

Coherent Control and Magnetic Detection of Divacancy Spins in Silicon Carbide at High Pressures

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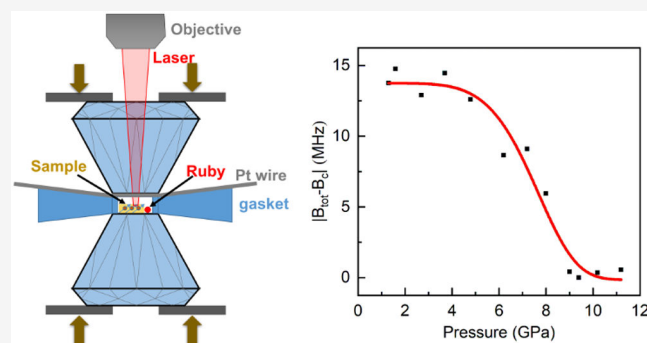
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ABSTRACT: Spin defects in silicon carbide appear to be a promising tool for various quantum technologies, especially for quantum sensing. However, this technique has been used only at ambient pressure until now. Here, by combining this technique with diamond anvil cell, we systematically study the optical and spin properties of divacancy defects created at the surface of SiC at pressures up to 40 GPa. The zero-field-splitting of the divacancy spins increases linearly with pressure with a slope of 25.1 MHz/GPa, which is almost two-times larger than that of nitrogen-vacancy centers in diamond. The corresponding pressure sensing sensitivity is about 0.28 MPa/Hz^{-1/2}. The coherent control of divacancy demonstrates that coherence time decreases as pressure increases. Based on these, the pressure-induced magnetic phase transition of Nd₂Fe₁₄B sample at high pressures was detected. These experiments pave the way to use divacancy in quantum technologies such as pressure sensing and magnetic detection at high pressures.

KEYWORDS: Divacancy, high pressure, coherent control, magnetic detection, silicon carbide



Silicon carbide (SiC) is a technology-friendly semiconductor that has been widely used for high power electronics.^{1,2} Various color centers, which can be used as bright single-photon-emitters^{3,4} or spin qubits,^{1,2,5–13} have been identified in SiC.^{1–14} The wavelengths of these single-photon-emitters range from visible to infrared,^{3,4,14} which could be used in quantum photonics.^{3,14} The spin qubits in SiC were reported to have silicon vacancy (V_{Si}),^{5–7} divacancy (V_{Si}V_C),^{1,2,8–10} nitrogen-vacancy (N_CV_{Si}) centers,^{11–13} and some others.^{15–17} The neutral divacancy is a spin defect consisting of a silicon vacancy adjacent to a carbon vacancy in SiC.^{1,2,8–10} There are seven types of divacancy defects in 4H-SiC, including PL1–PL7 divacancies,^{1,2} and the spin states of PL3 and PL5–PL7 can be controlled at room temperature.² They have two orientations: *c*-axis for PL1, PL2, and PL6, while basal for others. The divacancy defects have drawn much attention for their near-infrared photoluminescence (PL) spectrum, high optically detected magnetic resonance (ODMR) readout contrast, and long coherence time (millisecond)^{1,2,8–10} even at room temperature. Divacancy-based quantum technologies have been investigated in spin-photon interface,⁹ hybrid spin-mechanics,¹⁸ Purcell PL enhancement,¹⁹ quantum information processing,^{1,2,8,9} and quantum

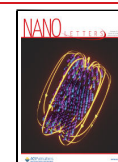
sensing including electric field,²⁰ strain,²¹ temperature,²² and magnetic field.²³

All studies of divacancy defects until now were performed at ambient pressure.^{1–14,18–23} Nothing is known about the optical and spin properties of divacancy at high pressures. Investigation of their properties can not only reveal novel physical phenomena of the divacancy but also provide new opportunities for quantum metrologies such as pressure and magnetic field sensing at high pressures.²⁴ Furthermore, SiC has mature inch-scale growth and micronano fabrication technologies. The material is much cheaper than diamond and easier to fabricate. Each type of divacancies has only one orientation. In some cases, the simplicity of the single orientation makes it easier to read out the ODMR frequencies. On the other hand, in recent years, the diamond anvil cell (DAC)-based high-pressure research got many exciting

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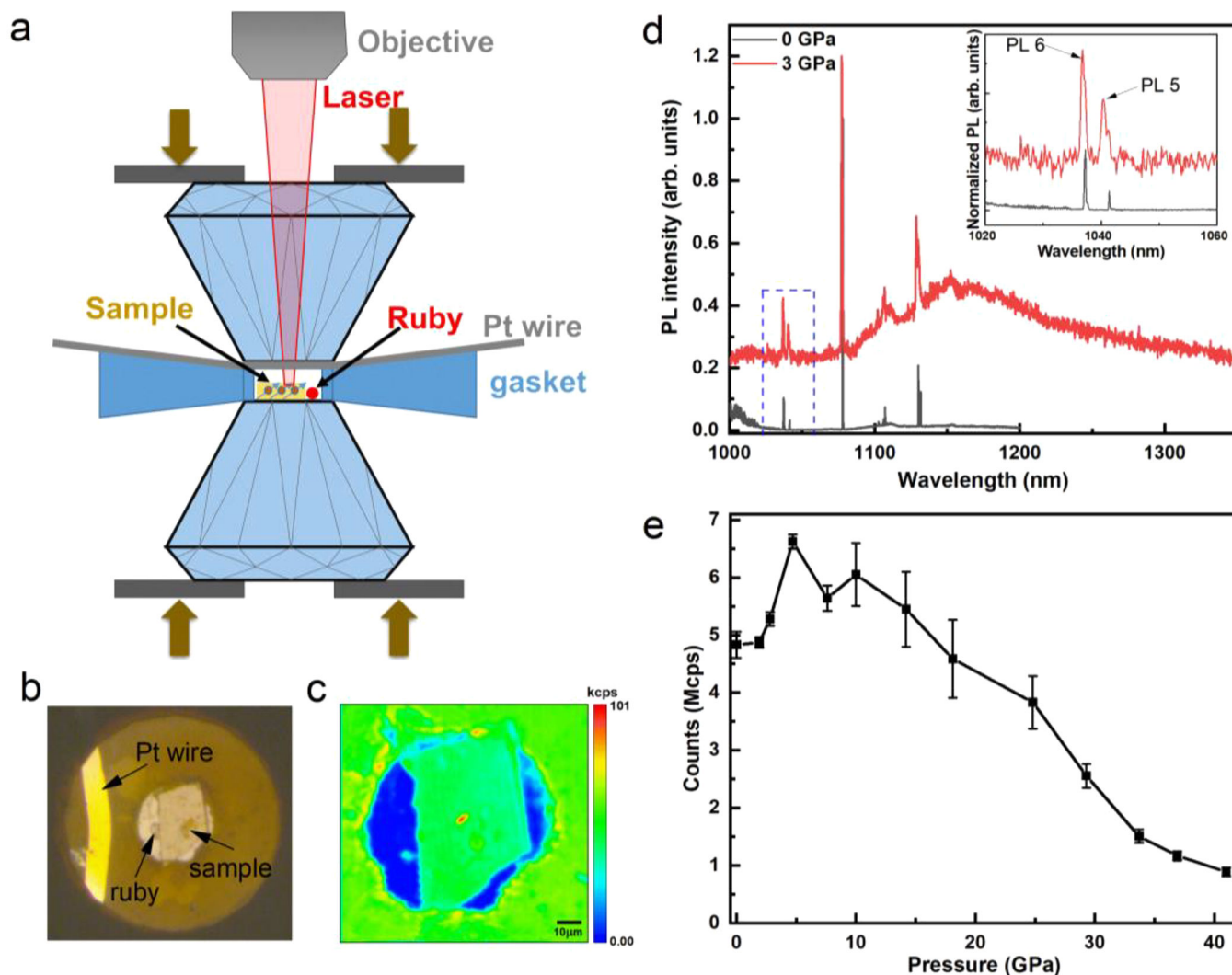


Figure 1. Schematic diagram of DAC and optical properties of the divacancy at high pressure. (a) Schematic diagram of DAC. High pressure is obtained by compressing two diamonds in opposition. A small thin piece of sample and a ruby ball are placed in the DAC chamber. A 10 μm Pt wire is crossed on the insulating gasket. A long work distance objective is used to excite the sample. (b) Optical microscopic image of the culet surface, including the sample, ruby, and Pt wire at 2.2 GPa. (c) Fluorescence scanning confocal image of the corresponding sample at 2.2 GPa. (d) Low-temperature fluorescence spectra of divacancy at 25 K. The data obtained at ambient pressure and 3 GPa are shown in black and red curves. Inset: enlarged content of the blue dotted box. The ZPL of PLS has a small blueshift. (e) Mean counts of divacancy ensemble in an $8 \times 8 \mu\text{m}^2$ area as a function of pressure at the laser power of 8 mW. The error bar is the standard deviation of the counts in the $8 \times 8 \mu\text{m}^2$ areas.

breakthroughs in multidisciplinary directions.^{24–31} There have been several claims of the superconducting temperatures approaching 300 K in the novel compounds synthesized at very high densities in DAC. For example, lanthanum hydride with T_c of 250 K was claimed at about 170 GPa.²⁷ Due to the extremely small volume of the DAC sample chamber, conventional magnetic detection technologies are difficult to implement.^{28–31} Until now, nanoscale quantum sensor nitrogen-vacancy (NV) centers in diamond have been used for *in situ* pressure sensing and magnetic environment detection in DAC.^{24,28–31} Thus, exploring the divacancy spins in SiC at high compressions could extend the scope of solid-state spin-based quantum metrology.

In this work, we realize the coherent control and magnetic detection of divacancy spin ensembles created at the surface of 4H-SiC under high pressure up to 40 GPa. By measuring the counts and zero-phonon-lines (ZPLs) of PLS as a function of pressure, we show that the ZPL blueshifts. We then detect the

ODMR signals of PLS, observing that the zero-field-splitting (ZFS) of the PLS spin increases linearly with pressure having a slope of 25.1 MHz/GPa. We also measure the Rabi oscillation and spin coherence time of the PLS spins above 36 GPa at room temperature. Finally, we measure the magnetic phase transition of $\text{Nd}_2\text{Fe}_{14}\text{B}$ under high pressure by using PL6 divacancy. These experiments demonstrate that this nanoscale divacancy quantum sensor can be applied for pressure sensing and magnetic detection at high pressures.

The experimental equipment comprises a homemade fluorescence scanning confocal microscope equipped with a microwave system, which are used to scan the sample and control the divacancy spins.^{10,12,22,23} As shown in Figure 1a, a continuous-wave 914 nm laser is used to excite the sample. Due to the large volume of the DAC device, a long work distance infrared objective (N.A. = 0.4) is applied to excite the divacancy and collect its resulting fluorescence. The low-temperature optical experiment is performed using a cryostat

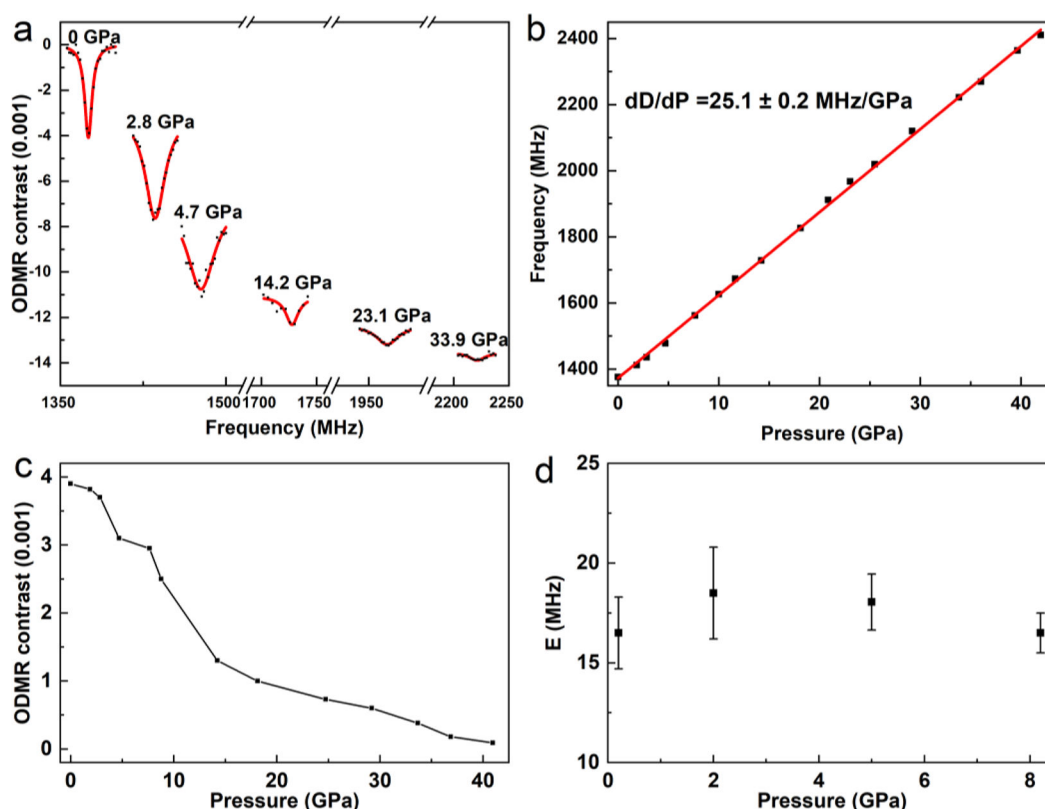


Figure 2. Pressure dependence of the ODMR spectra of PL5 divacancy at room temperature. (a) Several representative ODMR spectra (black dots) at different pressures under zero magnetic field. The red curves are the fits using the Lorentz functions. (b) Measured ZFS parameter increases linearly as the pressure increases up to 42 GPa with a slope of 25.1 MHz/GPa. (c) ODMR contrast as a function of the pressure. (d) E value of PL5 remains the same at different pressures. The error bar is the standard deviation. The pressure error bars are smaller than the size of data points.

combined with a fluorescence scanning confocal microscope system. A superconducting single-photon detector (SSPD) is used for the detection of divacancy fluorescence.¹² Standard lock-in technology is used for spin property measurements with a photoreceiver (Femto, OE-200-IN1).^{1,2,12,23} In order to generate high-density divacancy, 30 keV nitrogen ions with a dose of $1 \times 10^{14}/\text{cm}^2$ are implanted ~ 50 nm from the surface of a bulk high-purity 4H-SiC epitaxy layer sample. Then the sample is annealed at 1050 °C for 2 h to efficiently generate divacancy. Finally, the sample is polished mechanically to a thickness of 40 μm to place it in the DAC chamber.

In our experiment, we used a nonmagnetic beryllium copper DAC to generate high pressure. The DAC is mounted on a three-axis nanoscale piezoelectric stage (PI, P-561.3CD) to realize three-dimensional fluorescence confocal scanning of the divacancy sample. As shown in Figure 1a, the high-pressure device (DAC) is mainly composed of two aligned diamonds and a gasket with a sample chamber on it. Two diamonds with 200 μm diameter culet and an insulating rhenium gasket are used in this experiment. NaCl is applied as the pressure transmitting medium, which provides quasi-hydrostatic pressure environment. Figure 1b shows the optical microscopic image of a tiny piece of the 4H-SiC sample with the implanted divacancy defects in the pressure chamber at 2.2 GPa. A 10 μm ruby ball is set close to the sample, which is used to monitor the local pressure.³² A 10 μm Pt wire across the culet was used to transmit the microwave. The corresponding fluorescence scanning confocal image with a transverse diffraction-limited

spot size of ~ 3 μm of the sample is presented in Figure 1c, which is consistent with the microscopic image.

The divacancy defects have a ground electron spin $S = 1$, and the Hamiltonian can be written as $H = DS_z^2 + E(S_x^2 - S_y^2) + g\mu_B \mathbf{B} \cdot \mathbf{S}$, where D and E are the axially symmetric and anisotropic components of ZFS, g is Lande factor, μ_B is Bohr magneton, $\mathbf{B}$ is an external magnetic field, and $\mathbf{S}$ is electron spin.^{1,2,8–10} Since the spin state is well isolated from other ODMR peaks, we select the right-branch ODMR peak of the PL5 divacancy in the experiment. The PL5 divacancy is a basal defect, which has a C_{1h} symmetry, and its two ODMR peak positions are 1343.5 and 1375 MHz at room temperature.^{1,2}

At room temperature, the PL spectra of different types of divacancy defects are mixed together due to the phonon sideband. To distinguish PL5 and PL6, we measure the divacancy PL spectrum at about 25 K (Figure 1d). The experiments were performed in a cryostat combined with a fluorescence scanning confocal microscope system, and the PL spectra are measured with a spectrometer (Horiba, iHR550). The inset shows the ZPLs of PL5 and PL6 at ambient pressure and 3 GPa. At ambient pressure, the ZPLs of PL5 and PL6 are about 1041.4 and 1037.2 nm, which are consistent with the previous results.^{1,2,33} At 3 GPa, the ZPLs of PL5 and PL6 are about 1040.4 and 1036.8 nm, respectively. The results demonstrate that the ZPLs blueshift with pressure, in agreement with the NV centers in diamond.²¹ The mean counts of the divacancy as a function of pressure are summarized in Figure 1e. The counts first increase as the pressure increases from ambient pressure to ~ 8 GPa, which

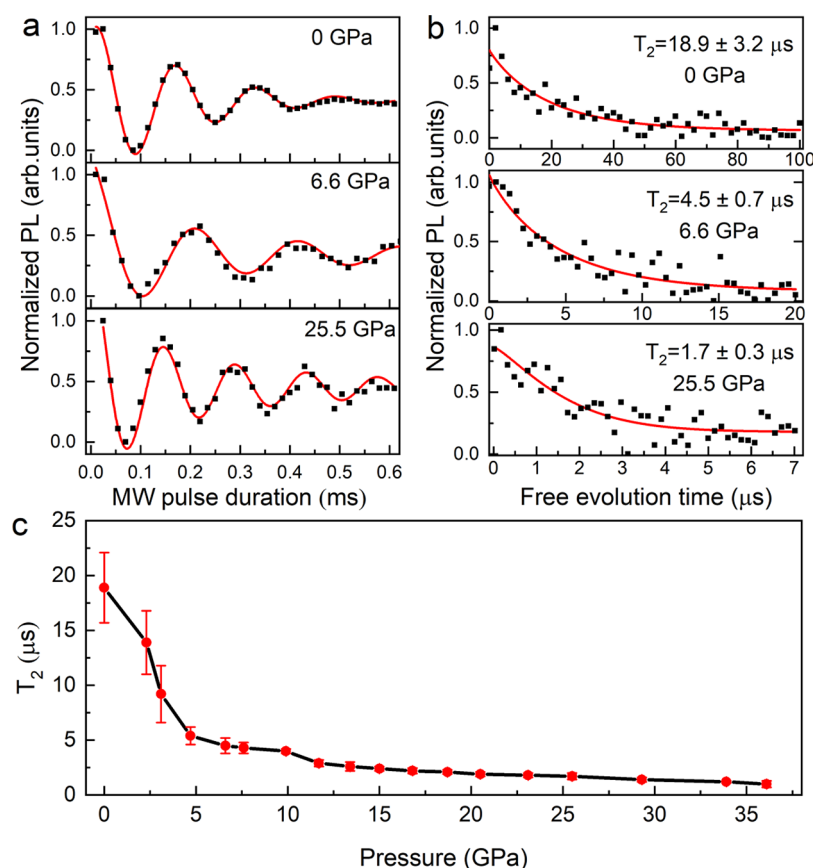


Figure 3. Coherent control of the PL5 divacancy spin at high pressures and room temperature. (a) Three representative Rabi oscillation measurements (black square) of PL5 at different pressures. The exponential decaying cosine function is used to fit the data (red lines). (b) Three typical spin echo measurements at different pressures. Decaying exponential functions are used to fit the data (red lines). (c) Measured spin coherence time T_2 as a function of pressure up to 36 GPa.

may be due to the higher detection efficiency of the SSPD at a lower wavelength. Then it decreases as the pressure increases up to ~ 42 GPa, which is due to the lattice distortion caused by compression.

The spin properties are the foundation for divacancy-based quantum sensing at high pressure. Given this, we then study the pressure-dependent ODMR spectrum at room temperature of the PL5 divacancy at zero magnetic field. Several representative ODMR spectra of the right branch of PL5 under different pressures are presented in Figure 2a. At ambient pressure, the resonance frequency is about 1376 MHz, which is consistent with previous studies,^{1,2,23} while the resonance frequencies at 14.2 and 33.9 GPa are 1727.7 and 2222.1 MHz. We find that the ODMR peaks gradually broaden at higher pressures, which is caused by the slightly crystal distortion at high pressures.^{24,34} These frequency results show that the ZFS parameter shifts to higher frequencies with compression. The ZFS parameter as a function of pressure up to ~ 42 GPa is shown in Figure 2b. The results demonstrate that the ZFS parameter D linearly increases as pressure is applied with a slope of about $dD/dP = 25.1 \pm 0.2$ MHz/GPa, which is around two-times that of the nitrogen-vacancy (NV) centers in diamond.^{24,28} We also measured the slope of PL6 (shown in Figure 4a) and PL7 (shown in Figure S2). The increase in D with pressure may be due to several reasons: the change in the localization of the electron density on the adjacent atoms of the divacancy and the shape of the localization at the divacancy sites; or structural relaxation at

the divacancy site while the compression of the SiC lattice induces a distance decrease in the spins in the SiC lattice. Additionally, the anisotropic component of ZFS E at high pressure is shown in Figure 2d. The results show that E does not change until 8.2 GPa, suggesting that the local symmetry around the defect sites does not significantly change under pressure.³⁵ The phenomenon is consistent with the results of NV centers in diamond.³⁵ The slope of the PL5 divacancy is larger than that of NV centers in diamond, which may be due to the lower hardness of the SiC lattice and the divacancy defect properties. We note that the larger slope of the PL5 divacancy is beneficial for pressure sensing. The estimated ODMR-based pressure sensing sensitivity of the divacancy ensemble at room temperature is²⁴

$$\eta(P) = \frac{1}{2\pi \frac{dD(P)}{dP} K \sqrt{T_2^*}} \quad (1)$$

where $K = (1 + 2(a_0 + a_1)/(a_0 - a_1)^2)^{-1/2}$, $a_0 = I \times \tau$ and $a_1 = I \times \tau \times (1 - c)$, $T_2^* \approx 0.1 \mu$ s³⁶ (shown in Figure S3). Here, I is the photon count (133 Mcps shown in Figure S4), τ is the Rabi readout duration time (350 ns), and c is the ODMR contrast (0.02). Using the data, the calculated pressure sensing sensitivity is $0.28 \text{ MPa}/\sqrt{\text{Hz}}$.

In addition to the resonant frequency, signal contrast is also an important parameter for the ODMR spectra. To compare the contrast at different pressures, we maintain the laser power (8 mW) and microwave power (Rabi frequency 2.2 MHz)

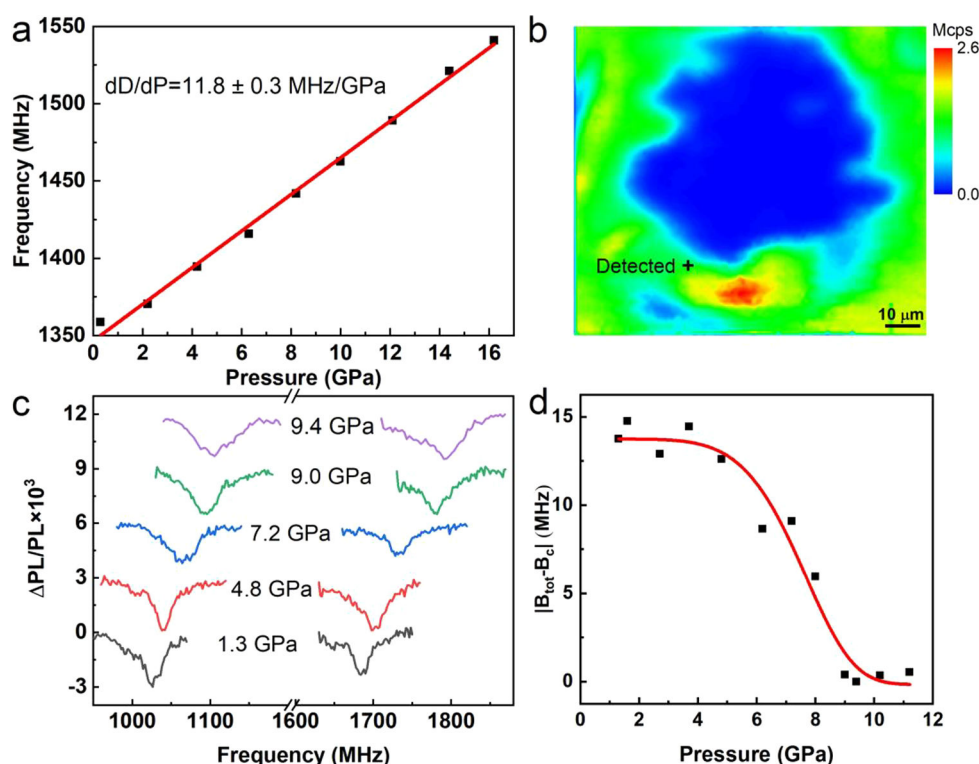


Figure 4. Detection of pressure-induced magnetic phase transition of a $\text{Nd}_2\text{Fe}_{14}\text{B}$ magnet using PL6 defects. (a) Measured ZFS parameter D increases linearly as the pressure increases up to 16.2 GPa with a slope of 11.8 MHz/GPa. (b) PL image of the PL6 defects and magnetic $\text{Nd}_2\text{Fe}_{14}\text{B}$ sample on the culet surface of a diamond. The blue part in the middle is the $\text{Nd}_2\text{Fe}_{14}\text{B}$ sample, and the rest is the silicon carbide micrometer slice. The scale bar is 10 μm . (c) ODMR spectra of PL6 defects during the compression process. (d) Inferred magnetic fields of the $\text{Nd}_2\text{Fe}_{14}\text{B}$ sample are detected by PL6 defects during the compression processes.

stable in the experiments. Figure 2c presents the ODMR contrast of PL5 with respect to pressure. The ODMR contrast decreases quickly as the pressure increases from ambient pressure to about 15 GPa and then decreases at a slower speed up to about 42 GPa, which is similar to the ODMR contrast of the NV centers in diamond.²¹ This may be due to the pressure changes the optically induced spin polarization process. This experiment demonstrates that the PL5 ODMR spectra can be used for *in situ* pressure probes in the DAC chamber.^{28–31}

Coherent control of divacancy spins at high pressures not only provides insight into the spin properties but also is important to detect phase transitions of the sample with extremely weak magnetic signals.^{30,31} We then study the coherent control of the PL5 spins at zero magnetic field, room temperature, and different pressures. The standard pulse sequence of the Rabi oscillation is used in experiments.^{1,2,12} When a resonant microwave pulse is applied to drive the divacancy spins, population oscillations occur between the resonant spin states. Three typical Rabi oscillations under different pressures are presented in Figure 3a. We can see that it still has a good coherent control under a high pressure of 25.5 GPa and the corresponding Rabi frequency is about 7.0 MHz. Spin coherence time (T_2) is a key parameter in multiple quantum technologies.^{1,2} Given this, we further measure the spin echo of the PL5 with pressures. As shown in Figure 3b, the coherence time T_2 decreases with pressure. Specifically, the T_2 at ambient pressure and 25.5 GPa are $18.9 \pm 3.2 \mu\text{s}$ and $1.7 \pm 0.7 \mu\text{s}$. Finally, the measured spin coherence time T_2 as a function of the pressure up to 36 GPa is summarized in Figure 3c. The results show that T_2 decreases quickly from 18.9 μs to about 5.4 μs as the pressure increases from ambient pressure to

about 5 GPa, then it slowly declines to about 0.9 μs as the pressure increases up to 36.1 GPa, which is different with that of NV centers in diamond.³¹ The decrease in T_2 may be due to the distance decreases of the divacancy and spin bath defects, which will increase the interactions between the divacancy and the spin bath. In addition, we measured T_2^* at 0.3 GPa, obtaining the value of $108 \pm 6 \text{ ns}$ (shown in Figure S3).

In situ magnetic detection at high pressure is a long-term and difficult task.^{28–31} The magnetism of numerous materials will obviously change under compression, for example,^{31,37} $\text{Nd}_2\text{Fe}_{14}\text{B}$ will change from ferromagnetic phase to paramagnetic phase under compression.^{31,38–40} We detect the target materials' phase transition by measuring the ODMR spectra under external magnetic field. Since the direction of the PL5 is basal, it is hard to add on-axis magnetic field. In view of this, we chose the *c*-axis PL6.^{1,2} External pressure and magnetic field acting on PL6 defects will lead to different changes in its ODMR spectrum. Figure 4a shows the trend of the ZFS parameter D of PL6 defects as pressure increases. D increases linearly with pressure, and the slope is $11.8 \pm 0.3 \text{ MHz/GPa}$. As shown in Figure 4b, we choose a measuring spot near the $\text{Nd}_2\text{Fe}_{14}\text{B}$ material to detect changes in its magnetism with pressure. We applied an external *c*-axis magnetic field of about 127 G (B_c). The probe process is illustrated in Figure 4c. The magnetism of $\text{Nd}_2\text{Fe}_{14}\text{B}$ material will decrease with pressure. Therefore, under different pressures, PL6 defects detect different magnetic fields, so we can use the ODMR spectrum to infer the magnetic changes of $\text{Nd}_2\text{Fe}_{14}\text{B}$ materials. The results are shown in Figure 4d. The B_{tot} is the total magnetic field on the PL6 defects, thus $|B_{\text{tot}} - B_c|$ stands for the magnetic field of $\text{Nd}_2\text{Fe}_{14}\text{B}$. As the experimental results show, magnetic

field of $\text{Nd}_2\text{Fe}_{14}\text{B}$ decreases significantly as the pressure increases from 5 GPa to about 9 GPa, which is in good agreement with the previous results.³¹ The experiments demonstrate that divacancy can be used as a quantum magnetometer for magnetic detection at high pressure.

In summary, we explore the optical properties and realize coherent control of the PL5 divacancy defects in 4H-SiC up to 40 GPa using a DAC device. The experiments show that the ZPL of the divacancy has a blueshift and the counts decrease under high pressure. The ZFS parameters increase linearly with the pressure with a slope of 25.1 MHz/GPa, which is almost two-times larger than that of the NV centers in diamond. The larger slope is beneficial to improve the sensitivity of SiC-based pressure sensing. The divacancy ensemble-based pressure sensing sensitivity is 0.28 MPa/ $\sqrt{\text{Hz}}$. The coherence time T_2 of the PL5 divacancy at high pressures decreases as the pressure increases. Finally, we successfully detected the magnetic phase transition of $\text{Nd}_2\text{Fe}_{14}\text{B}$ under high pressures using the PL6 defects. The experiments show that the nanoscale divacancy quantum sensor in SiC can be used in pressure sensing and *in situ* magnetic detection at high pressure.

■ ASSOCIATED CONTENT

SI Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acs.nanolett.2c03378>.

ODMR spectra of PL5 and PL7 with pressure, T_2^* of PL5, fluorescence saturation curve (PDF)

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Author Contributions

△L.L. and J.F.W. contributed equally to the work.

Notes

The authors declare no competing financial interest.

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