effect" was observed clearly (see Fig. 1(a)). It was found that the nuclear polarization time T^p_1 depends strongly on the phosphorus concentration N(P). At $T = 12$ K the time T^p_1 changes from 3.4 hr in the sample with N(P) $\approx 5\times10^{16}$ cm^{-3} to 16 h at N(P) $\approx 10^{15}$ cm^{-3}. No significant dependence of T^p_1 on the ^{29}Si abundance was observed.

4 Conclusions The DNP of ^{29}Si nuclei due to the "solid-effect" was observed in silicon containing 1 %, 4.7 %, and 99.3 % of ^{29}Si isotopes. It was found that DNP is more effective in the temperature range of 10 - 14 K. The band gap illumination increases the nuclear polarization at low temperatures $T < 10$ K.

At higher temperatures the DNP degree decreases following the decrease of the equilibrium electron spin polarization and phosphorus EPR signal intensity.

The well-resolved "solid-effect" was observed in the 1 % abounded silicon, while the "differential solid-effect" takes place in the naturally abounded and 99.3 % enriched silicon samples.

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