High Field ESR of P-doped Si for Quantum Computing Application

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Abstract. We measured ESR of phosphorous-doped silicon with a low concentration of P, \( n \), at high magnetic fields and low temperatures to investigate the states of nuclear spin. A sample with \( n = 6.52 \times 10^{16} / \text{cm}^3 \) was studied at 2.85 T (80 GHz) from 30 K to 2.3 K by field-modulating cw-ESR for a fixed 0 dB power. As the temperature was lowered, the out-of-phase signal appeared around 18 K, reached at a maximum intensity at 13 K, and disappeared around 6 K. The out-of-phase signal is referred to the field modulation. The in-phase signal started to change from the derivative of absorption spectrum at high temperatures to absorption-like shape around 15 K and asymmetry of intensity for two peaks of hyperfine-separated signals increased as temperatures was lowered. Below 10 K, the saturation of the in-phase signal started to appear. We speculate that the asymmetry is caused by saturation effect and dynamic nuclear polarization of \( ^{31} \text{P} \) nuclear spin due to drastic change of electron \( T_1 \).

1. Introduction
Among various quantum computing architectures, silicon-based nuclear spin system proposed by Kane [1] has been pursued most intensively because of its compatibility with classical computers. In the Kane’s model, the \( ^{31} \text{P} \) nuclear spins are used as qubits, and their interactions and measurements are achieved by manipulating electron clouds surrounding \( ^{31} \text{P} \) nucleus. In order to assure reversible interactions between nuclear and electron spins, the electrons should be in lowest energy-bound state and fully spin-polarized. Therefore, the Kane’s model requires low temperature and high magnetic field environments. We performed ESR of phosphorous-doped silicon, Si:P, at low temperatures and high magnetic fields to investigate the \( ^{31} \text{P} \) nuclear spin states.

The typical ESR spectrum of Si:P is consist of two side peaks separated by 4.2 mT and central peaks [2, 3]. The two side peaks originate from isolated P and the hyperfine interaction with \( ^{31} \text{P} \) nuclear spins shifts the peaks from the Larmor field. The central peaks originate from clusters of P-impurities. As the \( ^{31} \text{P} \) donor concentration, \( n \), is lowered, the side peaks
grow and the central peak intensity decreases. Most of ESR measurements have been done at low magnetic fields. Morigaki and Rosso [4], however, have measured ESR of Si:P with $n = 1.9 \times 10^{17} / \text{cm}^3 \sim 2.8 \times 10^{18} / \text{cm}^3$ at 100 GHz, from 1.4 K to 4.2 K. In their donor concentration range, the ESR spectra had a large central peak. They found asymmetry of the central peak grew with increase of the microwave power and the donor concentration. In quantum computing, the situation of localized electrons is more relevant. We used the Si:P sample with lower concentration than Morigaki’s in order to eliminate contributions from non-localized electrons, and investigated two side peaks even though we still had ESR spectrum from a small central peak.

2. Experimental details
The ESR measurements were performed in the temperature range from 2.3 K to 30.1 K at 80 GHz. The microwave was generated using a GUNN oscillator and its frequency was locked by using a vector network analyzer (AB Millimetre MVNA 8 – 350 GHz). The maximum microwave power at the output of the GUNN oscillator was 27 mW around 80 GHz. An attenuator was inserted between the GUNN oscillator and a waveguide. A sample of Si:P was put in the cylindrical waveguide at the bottom of the $^4\text{He}$ cryostat. The transmitted microwave power through the Si:P sample was measured by a cryocooled InSb detector at the outside of the cryostat. The ESR signals were obtained by the steady state method with magnetic field modulation. The magnetic field of 2.85 T was applied by a superconducting magnet and its sweep speeds of 1 mT/min and 0.3 mT/min were used. The magnetic field modulation was done by using a small coil wound on the waveguide, and its frequency and field were 16.0 kHz, and 13 $\mu$T. The modulation field was sufficiently smaller than the inhomogeneous broadening width of the ESR side peaks, 0.5 mT. We observed the in-phase and the 90° out-of-phase signals of the transmitted microwave power referred to the phase of the field modulation. Usually, the in-phase signal corresponded to the derivative of the power absorption spectrum. The phase adjustment was done at 20 K so as to eliminate the out-of-phase signal. The sample had a rectangular shape (3 mm $\times$ 3 mm $\times$ 237 $\mu$m). The donor concentration, $n$, was determined from

Figure 1. Temperature dependence of the ESR spectrum at the microwave power level of 0 dB. (a) the in-phase signals, (b) the out-of-phase signals, referred to the field modulation.
Figure 2. Temperature dependence of the signal intensities. ○: the in-phase signal (IN) of the lower magnetic field side peak (LFP), □: IN of the higher magnetic field side peak (HFP), ●: the out-of-phase signal (OUT) of LFP, ■: OUT of HFP. The dashed line corresponds to fitted normalized magnetization curve. (a) intensity vs $T$ plot, (b) intensity vs $1/T$ plot.

the room temperature resistivity.

3. Temperature dependence
With 0 dB microwave power level (corresponding to 0.6 mT microwave field intensity at the sample), we measured the ESR spectra of the in-phase and the out-of-phase signals as a function of temperature. There was no difference in the line shapes due to the sweep direction of magnetic field. We plotted the ESR spectra of the up-sweep in Figure 1. There were three temperature regions in the spectrum. In the high temperature region, $T > 16$ K, the in-phase signal behaved as the the derivative of the power absorption spectrum and there were no out-of-phase signals. In the intermediate temperature region, $16$ K $> T > 10$ K, the in-phase signal changed its shapes from the derivative to an absorption-like form and the signal intensity grew as the temperature was lowered. The out-of-phase signal appeared and reached to the maximum intensity which was comparable to the in-phase signal intensity and then disappeared. In the low temperature region, $T < 10$ K, the in-phase signal intensity became smaller and there were no out-of-phase signal.

Although the shapes of the signals changed with the temperature, we plotted temperature dependence of the ESR signal intensities in Figure 2. In the high temperature region, the peak to peak value was taken as the signal intensity. In the intermediate and low temperature regions, the peak values were taken. Since the expected shape was observed, we fitted data in the high temperature region to the normalized magnetization curve, $m(T) = \tanh(\mu_B H/k_BT)$. In the high temperature region, the intensity followed the Curie law well. In the intermediate temperature region, however, the in-phase signal intensity deviated from the curve, and in the low temperature region the intensity became smaller. Only in the intermediate temperature, the out-of-phase signal existed.

4. Power dependence
In order to investigate saturation effects, we measured the signal intensities as a function of the microwave power at various temperatures. Figure 3 shows the power dependence of the signal intensities at 3.0 K and 10.0 K. We could not observe the saturation effects in our measured microwave power range above 10.0 K. Therefore, the shape change from the high temperature region to the intermediate temperature region should not be caused by the saturation effects. Below 10.0 K, the saturation was observed at 5.2 K and 3.0 K. The saturation became larger as the temperature was lowered. If we corrected the saturation effect on ESR intensities, the intensities are smoothly connected to the values around $T \approx 10.0$ K within an experimental
Figure 3. Microwave power dependence of the ESR signal intensities of two hyperfine peaks. •:LFP, ○:HFP. The broken lines are results of fitting. The microwave power was normalized to the power with 0 dB attenuator. (a) power dependence at 10.0 K, (b) at 3.0 K.

accuracy and thus the drops of intensity below 10.0 K are related to the saturation effects. Although the saturation existed at 5.2 K and 3.0 K, there was no effect on the signal shapes and the out-of-phase signal.

5. Discussions
Although there were no measurement of $T_1$ at the high magnetic field, $T_1$ of Si:P below $n_{T_1} = 1 \times 10^{16} \text{ /cm}^3$ at the low magnetic field ($\sim 1 \text{ GHz}$), was measured to be $10^{-6} \text{ sec}$ at 20 K and $10^4 \text{ sec}$ at 1 K by several groups [5–7]. There was no donor-concentration dependence below $n_{T_1}$ [5]. Assuming that the measured donor-concentration dependence above $n_{T_1}$ is same for higher temperature and that there is no magnetic field dependence, $T_1$ for the sample with $n = 6.52 \times 10^{16} \text{ /cm}^3$ becomes of the order of $10^{-8} \text{ sec}$ at 20 K and $10^5 \text{ sec}$ at 1 K. Due to the strong dependence of $T_1$ on temperature, the passage condition, $\Omega_m \times T_1$, changes drastically with temperature, where $\Omega_m/2\pi$ is the modulation frequency of the field. We speculate the observed ESR spectrum change are caused by the transition from the slow passage, $\Omega_m \times T_1 \ll 1$, in the higher temperature region to the fast passage, $\Omega_m \times T_1 \gg 1$, in the lower temperature region.

As shown in Figure 2(a), both the in-phase and out-of-phase signals of the lower field peak below 15 K are larger by 20% than those of the higher field peaks. Even though we still don’t understand the ESR spectrum at low temperature, we believe that this phenomena is originated from dynamic nuclear polarization (DNP) due to the saturation of electron spin and long $T_1$ of the localized nuclear spin. If this is the case, the DNP would open up new possibilities of observing isolated nuclear spins directly.

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