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Electron paramagnetic resonance and dynamic nuclear polarization of ^{29}Si nuclei in lithium-doped silicon

M.R. Rahman [a], L.S. Vlasenko [b], E.E. Haller [c], K.M. Itoh [a,*]

[a] School of Fundamental Science and Technology, Keio University, Yokohama 223-8522, Japan
[b] A.F. Ioffe Physico-Technical Institute, 194021 St. Petersburg, Russia
[c] Lawrence Berkeley National Laboratory and UC Berkeley, 1 Cyclotron Rd., Berkeley, CA 94720, USA

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ABSTRACT

Electron paramagnetic resonance (EPR) and dynamic nuclear polarization (DNP) experiments with Li-doped FZ silicon wafers are reported. The Li related EPR spectrum of tetrahedral symmetry was detected clearly without external stress even at low temperatures ($T<5$ K) implying that the Li electron spin-lattice relaxation time is much shorter than that of other shallow donors, e.g. phosphorus. The solid-effect was found to be responsible for the DNP and the polarization was enhanced by a factor of 87 at 3.2 K to reach 0.17%.

1. Introduction

Recently silicon-based quantum computing utilizing ^{29}Si nuclear spins as qubits has been proposed [1,2]. While ^{29}Si nuclear spins have been shown to have more than 25 s of coherence time, even at room temperature, [3] it is not straightforward to polarize all the nuclear spin qubits in the same orientation because of the small magnetic moment of the nuclear spins. Since application of a large magnetic field and lowering of temperature (e.g., $B>10$ T, $T<1$ K) alone cannot lead to 5% polarization needed for the initialization step of quantum computation, it is necessary to control the average distance between ^{29}Si nuclear spin qubits through isotope engineering [4–6] and utilized photons [7] and/or electron spin [8,9] to polarize nuclear spin qubits dynamically.

The DNP process transfers the equilibrium Boltzmann electron spin polarization P_{e0} at temperature T and magnetic field B to the nuclear spin by electron paramagnetic resonance (EPR) of paramagnetic centers that are in hyperfine contact with nuclear spins. The largest nuclear spin polarization enhancement obtainable is given by $E_{\max}=P_N/P_{N0}=\gamma_e/\gamma_N$ where P_{N0} is equilibrium Boltzmann nuclear polarization and γ_e and γ_N are the electron and nuclear gyromagnetic ratios, respectively [11,12]. For silicon the maximum enhancement ($E_{\max}$) is 3310.

The first experiments on DNP of ^{29}Si nuclei were performed by Abragam et al. [12] using phosphorus-doped silicon. A DNP enhancement of $E=30$ and a nuclear polarization of 0.048% were obtained under saturation of the phosphorus EPR lines in a microwave field of 9 GHz at 4.2 K [10]. Such low efficiency of DNP with phosphorus-doped silicon at low temperatures ($T<10$ K) is due to the long electron spin-lattice relaxation time $T_{1e}\sim1-10$ s [8]. The maximum value of the nuclear polarization $P_{N\max}$ that can be achieved by complete saturation of EPR transitions is; [13]

$$P_N=P_{e0}(1+f)^{-1}, \tag{1}$$

where

$$f=N_n T_{1e}/N_e T_1^p. \tag{2}$$

Here N_n is the number of nuclei interacting with one paramagnetic center, N_e is the number of paramagnetic centers, and T_1^p is the nuclear polarization time ranging from a few minutes to a few hours depending on the concentration of paramagnetic centers. When the electron relaxation time is short, $T_{1e}<N_eT_1^p/N_e$, the maximum nuclear polarization can be achieved when $f<1$. In this regard, lithium donors in silicon some of the most attractive candidates for effective DNP of ^{29}Si nuclear spins. In contrast to a phosphorus donor that has a non-degenerate singlet ground A_1 state, the five-fold degenerate ground state (doublet E+triplet T_2) of Li in Si [14,15] leads to shorter electron spin-lattice relaxation time than that for phosphorus. EPR investigations of Li in Si with externally applied stress have shown that Li prefers to occupy interstitial tetrahedral sites leading to an EPR spectrum with axial symmetry about the $\langle 100\rangle$ axis with $g_\parallel=g_{[100]}=1.9997$ and $g_\perp=1.9987$ [15].

2. Experimental

Experiments were performed with FZ silicon doped with Li by diffusion at 420 °C in vacuum for 15 min. The sample was quenched in alcohol immediately after annealing to prevent

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[*] Correspondence to: Department of Applied Physics and Physico-Informatics, Keio University, 3-14-1, Hiyoshi, Kohoku-Ku, Yokohama 223-8522, Japan.
Tel.: +81 45 566 1594; fax: +81 45 566 1587.
E-mail address: kitoh@appi.keio.ac.jp (K.M. Itoh).

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oxidation of Li. The concentration of shallow donors after diffusion was $\sim 1.0\times10^{16}\,\mathrm{cm}^{-3}$.

A *X*-band EPR spectrometer was used for the detection and saturation of the Li EPR lines. The temperature of the samples was controlled by an Oxford Instruments He gas flow cryostat in the range of 3.2–50 K. In order to determine the *g*-values of the observed EPR lines precisely, the EPR measurements were performed together with a Si:P reference sample having the well known isotropic *g*-value of 1.9985 ± 0.0001. The DNP of the ^{29}Si nuclei were performed using the EPR spectrometer for saturation of the EPR lines at the microwave power of 1–200 mW. The time of saturation, *t*, was varied between 10 min and 15 h. The long nuclear spin-lattice relaxation time $T_1 > 30$ min for all investigated samples at room temperature allows us to transfer the sample from EPR to the pulse nuclear magnetic resonance (NMR) spectrometers to evaluate the polarization of ^{29}Si nuclear moments.

3. Results and discussion

An EPR spectrum recorded without any externally applied stress at 3.2 K with $B\parallel\langle 1\,0\,0\rangle$ is shown in Fig. 1(a). It agrees very well with previously reported spectra for isolated Li [15]. The angular dependence of this EPR spectrum (Fig. 1(b)) shows axially symmetric g-tensors about $\langle 1\,0\,0\rangle$ with $g_\parallel = g_{[1\,0\,0]} = 1.9996\pm0.0001$ and $g_\perp = 1.9986\pm0.0001$ and they agree very well with previously reported values for Li atoms occupying tetrahedral interstitial positions [15].

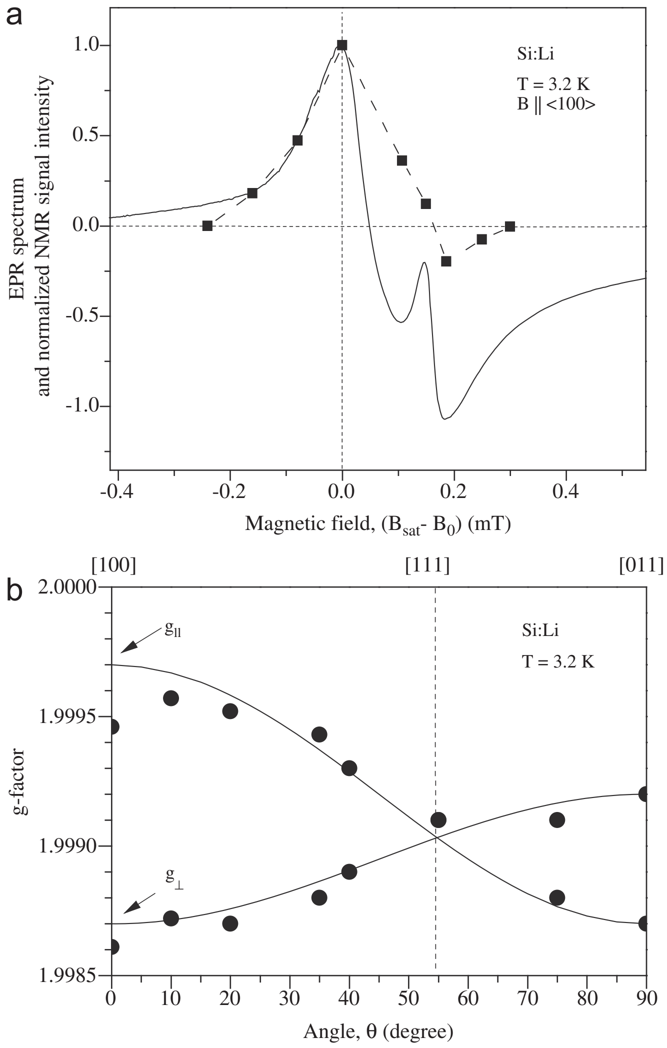

Fig. 1. (a) Solid curve shows the EPR signal of isolated Li measured with $B\parallel\langle 1\,0\,0\rangle$. Here B_{sat} is the saturation magnetic field and B_0 is the center magnetic field. Solid squares represent the normalized ^{29}Si NMR intensity after saturation of isolated-Li at $B_{\mathrm{sat}}-B_0$. (b) The angular dependence of EPR signals position where θ is the angle between the symmetry axis $\langle 1\,0\,0\rangle$ and applied magnetic field direction.

Strong enhancement of ^{29}Si NMR nuclei was observed after saturation of the Li EPR transitions. The dependence of the NMR signal amplitude on the magnetic field B_{sat} that has been used for the saturation in the EPR spectrometer is shown in Fig. 1(a) together with the first derivative shape of the EPR spectrum. The shape of the enhanced NMR signal represented by filled square in Fig. 1(a) resembles that of the EPR curve shown by the solid curve. This shows that the “differential” solid-effect [11–13] is responsible for the DNP of ^{29}Si nuclei.

Temperature dependences of the Li EPR intensity and ^{29}Si DNP degree are shown in Fig. 2(a). In contrast to the phosphorus EPR spectrum [16], the intensity of the Li EPR spectrum increases with decreasing temperature even below $T\approx 12$ K. This suggests that isolated interstitial Li atoms have a much shorter electron spin-lattice relaxation time than that of phosphorus. The temperature dependence of the ^{29}Si DNP degree correlates with the temperature dependence of the Li EPR spectrum.

The dependence of the DNP degree on the duration of the EPR saturation measured at $T = 3.2$ K and $B_0 = 323.16$ mT is shown in Fig. 2(b). The DNP degree extrapolated to infinite saturation time reaches the NMR signal enhancement of 87 corresponding to the polarization of 0.17% that is three orders better than that of thermal equilibrium. The nuclear polarization time (T_1^{p}) of ^{29}Si

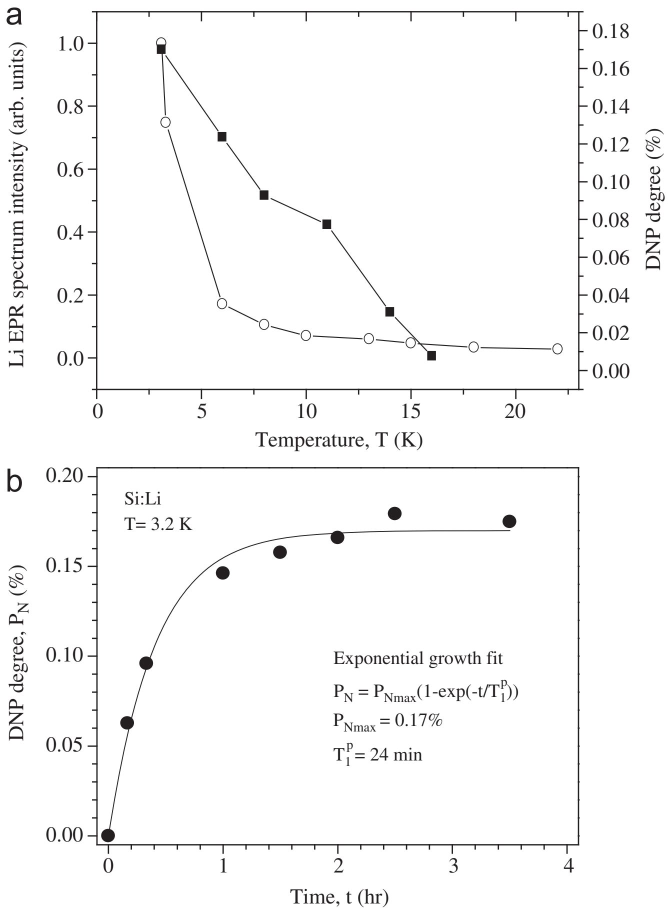

Fig. 2. (a) The temperature dependence of the Li EPR intensity (open circle) and the degree of polarization of ^{29}Si nuclei (square). (b) Dependence of the DNP degree on the time of EPR saturation. Solid line is the single exponential fit.

nuclei is 24 min, which is significantly shorter than 82 min obtained for DNP of ^{29}Si via phosphorus donors [16] for the same concentration of paramagnetic centers. Inhomogeneous broadening of the EPR line in the present sample does not allow for an efficient DNP through the pure solid effect. It is important to identify the origin of the inhomogeneous broadening to improve the efficiency of DNP via Li in Si.

4. Conclusions

The EPR spectrum observed in FZ silicon without external stress after diffusion of Li atoms was found to agree with the EPR spectrum observed earlier under uniaxial stress [15]. Temperature dependence of EPR and field dependence of NMR intensity shows that Li has a short enough electron spin relaxation time for efficient DNP at liquid helium temperature. It was shown that DNP is a result of the differential solid-effect. ^{29}Si nuclear polarization of 0.17% has been achieved via saturation of electron paramagnetic resonance transitions related to isolated lithium donors in silicon.

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