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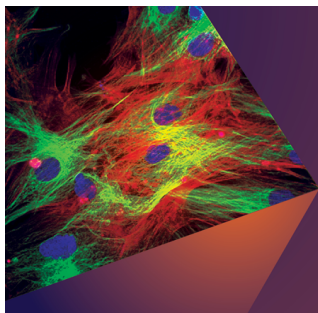
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Control of impurity incorporation into CVD diamond synthesized with *tert*-butylphosphine for quantum applications

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ABSTRACT

Nitrogen-vacancy (NV) centers in phosphorus-doped diamond have potential applications in quantum technologies because their electron spin has the longest spin coherence time among those in solid systems at room temperature (RT) and because their negative charge state is more stable than that in undoped diamond. However, the conventional phosphorus source, phosphine, is toxic and explosive; a safer precursor is therefore preferred. We consequently attempted to synthesize phosphorus-doped diamonds using *tert*-butylphosphine (TBP), which has lower toxicity and explosivity than phosphine. However, controlling phosphorus incorporation during doping with TBP is difficult because it condenses easily and remains in the synthesis reactor due to its low boiling point. Here, we report controlling the incorporation of impurities by optimizing the synthesis conditions, such as the synthesis pressure. Comparing diamonds synthesized at 15 and 50 kPa reveals that the phosphorus concentration was reduced by 93% and the nitrogen concentration by 72% in the diamond synthesized at 50 kPa. For diamonds prepared under optimized conditions in our synthesis reactor, a long spin coherence time ($T_2 = 2.23$ ms) for NV centers with almost perfect selective alignment of the NV axes was achieved. The T_2 was comparable to the longest reported length for NV centers in a sample synthesized with phosphine, indicating that TBP is a promising phosphorus source for obtaining NV centers with excellent properties. We also achieved high electron mobility [$580 \text{ cm}^2/(\text{V s})$ at RT] in diamonds synthesized with TBP; this value is comparable to that for diamonds synthesized with phosphine.

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Quantum technologies based on nitrogen-vacancy (NV) centers in diamonds have been extensively investigated and are expected to be applied to quantum sensors and quantum information devices.^{1–7} One technique for producing NV centers with excellent properties is diamond synthesis using chemical vapor deposition (CVD). This technique enables good control of impurities⁸ and can produce *n*-type diamonds.⁹ In addition, the CVD technique is expected to enable the synthesis of large diamonds.¹⁰

Phosphorus doping in *n*-type diamonds has been reported to lead to long spin coherence times¹¹ and stabilization of the negative charge state of NV centers.^{12–15} The spin coherence times are the longest ones reported among solid-state electron spin at room

temperature (RT).¹¹ Intriguingly, long coherence times are achieved in diamonds with semiconducting properties. The mechanism of long NV spin coherence in donor-doped^{11,13} and acceptor-doped¹⁶ diamonds has been attributed to the reduction of magnetic noise sources such as vacancy complexes and vacancy-related defects by Coulomb repulsion between the charged vacancies and impurity defects. Increasing the coherence times in *n*-type semiconductor diamonds is important for developing diamond-based electronic devices for quantum information science and quantum sensing.^{17–22} Another unique feature of the CVD technique is the selective alignment of the NV centers along the [111]-axis, which improves the readout contrast of ensemble NV centers.^{23–25} This selective alignment increases the

magnetic field sensitivity of quantum sensors and enhances the read-out efficiency of the quantum devices.

As the aforementioned reports show, *n*-type diamonds synthesized by the CVD technique are important for obtaining NV centers with excellent properties. Diamonds with *n*-type conductivity, as indicated by Hall measurements,²⁶ have only been achieved by phosphorus doping in CVD synthesis, and phosphine is used as the conventional doping gas. However, phosphine is both highly toxic and explosive, preventing the introduction of *n*-type diamond synthesis to research laboratories and increasing the cost of the equipment. By contrast, *tert*-butylphosphine (TBP) is less toxic and less explosive than phosphine. It is used as a source for phosphorus doping and enables the synthesis of *n*-type diamonds without costly detoxification equipment.²⁷ However, TBP easily condenses and adheres to the reactor wall because of its low boiling point (56.1 °C), making the amount of phosphorus incorporated into *n*-type diamond difficult to control. In our previous study,²⁸ the phosphorus concentration was $\sim 10^{17} \text{ cm}^{-3}$ even without TBP addition and the phosphorus doping level could not be controlled at low phosphorus concentrations. This lack of control made finding the appropriate concentration for a longer spin coherence time (T_2) and attaining a high electron Hall mobility difficult.

In the present study, we report that the amount of impurities incorporated into *n*-type diamond was reduced with increasing CVD synthesis pressure. In particular, the amount of phosphorus incorporated could be substantially reduced even when residual TBP was present in the growth environment, indicating that this method is effective for controlling the incorporation of impurities in *n*-type diamond. In addition, we achieved long T_2 for NV centers in samples synthesized with favorable impurity concentrations. The T_2 is comparable to the longest one reported for a sample synthesized with phosphine,¹¹ thus demonstrating that TBP is a promising phosphorus source gas for attaining excellent coherence. We also investigated the NV axis orientation and carried out Hall effect measurements to characterize the electrical properties of the phosphorus-doped diamonds prepared using TBP.

The diamond samples were epitaxially grown on Ib-type (111) diamond substrates using a microwave-plasma-assisted chemical vapor deposition (MPCVD) system. Diamond substrates with dimensions of $2 \times 2 \times 0.5 \text{ mm}^3$ were prepared via the high-pressure-high-temperature (HPHT) method. The substrate surfaces were polished by

chemical-mechanical polishing with a misorientation angle of 1° – 2° . The substrate temperature was maintained at 900 °C by adjusting the microwave power between 750 and 950 W. The pressure during synthesis was varied in each synthesis. The source gas consisted of hydrogen, ^{12}C -enriched methane, and TBP. Even without the addition of TBP, phosphorus incorporation occurred from the residual TBP in the reactor because TBP is easily condensed because of its high boiling point. The growth conditions are listed in Table I. The film thicknesses of samples for SIMS measurement are about 7–10 μm , and that for NV center measurement are about 2–10 μm . After growth, samples were cleaned with mixed acid ($\text{H}_2\text{SO}_4\text{:HNO}_3 = 3\text{:}1$) at 150 °C for 30 min. The impurity concentration was measured by secondary-ion mass spectrometry (SIMS). The background level of each SIMS measurement varies depending on the vacuum level, the cleanliness of the SIMS equipment, and the etching rate. Therefore, when measuring the low nitrogen concentration, we etched only a few micrometers from the sample surface at a fast rate to suppress the background level as low as possible. The actual background level for each measurement is shown in Fig. S1 in the [supplementary material](#).

The surface morphology of the samples was observed with an optical microscope. The photoluminescence (PL) from the NV centers was measured at room temperature using a custom-built confocal microscope system. The NV centers were excited with a laser ($\lambda = 532 \text{ nm}$), and PL was detected using an avalanche photodiode. We obtained PL imaging maps to estimate the NV concentration and counted the number of single NV centers. The estimation method is detailed in the [supplementary material](#). Microwaves were applied via a thin copper wire fabricated on the sample surface. A static magnetic field ($B \sim 5.8 \text{ mT}$) was applied along the [111] direction using a permanent magnet.

The spin properties of the NV centers were characterized via optically detected magnetic resonance (ODMR). The NV axis orientations were confirmed by the peak splitting width in the ODMR spectrum, which is governed by the strength of the magnetic field component parallel to the NV axis. We applied the static magnetic field in the [111] direction and could therefore judge whether the NV axis was oriented along the [111] direction or along one of the other three directions ($[\bar{1}\bar{1}\bar{1}]$, $[\bar{1}\bar{1}1]$, or $[\bar{1}1\bar{1}]$). The T_2 was measured using the Hahn echo method. Two types of signals ($S_{|0\rangle}$ and $S_{|1\rangle}$) were obtained with two different pulse sequences ($\pi/2, \pi, \pi/2$ and $\pi/2, \pi, 3\pi/2$,

TABLE I. Growth conditions, nitrogen and phosphorus concentration, and longest T_2 for each sample. To obtain actual impurity concentrations, the background level should be subtracted from the measured value shown in Fig. 1; the values after subtraction are shown in this table.

Sample No.	Total gas flow rate (sccm)	CH ₄ /H ₂ (%)	TBP/CH ₄ (ppm)	Pressure (kPa)	[N] (cm ⁻³)	[P] (cm ⁻³)	Longest T_2 (μs)
15A	400	0.25	0	15	N/A	3.9×10^{17}	930 ± 60
25A	400	0.25	0	25	N/A	N/A	1620 ± 100
40A	400	0.25	0	40	N/A	N/A	1130 ± 60
50A	400	0.25	0	50	2.1×10^{15}	1.1×10^{15}	1790 ± 70
50B	400	1.0	0	50	1.2×10^{15}	4.6×10^{14}	2230 ± 80
50C	400	2.0	0	50	N/A	N/A	2060 ± 70
15P	400	0.25	1100	15	1.5×10^{16}	3.9×10^{17}	N/A
25P	400	0.25	1100	25	1.1×10^{16}	1.9×10^{17}	N/A
40P	400	0.25	1100	40	6.2×10^{15}	3.5×10^{16}	N/A
50P	400	0.25	1100	50	4.2×10^{15}	2.7×10^{15}	N/A

respectively), and they were subtracted as $(S_{|0\rangle} - S_{|1\rangle}) / (S_{|0\rangle} + S_{|1\rangle})$ to remove the common-mode noise. Echo signals were normalized by referring to the Rabi contrast and were fitted with decay curves ($\exp(-(\tau/T_2)^n)$) to estimate T_2 . Each NV center exhibits a different T_2 depending on the surrounding impurity distribution, necessitating numerous Hahn echo measurements at different NV centers. However, Hahn echo measurements for T_2 longer than 1 ms are time-consuming; thus, measuring several dozen NV centers is impractical. We therefore preselected an NV with a relatively long T_2 by comparing numerous NV centers using less-precise but short measurements; the details of this method are described in the [supplementary material](#).

Hall effect measurements were performed to confirm the n -type conductivity. To achieve a reliable contact with low resistance, heavily phosphorus-doped n^+ -type diamond with a doping concentration of $\sim 2 \times 10^{20} \text{ cm}^{-3}$ was selectively grown between n -type diamond and Ti contacts.²⁹ A Ti metal mask was fabricated on the diamond surface by conventional photolithography and liftoff processes, and n^+ -type diamond was grown by MPCVD on the patterned surface morphology of the van der Pauw configurations. A different CVD reactor was used for doping to achieve n^+ -selective growth to avoid the memory effect of phosphorus doping. Contacts with the structure Ti (30 nm)/Pt (30 nm)/Au (100 nm) were deposited onto diamond by electron-beam evaporation followed by thermal annealing at 420 °C for 30 min.

We investigated the effect of pressure during CVD synthesis on the incorporation of impurities in an environment where TBP remained on the reactor wall. [Figure 1\(a\)](#) shows the synthesis pressure dependence of the impurity concentrations of phosphorus, nitrogen, and hydrogen for samples grown under flowing TBP (15P, 25P, 40P, and 50P) and without flowing TBP (15A and 50A). The dotted lines in [Fig. 1\(a\)](#) indicate the background levels of each element to show that the obtained impurity concentrations are above the background levels, and the raw data from the SIMS measurements are plotted. The actual impurity concentration is obtained by subtracting the background level from the plotted raw SIMS data. Note that the actual background level in the case of the low concentration of nitrogen is lower than it, as shown in [Fig. S1](#) in the [supplementary material](#), and the estimated concentration is shown in [Table I](#). In the samples grown at 15 kPa, the phosphorus concentrations were found to remain the same irrespective of the TBP flow, indicating that the residual TBP has a greater contribution to phosphorus incorporation than the flowing TBP. At higher pressures, the impurity concentration decreased. The impurity concentrations in the sample synthesized at 50 kPa under flowing TBP were reduced by 79% for hydrogen, 72% for nitrogen, and 93% for phosphorus compared with those in the sample synthesized at 15 kPa. In the absence of the TBP flow, we found that the phosphorus concentration was reduced to $1 \times 10^{15} \text{ cm}^{-3}$ in the sample synthesized at 50 kPa (50A), which is more than two orders of magnitude lower than the phosphorus concentration in the samples synthesized at 15 kPa. From these results, we found that the unintentional incorporation of phosphorus due to the residual TBP could be substantially reduced by increasing the pressure. High synthesis pressures did not affect the surface quality. [Figures 1\(b\)](#) and [1\(c\)](#) shows optical images of the surface of the sample synthesized at 50 kPa, showing a good surface condition without nucleation or cracks.

We speculate that the higher pressure reduces the unintentional incorporation of impurities because the ratio of the pressure of the impurities to the total pressure is relatively decreased when the

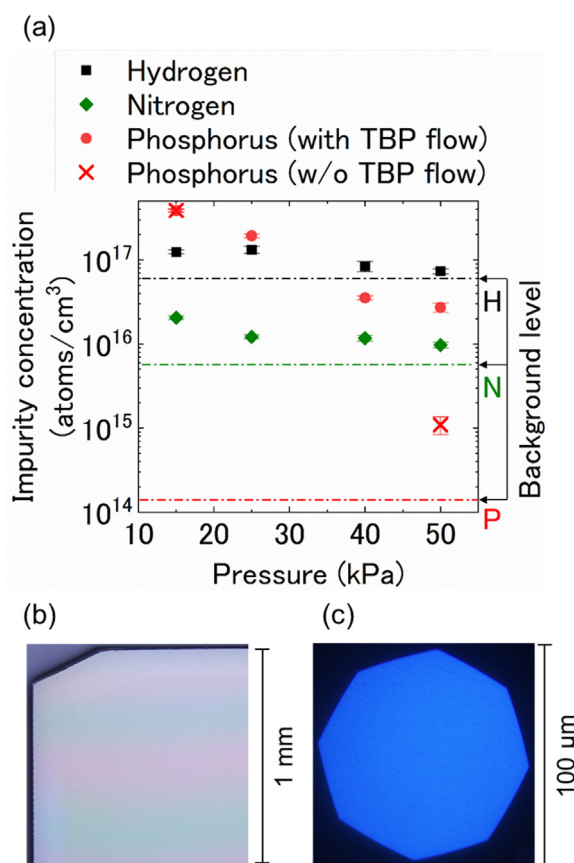


FIG. 1. (a) Impurity concentration of samples synthesized at different pressures. The plots show the raw data of the SIMS measurements. The red circles (red filled circle) and the crosses (red cross) represent phosphorus with and without TBP flow, respectively. The green rhombuses (green filled diamond) and the black squares (■) represent nitrogen and hydrogen, respectively. The actual impurity concentrations are obtained by subtracting the background level, which is represented by the dotted lines from the plotted raw data. Note that the actual background level in the case of the low concentration of nitrogen is lower than it, as shown in [Fig. S1](#) in the [supplementary material](#), and the estimated concentration is shown in [Table I](#). The samples were synthesized with the following gas ratio: CH_4/H_2 0.25%, TBP/CH_4 1100 ppm in the case of with TBP flow. (b) and (c) Optical images of the sample synthesized at 50 kPa [(b) 10 $\times$ and (c) 100 $\times$ objectives with a Nomarski prism].

pressure of each impurity is constant. When the synthesis pressure is varied from 15 to 50 kPa, the vapor pressure of the residual TBP is almost constant; it varies by only $\sim 1\%$ (details are provided in the [supplementary material](#)). If nitrogen is incorporated from leakage, its partial pressure is speculated to be constant or lower at higher total pressures. Therefore, under a higher total pressure, the ratio of the partial pressure of the leakage nitrogen is reduced, along with the amount of nitrogen incorporated.

To investigate phosphorus incorporation at varied pressures, we performed optical emission spectroscopy and attempted to detect the signal from the active phosphorus species such as PH and PH_2 radicals, but we could not observe any signal from them in our equipment. Previously, Lobaev *et al.* detected the PH radical peak with a high-resolution spectrometer in a diamond synthesis environment, where

strong emission from hydrogen and methane is observed, and they showed that the PH radical peak decreased with increasing pressure at 50–100 Torr, which indicates that the phosphorus incorporation was suppressed by increasing the pressure.³⁰ Our synthesis conditions differed from those in this paper,³⁰ so we cannot conclude whether the same phenomena occurred; however, if the active phosphorus species decreased with increasing pressure, this may be one of the mechanisms explaining our observation. We also speculate that the plasma ball's size may depend on the pressure, where a higher pressure results in a smaller plasma ball on the diamond sample. The smaller plasma ball can suppress the temperature rise of the reactor wall around the plasma ball, which may in turn reduce the volatilization of the residual TBP. The plasma density also increases under higher pressure; thus, the reactions in the plasma may be affected.

Figure 2(a) shows the NV concentrations in the samples synthesized under various pressures. When the synthesis pressure was increased, the NV concentration and the nitrogen concentration decreased; thus, the conversion efficiency ($[NV]/[N]$) was almost constant with respect to the synthesis pressure. When the methane flow rate was increased to further reduce the nitrogen incorporation, the NV concentration was substantially reduced [Fig. 2(b)]. In the samples synthesized with 0.25% and 1% methane ratios, the NV conversion efficiency was almost constant [i.e., $(3\text{--}7) \times 10^{-5}$]. However, in the sample synthesized with a 2% methane ratio, the NV concentration was so low that we did not find an NV in a $30 \times 30 \mu\text{m}^2$ area [Fig. 2(b)]. This observation corresponds to an NV concentration of less than 10^9 cm^{-3} . The nitrogen concentration

in this sample was below the background level of the SIMS measurement ($6.5 \times 10^{15} \text{ cm}^{-3}$).

Next, we investigated the T_2 for the NV electron spins with the Hahn echo sequence shown in Fig. 3(a). In our samples, most NV centers have a T_2 longer than 1 ms. Therefore, we preselected the NV centers with a relatively long T_2 using a simplified method described in the supplementary material to compare numerous NV centers. As a result, we observed the longest T_2 for the sample synthesized at 50 kPa and 1% methane flow (50B), which was estimated to be $2.23 \pm 0.08 \text{ ms}$ [Fig. 3(b)]. This T_2 is comparable to the longest T_2 reported for an NV center at RT.¹¹ In our synthesis, impurity incorporation was reduced by increasing the synthesis pressure; thus, it is reasonable that a long T_2 was attained for the sample synthesized under high pressure.

Bauch *et al.* reported the T_2 limitation by the electron spin bath of substitutional nitrogen (P1 center).³¹ We plot our result in Fig. 3(c) to compare it. The blue crosses correspond to the results obtained with the samples synthesized at 50 kPa. This figure also includes our previously reported results for samples synthesized at 25 kPa²⁸ (orange crosses) and shows the nitrogen spin-bath contribution of NV centers to T_2 (straight line).³¹ The T_2 s obtained in this research are longer than those previously reported^{28,31} and deviate from the straight line corresponding to the P1 spin-bath limitation.³¹ Hence, our results indicate that the P1 spin bath is no longer the main source of decoherence; that is, some defects or impurities other than the P1 center caused decoherence in the samples synthesized at 50 kPa. Bauch *et al.* quantitatively investigated the causes of decoherence by fitting the data points according to the following equation:

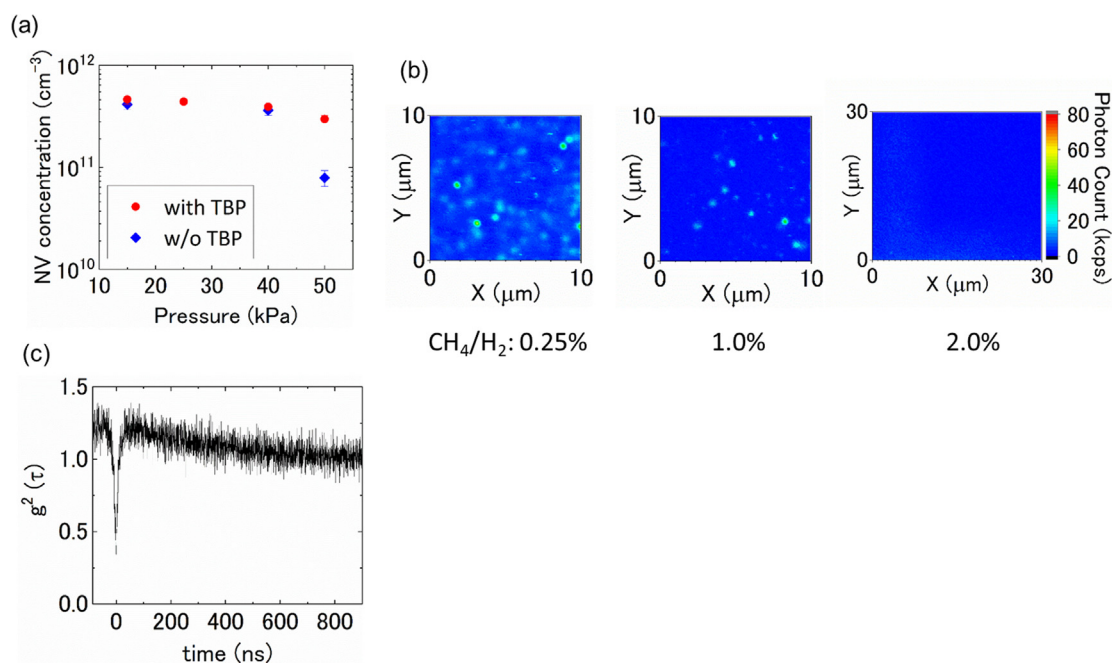


FIG. 2. (a) NV concentration of samples synthesized at different pressures. At higher pressures, the NV concentration was also reduced along with the nitrogen concentration, and the NV conversion efficiency was almost constant ($[NV]/[N] = (2\text{--}3) \times 10^{-5}$). The NV concentration was estimated by counting the number of NV centers in the PL scanning images. The samples were synthesized with the following gas ratio: CH₄/H₂ 0.25%, TBP/CH₄ 1100 ppm in the case of with TBP flow. (b) PL scanning images of samples synthesized with different methane flow ratios. The synthesis pressure was 50 kPa, and the TBP flow rate was 0 sccm. (c) Second-order autocorrelation function of the photons emitted by a single NV center located in the synthesized CVD film.

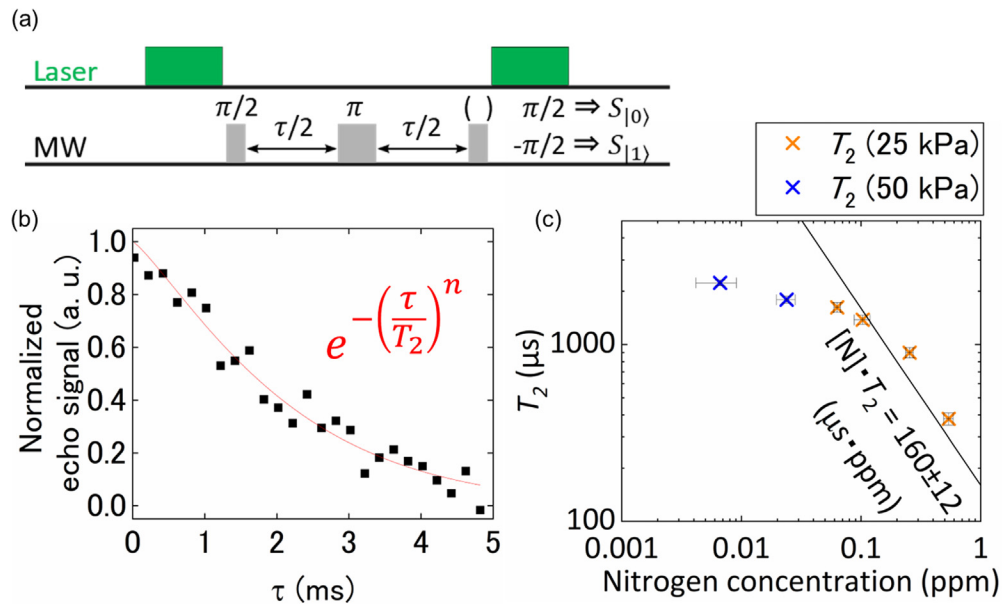


FIG. 3. (a) Pulse sequence for a Hahn echo measurement. We used a $\pi/2$ ($-\pi/2$) pulse for the last pulse to rotate the spins to the $|0\rangle$ ($|1\rangle$) state and read out the spin states $S_{|0\rangle}$ ($S_{|1\rangle}$). (b) The result of Hahn echo measurements, showing the longest T_2 obtained in this research. The red line is the fitting curve, and T_2 was estimated to be 2.23 ms. Common-mode noise was removed by subtracting $S_{|1\rangle}$ from $S_{|0\rangle}$. The signal was normalized by the Rabi contrast. (c) Correlation between the nitrogen concentration and T_2 in each sample. The line shows the limitation of T_2 by the nitrogen concentration, as reported by Bauch *et al.* For samples synthesized at 50 kPa, each plot clearly deviates from this line. This result indicates that the nitrogen concentration can be sufficiently reduced in the sample synthesized at 50 kPa and that the nitrogen spin bath is not the determining factor for T_2 .

$$\frac{1}{T_2([N])} = B_{NV-N} \times [N] + \frac{1}{T_{2,other}}, \quad (1)$$

where B_{NV-N} is the nitrogen-dominated NV decoherence rate per unit density, and $T_{2,other}$ is the decoherence parameter due to defects other

than nitrogen. We also obtained these parameters by fitting the results in Fig. 3(c), obtaining $1/B_{NV-N} = 340 \pm 46 \mu\text{s ppm}$ and $T_{2,other} = 2.24 \pm 0.10 \text{ ms}$ ($1/B_{NV-N} = 160 \pm 12 \mu\text{s ppm}$ and $T_{2,other} = 694 \pm 82 \mu\text{s}$ in the report of Bauch *et al.*³¹). The longer $T_{2,other}$ indicates that the

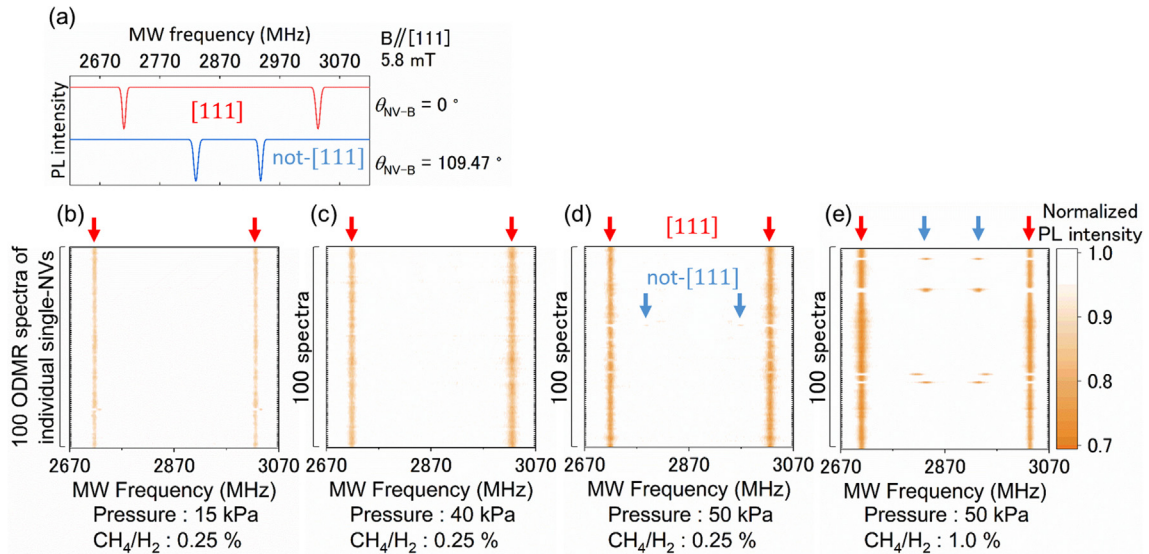


FIG. 4. (a) Simulation of ODMR spectra under a static magnetic field in the [111] direction. The upper red spectrum corresponds to the case where the NV axis is oriented in the [111] direction, and the lower blue spectrum corresponds to the case where the NV axis is oriented in three directions other than [111]. (b)–(e) ODMR spectra obtained from 100 individual single-NVs in samples 15A, 40A, 50A, and 50B, respectively; almost all of the spectra correspond to [111]-directed NV, and the NV axes were confirmed to be highly aligned in all of the samples. ODMR measurements were performed under a 5.8 mT static magnetic field oriented in the [111] direction.

amount of defects other than nitrogen could be reduced. The B_{NV-N} did not correspond with high accuracy because our data were obtained only at low nitrogen concentrations and over a narrow range of concentrations, where the nitrogen spin bath is not dominant.

We acquired the ODMR spectra of the NV centers in some samples to confirm that the NV axes are preferentially oriented in the [111] direction, which is important for quantum applications^{23–25} and provides insights into the growth mechanism.³² Figure 4(a) shows the ODMR spectra simulated under the assumption that a static magnetic field of 5.8 mT is applied in the [111] direction as well as in the actual experimental condition. The upper red spectrum corresponds to the NV oriented in the [111] direction, and the lower blue spectrum corresponds to those oriented in the other three directions. Figures 4(b)–4(e) shows the ODMR spectra acquired experimentally. Each figure includes spectra of 100 single NV centers found in each sample, and these spectra are stacked vertically. The samples synthesized at a 0.25% methane flow rate (15, 40, and 50 A) showed almost perfect alignment of the NV axes in the [111] direction. In the sample with the longest T_2 in this study (50B),

increasing the methane flow rate slightly decreased the alignment ratio; however, it remained at a high level (95%). These results indicate that the step-flow growth was maintained under the conditions used in the present study.³²

We carried out a Hall effect measurement to characterize the electrical properties of the phosphorus-doped diamond samples. We measured the Hall mobility of the sample synthesized at 15 kPa (15A), which exhibits the highest phosphorus concentration and the lowest sheet resistance among the prepared samples. Figures 5(a) and 5(b) show the electron concentration and mobility results at each temperature. The temperature dependence of the electron concentration is plotted in Fig. 5(a), and the donor level E_d determined from the slope of the fitting line is close to the standard value of 0.58 eV for phosphorus-doped diamond. As shown in Fig. 5(b), the mobility was higher at lower temperatures, and a mobility of 581 $\text{cm}^2/(\text{V} \cdot \text{s})$ was obtained at RT, which is the highest electron mobility reported for a diamond synthesized with TBP.³³ The mobility decreases because of acoustic phonon scattering ($\mu \propto T^{-3/2}$) at temperatures less than 500 K. Furthermore, it is approximately the same as the previously

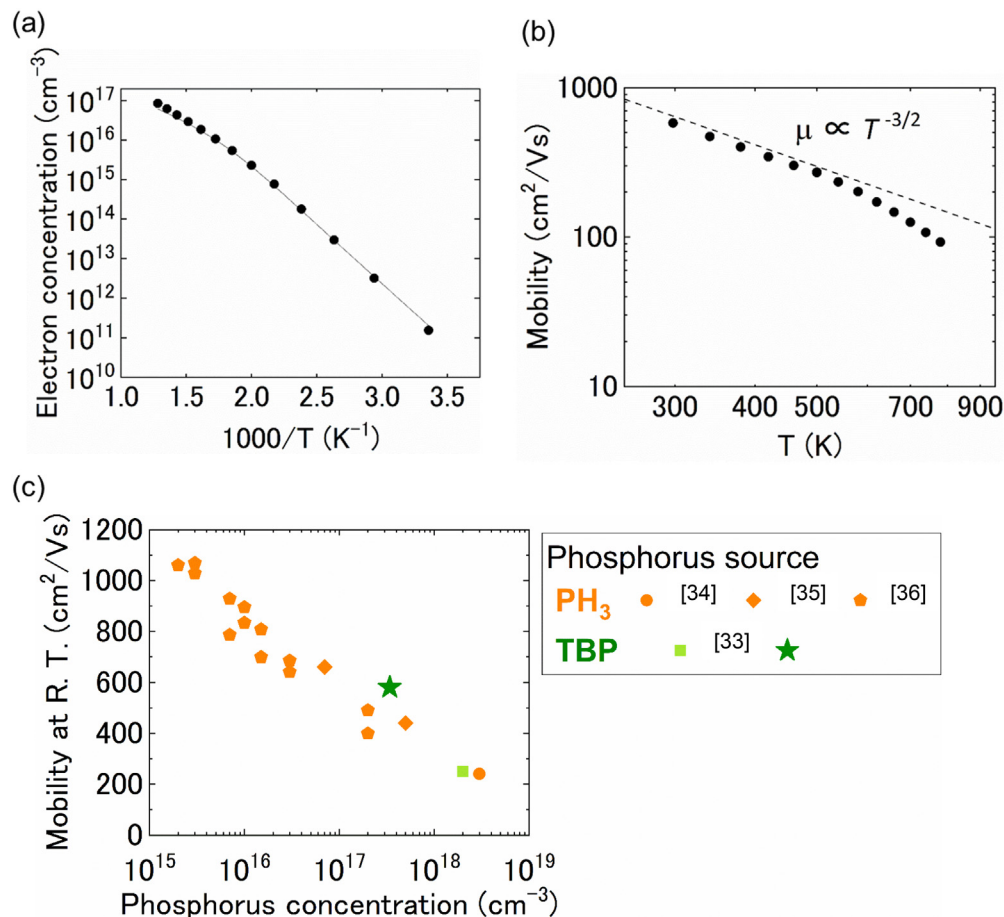


FIG. 5. (a) Temperature dependence of the electron concentration. We estimated the activation energies from the slope of the linear line and confirmed that these values approximately correspond to the donor level of phosphorus. (b) Temperature dependence of the electron mobility. The mobility was dominated by the acoustic phonon scattering ($\mu \propto T^{-3/2}$) at temperatures less than ~ 500 K. (c) Correlation between the phosphorus concentration and electron mobility at room temperature. The mobility against phosphorus concentration obtained in this study ($\star$) is comparable to previous results ($\bullet$, $\blacklozenge$, $\blacksquare$, $\blacklozenge$)^{33–36} obtained in samples synthesized with phosphine and TBP.

reported values^{33–36} for diamond with a similar phosphorus concentration achieved using phosphine as a doping gas [Fig. 5(c)].

The T_2 for sample 15A, which exhibited high mobility, was shorter than the longest T_2 in our research. This result suggests that the magnetic noise that dominates T_2 was not sufficiently removed compared with the noise for sample 50B. However, in the samples that achieve the longest T_2 , the carrier density is not sufficiently high for the mobility to be measured. Achieving high mobility at low doping levels has been challenging, even in research using phosphine, because a small amount of impurities or defects severely affects mobility.^{35,36} In the next step, it will be important to systematically investigate the synthesis conditions that achieve both ultra-long T_2 and high mobility.

To advance quantum technologies based on the use of NV centers, it is important to precisely control the incorporation of impurities and improve the coherence of NV centers. CVD synthesis of phosphorus-doped diamond using TBP has advantages in terms of safety and cost; however, controlling the phosphorus incorporation is difficult. In the present research, we investigated the effect of synthesis pressure on impurity incorporation under an environment with residual TBP. Increasing the synthesis pressure reduced impurity incorporation, particularly unintentional phosphorus incorporation from the residual TBP. We obtained a T_2 of 2.23 ms with a single NV center in the synthesized sample. This length is comparable to the best reported length (2.43 ms) obtained for a sample synthesized with phosphine as the phosphorus source. We also conducted Hall effect measurements and obtained the highest reported electron mobility [580 cm²/(V s) at RT] among the samples synthesized with TBP, which is comparable to those synthesized with phosphine in similar phosphorus concentrations. These results indicate that the synthesis of n-type diamond using TBP is promising and shows good potential for enabling the formation of NV centers with excellent coherence properties and good semiconductor properties for quantum applications.

See the [supplementary material](#) for additional details related to this work.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Riku Kawase: Formal analysis (equal); Investigation (lead); Writing – original draft (lead). **Hiroyuki Kawashima:** Formal analysis (equal); Investigation (equal); Validation (equal); Writing – review & editing (supporting). **Hiromitsu Kato:** Formal analysis (supporting); Investigation (supporting); Validation (equal); Writing – review & editing (supporting). **Norio Tokuda:** Formal analysis (supporting); Investigation (supporting); Validation (supporting). **Satoshi Yamasaki:** Project administration (supporting); Validation (supporting); Writing –

review & editing (supporting). **Masahiko Ogura:** Formal analysis (supporting); Validation (supporting); Writing – review & editing (supporting). **Toshiharu Makino:** Formal analysis (supporting); Validation (supporting); Writing – review & editing (supporting). **Naoya Morioka:** Formal analysis (supporting); Investigation (supporting); Validation (supporting); Writing – review & editing (supporting). **Norikazu Mizuochi:** Conceptualization (lead); Funding acquisition (lead); Supervision (lead); Validation (lead); Writing – review & editing (lead).

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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