

Probing the nitrogen vacancy center using the Faraday effect at room temperature

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The spin states of the nitrogen vacancy (NV^-) center in diamond are used in, among others, quantum computing, nanoscale imaging, and magnetic field sensing, and have been proposed for fundamental physics experiments. The states of NV^- centers are traditionally measured using the spin-state-dependent photoluminescence—a destructive and often inefficient measurement scheme. In this article, using a single laser beam as a pump and a probe, we measure the spin states of the NV^- centers nondestructively using the Faraday effect at room temperature. Using the optical selection rules and the helicity-dependent absorption, we identify the optical transitions of the NV^- center responsible for the Faraday effect and confirm the existence of spin-orbit coupling in such centers at room temperature. Our results indicate that experiments such as NV spin-photon entanglement and electromagnetically induced transparency are viable in ambient conditions. Finally, using the Faraday effect, we develop a magnetic field sensor and argue that, with the inclusion of an optical cavity, a femtotesla-level magnetic field sensitivity can be achieved.

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I. INTRODUCTION

The nitrogen vacancy (NV^-) center in diamond is an atomlike system [1] that is used for quantum computing and communications [2,3], nanoscale imaging [4,5], magnetic field sensing [6–9], and thermometry [10]. Such centers have also been proposed in experiments for probing quantum gravity [11] and testing the macroscopic limit of quantum mechanics [12,13]. A key attribute of an NV^- center is its long spin coherence time (over a millisecond) [14], which enables it to interrogate its environment for a prolonged period, thereby improving measurement precision and fidelity. The spin state of an NV^- center is conventionally measured using its spin-state-dependent photoluminescence (PL), which is emitted in all directions. Due to the omnidirectional emission, not all photons emitted by an NV^- center can be collected, implying information loss. This limits the sensitivity of NV^- center-based magnetic field sensors [6] and the readout fidelity in experiments such as spin-photon entanglement [15] and the proposed test of quantum gravity [11]. The limited sensitivities of NV^- magnetometers also preclude them from applications such as magnetoencephalography (MEG), despite their high spatial resolutions and the ability to work in ambient conditions. Moreover, the existing PL-based measurement technique is destructive in the sense that the same state cannot be measured more than once to enhance the fidelity [16]. Techniques that can measure the NV^- spin states in a single shot at room temperature are

ideal, but any measurement scheme that has the potential to overcome the limitations of the photoluminescence-based measurements would benefit applications such as magnetic field sensing, biomedical imaging, and magnetic navigation.

In this article, we measure the spin states of NV^- centers using the Faraday effect (FE) at room temperature using a single laser as a pump and a probe. Our measurement scheme is inherently nondestructive [16,17], can be repeated many times with the inclusion of an optical cavity [18,19], and confirms the existence of spin-orbit coupling in NV^- centers at room temperature [20]. We investigate the spin-light interaction using the polarization degree of freedom of the probe laser and the optical dipole selection rules, and identify transitions responsible for FE. To show the utility of our NV^- center readout technique, we develop a magnetic field sensor.

II. EXPERIMENTAL DETAILS

In our experiment, we use a $2 \times 2 \times 2 \text{ mm}^3$ high pressure high temperature (HPHT) diamond with an NV^- concentration of 0.22 ppm and a nitrogen concentration of 50 ppm. Figure 1(a) shows an NV^- center oriented along the [111] crystallographic axis and its energy-level diagram. Existing room-temperature experiments utilize three excited energy levels to explain data [1]. However, to interpret our results, the six-level excited-state model, typical of low-temperature experiments [15,21], is more appropriate [1]. As we use an ensemble of NV^- centers, some strain is inevitable [1,21–24], which splits the excited state into two branches: $^3E_x (A_1, A_2, E_x)$ and $^3E_y (E_1, E_2, E_y)$. At relatively low strain, the $A_{1,2}$ and $E_{1,2}$ states are spin-orbital entangled states, i.e., $A_{1,2} = |E_- \rangle \otimes | + 1 \rangle \mp |E_+ \rangle \otimes | - 1 \rangle$ and $E_{1,2} = |E_- \rangle \otimes | - 1 \rangle \mp |E_+ \rangle \otimes | + 1 \rangle$ [15,21,25], where $E_{\pm}$ is the excited-state orbital angular momentum and $| \pm 1 \rangle$ represent the spin states for $m_s = \pm 1$. A schematic of our experimental setup is shown in Fig. 1(b). The diamond in our experiment is

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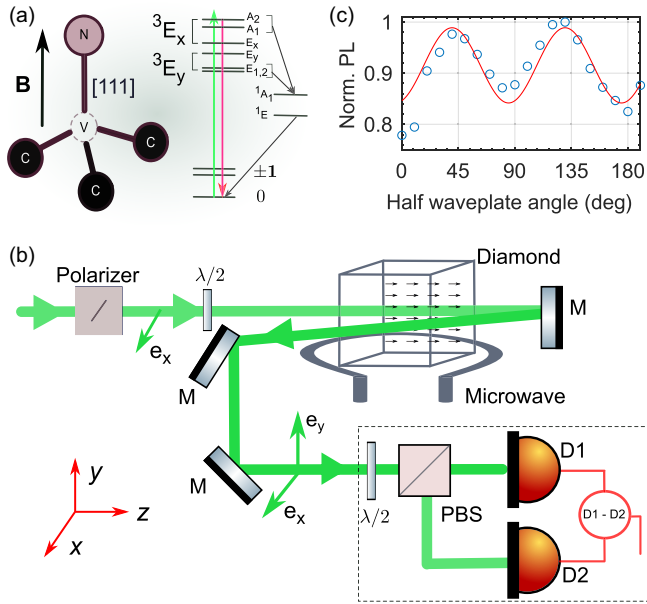


FIG. 1. (a) The nitrogen vacancy center in diamond and its energy-level diagram. Here, an NV^- center oriented along the $[111]$ direction has been shown. A dc magnetic field B is applied along the $[111]$ crystallographic direction for lifting the degeneracy of the NV^- center's ground-state manifold. (b) A schematic of our experimental setup. A green laser (532 nm) is used for exciting the NV^- centers from their ground-state manifold ($^3A_2$) to the excited-state manifold (^3E). Before interacting with the NV^- centers, the laser passes through a linear polarizer and a half-waveplate ($\lambda/2$). Components within the dotted box constitute a balanced polarimeter. Different components are as follows: M, mirror; $D_{1,2}$, photodiode; and PBS, polarizing beam splitter. $e_{x,y}$ represents the polarization of the laser along the x , y axes. (c) Photoluminescence as a function of the angle α of the half-waveplate located before the diamond and measured from the x axis.

oriented such that the $[111]$ crystallographic direction is along the z axis. A green laser (532 nm) propagating along the z axis excites the nitrogen vacancy center nonresonantly from the ground state to the excited state and spin polarizes the NV^- centers to the $m_s = 0$ state [1]. As well as using the green laser as an excitation source, we use it as a probe to measure the spin states of NV^- centers via the Faraday effect. To excite more NV^- centers and enhance FE, the green laser is retroreflected and picked up using a pickoff mirror. The double-passed laser is then inputted to a balanced polarimeter [17,26]. A fraction of the retroreflected green laser is used for absorption-detected magnetic resonance (ADMR) [27]. Photoluminescence emitted by NV^- centers is detected using a detector placed along the x axis. Figure 1(c) shows PL as we change α , the angle between the x axis and the laser polarization, with no microwaves in use. By fitting dipole radiation pattern with the PL [22,28,29], we identify the orientations of the NV^- dipoles, which play critical roles in FE. From fit, we find that two dipoles belonging to NVs parallel to $[111]$ are located at 72° (^3E_y) and -18° (^3E_x) from the x axis. Note that PL is independent of the orientations of the dipoles belonging to the NV centers not aligned to the $[111]$ direction (see Appendix).

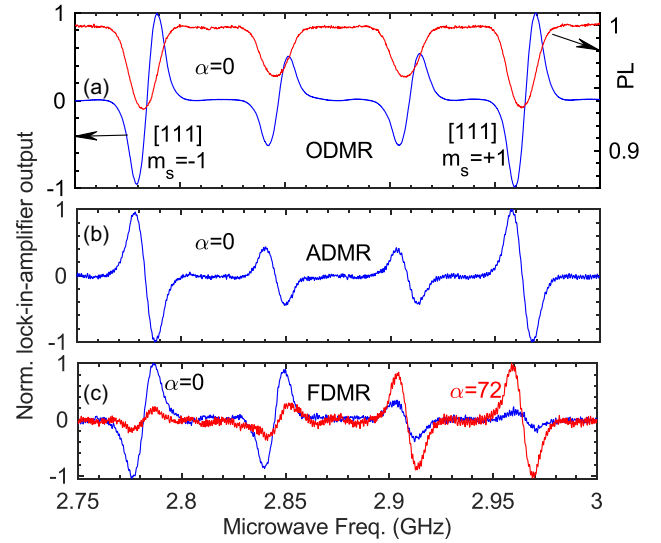


FIG. 2. Magnetic resonances detected using a lock-in amplifier. (a) PL-based optically detected magnetic resonance when the angle α between the axis of the polarizer (x axis) and the half-waveplate after the polarizer is zero. Resonances at 2.785 and 2.965 GHz are due to the NV centers aligned to the $[111]$ direction, while resonances at other frequencies belong to NV centers not parallel to the $[111]$ direction. Raw PL as a function of microwave frequency is also shown (right, red). (b) The green laser absorption-detected magnetic resonance for $\alpha = 0$. (c) The Faraday detected magnetic resonance for $\alpha = 0$ and 72° .

III. THE NV^- CENTER AND THE POLARIZATION OF LIGHT

Figure 2(a) shows the raw photoluminescence (red) and the same when detected using a lock-in amplifier (LIA, blue) with microwaves on and $\alpha = 0$. Due to $B \parallel [111]$, the optically detected magnetic resonances (ODMRs) belonging to the NV^- centers aligned to the $[111]$ direction appear at $(D + \gamma m_s B)/2\pi$, while the ODMRs of the remaining NV^- centers cluster at $(D + \gamma m_s B \cos \theta)/2\pi$ with $\theta \approx 109^\circ$. The ODMRs at $(D + m_s \gamma B)/2\pi$ are stronger than others due to the orthogonality of the microwave magnetic field with these centers, allowing efficient excitation. Figure 2(b) shows ADMR [27]. The polarities of all ADMR peaks are identical but are opposite to those due to PL.

Figure 2(c) shows the output of LIA when we probe the spin states of the NV^- centers using the retroreflected green laser and the balanced polarimeter. Four peaks are visible, which we call the Faraday detected magnetic resonance (FDMR). The two peaks related to the $m_s = 0 \rightarrow +1$ transitions at higher microwave frequencies are stronger than those due to the $m_s = 0 \rightarrow -1$ transitions for $\alpha = 72^\circ$ (red). This is expected as we are only exciting the ^3E_y dipole [21]. The situation changes when the polarization of the laser is parallel to the ^3E_x dipole ($\alpha = 0$, blue). Now the peaks belonging to the $m_s = 0 \rightarrow -1$ transitions are dominant. Most importantly, in both cases, the polarities of the peaks originating from the $m_s = 0 \rightarrow -1$ transitions and the $m_s = 0 \rightarrow +1$ transitions are opposite. This contrasts with ADMR and ODMR, which have peaks of the same polarities.

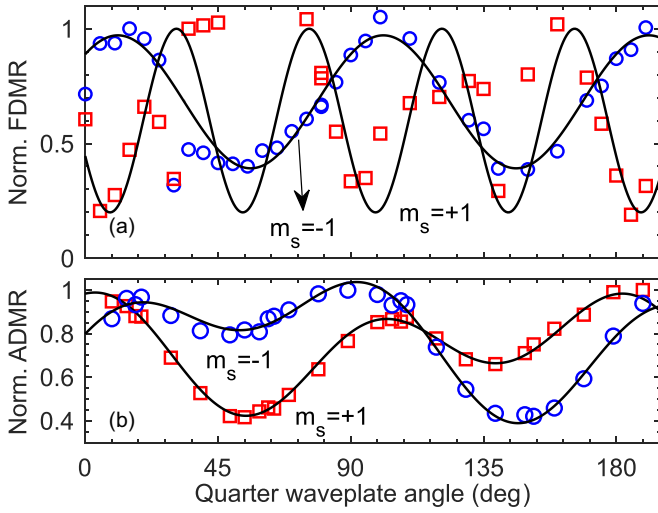


FIG. 3. Peak-to-peak amplitudes of the lock-in-amplifier outputs of (a) FDMR and (b) ADMR as a function of the QWP angle α when the $m_s = 0 \rightarrow -1$ ($+1$) transitions, blue circles (red squares), are pumped for the NV^- centers parallel to the $[111]$ direction. For these experiments, the half-waveplate before the diamond [see Fig. 1(b)] was replaced by a QWP. Light is σ_- (σ_+) polarized at $\alpha = 57^\circ$ (147° , an offset of 12° due to different optical components), where α is the angle between the x axis and the slow axis of the QWP. Solid lines are guides to the eyes only.

Linearly polarized light is composed of left ($\hat{\sigma}_+$) and right ($\hat{\sigma}_-$) circularly polarized light, i.e., $\hat{x} = (\hat{\sigma}_+ + \hat{\sigma}_-)/\sqrt{2}$ and $\hat{y} = -i(\hat{\sigma}_+ - \hat{\sigma}_-)/\sqrt{2}$. NV^- centers in the $m_s = -1$ state add a phase to the $\hat{\sigma}_+$ component of the linearly polarized light, keeping the $\hat{\sigma}_-$ component intact [16,30,31]. The spin-state-dependent phase rotates the plane of polarization of linearly polarized light in the opposite direction, giving rise to FDMR peaks of opposite polarities. For a fixed microwave power, the strength of FDMRs increases linearly without saturation as the green laser power goes up (see Appendix). In contrast, FDMRs saturate as the microwave power increases for a fixed green laser power (see Appendix). We note that in our Faraday measurements, the ^3E_y dipole appears at $\alpha = 0$ instead of $\alpha = -18^\circ$ predicted by the fluorescence measurement [Fig. 1(c)]. This might be due to the misalignment of our probe with respect to the facet of the diamond.

The Faraday effect depends on the helicity of the laser beam [26]. Ideally, when the probe light is circularly polarized, no FE is expected. Figure 3(a) shows the peak-to-peak amplitudes of FDMRs as we change the helicity of the green laser using a quarter-waveplate (QWP) immediately after the polarizer [Fig. 1(b)]. The reflection from the facets of the diamond varies by $<5\%$ as we change α . As light becomes σ_- (σ_+) polarized at $\alpha = 57^\circ$ ($\alpha = 147^\circ$, an offset of 12° due to different optical components (see Appendix)), the strengths of the FDMRs belonging to $m_s = \pm 1$ are minimized. We measure a contrast of 60%(80)% between linearly and circularly polarized light for $m_s = -1$ ($+1$), respectively. Considering that we are using an ensemble of NV^- centers and a double-passed laser beam that we purposely misalign to facilitate pick off (light appears elliptically polarized to the NV^- centers), this level of contrast is excellent. Moreover, this

helicity-dependent oscillation of our signals confirms that their origin lies in the Faraday effect, which has not been detected in NV center prior to our experiment at room temperature. Our experiment also confirms the existence of the spin-orbit coupling, the underlying phenomenon behind FE, despite the prediction of thermal averaging-induced suppression of such coupling at ambient conditions [20].

We now identify transitions [21], e.g., $m_s = \pm 1 \rightarrow A_{1,2}$ and $E_{1,2}$, responsible for the Faraday effect. Among these transitions, $m_s = \pm 1 \rightarrow A_{1,2}$ happen when the spin and the angular momentum of light have the opposite polarities, i.e., $m_s = +1$ and σ_- . The opposite is true when the $m_s = \pm 1 \rightarrow E_{1,2}$ transitions are pumped [21,25]. In Fig. 3(b), we show the peak-to-peak amplitudes of ADMRs detected using an LIA. As light becomes σ_- polarized and the $m_s = 0 \rightarrow +1$ transition is pumped, the strength of the ADMRs and hence transmission through the diamond is reduced by 55% compared to when light is linearly polarized. The situation is reversed when light is σ_+ polarized and the transmission through the diamond is now suppressed by 55% when the $m_s = 0 \rightarrow -1$ transition is pumped. In both cases, photoluminescence increases, implying more absorption. A relatively small drop in transmission ($\approx 15\%$) when light and spins have the same polarity is attributed to the misalignment of the probe beam with respect to the NV^- centers, which makes light appear to be elliptically polarized. This undesired ellipticity has the effect of reducing (increasing) the strength of the $m_s = \pm 1 \rightarrow \sigma_\mp$ ($\sigma_\pm$) transitions. Nevertheless, the relatively strong coupling between light and spins of opposite helicity indicates that in our Faraday experiment, the $A_{1,2}$ states play the pivotal roles. To differentiate between A_1 and A_2 , however, requires resonant light. Importantly, to our knowledge, this is the first time $A_{1,2}$ levels of the NV center have been identified at room temperature. Moreover, the identification of the A_2 level at room temperature, which forms a Λ system in combination with $m_s = \pm 1$ [Fig. 1(b)], might open the door for spin-photon entanglement for quantum communications [15] and demonstrate electromagnetically induced transparency for magnetometry at room temperature [32].

The rotation angle ϕ of the plane of polarization of the green light after it has traveled through the spin polarized ($m_s = \pm 1$) diamond can be found from the balanced detector signal [26] $\phi \approx \frac{\Delta P}{2P}$, where P is the input power to the polarimeter, ΔP is the difference in the input power to each photodiode [Fig. 1(b)] on and off microwave resonance, and $|\phi| \ll \pi/4$ is assumed. Due to the smallness of ϕ , ΔP is small and often perturbed by vibrations and laser noise, and hence, we use the balanced polarimeter output in combination with an LIA. The output of LIA is appropriately calibrated to find ϕ (see Appendix). Figure 4(a) shows the LIA output, while Fig. 4(b) presents the corresponding ϕ . On microwave resonance, we have $\phi = -6 \mu\text{rad}$ or equivalently $\Delta P = 5 \text{ nW}$ for $P = 4 \text{ mW}$. When the $m_s = 0 \rightarrow +1$ transition is pumped, the plane of polarization of the green laser is rotated by the same amount but in the opposite direction.

ϕ can be calculated phenomenologically [17,26]. When the $m_s = 0 \rightarrow -1$ ($+1$) transition is pumped using microwaves, NV^- centers become spin polarized, i.e., $\langle S_z \rangle < 0$ (> 0), and the left and right circularly polarized light encounters different indices of refraction. This difference in index of refraction

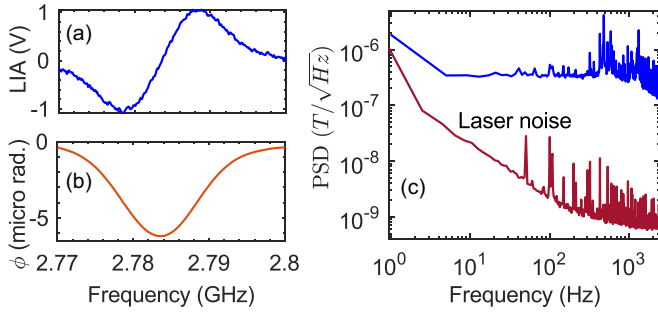


FIG. 4. The strongest FDMR [panel (a)] and the corresponding rotation angle [panel (b)] when the $m_s = 0 \rightarrow -1$ transition belonging to the NV centers parallel to the [111] crystallographic direction is pumped. In this case, a half-waveplate after the polarizer [see Fig. 1(b)] is used with $\alpha = 0$. ϕ is found from panel (a) after an appropriate calibration (see Appendix). The corresponding magnetic field sensitivity (blue line) of the FDMR-based sensor is shown in panel (c) with the microwave set on the $m_s = 0 \rightarrow -1$ resonance. For comparison, laser noise is also shown (red line). We average ten FFTs obtained from 0.5 s long time traces for finding the magnetic field sensitivity.

rotates the plane of polarization of light by

$$\phi = 2n_t k L \rho_n \text{Re}(\zeta) \langle S_z \rangle \hat{z} \cdot \hat{k}, \quad (1)$$

where n_t is the number of times probe light of wavelength λ travels through the diamond of length L , $k = 2\pi/\lambda$, ρ_n is the number density of NV[−] centers, and $\zeta = \frac{r_e f_s c^2 L(\nu)}{4\pi \nu}$ with c being the speed of light and r_e being the electron radius. f_s and $L(\nu) = 1/(\nu - \nu_0 + i\Gamma)$ are the oscillator strength and the absorption spectral profile of an NV[−] center with $\nu = c/\lambda$ being the laser frequency, ν_0 is the optical resonance frequency of the NV[−] center, and Γ is the linewidth of the optical resonance. $\langle S_z \rangle = \sum_s m_s P_s$ is the average spin polarization of NV[−] centers with P_s being the probability of an NV[−] center being in the s state. Using the analytical and experimental ϕ , we now estimate the number density of the NV[−] centers. For our calculation, we consider the NV[−] center to have a Lorentzian absorption profile with $\Gamma \approx 10^{14}$ Hz (≈ 98 nm) and $\nu_0 = 5.4 \times 10^{14}$ Hz (555 nm) [33,34], $f_s = 0.02$ [35]. From the rate-equations-based simulation [23], for our microwave and laser power, we find ≈ 43 (13)% of the NV[−] population is in the $m_s = -1(+1)$ state implying $\langle S_z \rangle = -0.30$ (see Appendix). With $|\phi| = 6 \times 10^{-6}$ rad [Fig. 4(b)] and taking into account that we are only exciting $\approx 1\%$ of the NV[−] centers due to the limited amount of green laser power available (see Appendix), we get $\rho_n = 1.6 \times 10^{22} \text{ m}^{-3}$. From our EPR measurements, we have $\rho_n = 4.01 \pm 0.36 \times 10^{22} \text{ m}^{-3}$. Considering the uncertainty in different parameters, including the NV[−] absorption profile and oscillator strengths, ρ_n found from ϕ is reasonable. Using this ρ_n and taking the laser illuminated volume of the diamond as that of a cylinder of diameter 1 mm and length 2 mm, we calculate the number of NV[−] centers interacted with the green probe beam is $n_s = 3.1 \times 10^{10}$. In other words, a single photon has interacted with 2.5×10^4 centers.

IV. THE FARADAY EFFECT AND A VECTOR NV[−] MAGNETOMETER

We now use our ability to read the spin states of the NV[−] centers using FE to develop a magnetic field sensor. To perform magnetometry, we use the $m_s = 0 \rightarrow -1$ transition and continuously drive it using resonant microwaves. Magnetic fields to be detected shift the resonance frequency via the Zeeman effect. This changes the NV[−] center population in the $m_s = -1$ state. This, in turn, changes ϕ , which then perturbs the balance of the balanced detector. Figure 4(c) shows the Fourier transforms of the calibrated outputs of the lock-in amplifier. Between 5 and 400 Hz, our magnetometer has a sensitivity of 350 nT/ $\sqrt{\text{Hz}}$. The spin-projection-limited minimum detectable magnetic field [26,36,37] is

$$\Delta B = \frac{1}{\gamma \sqrt{n_t n_s T_2^* t}}, \quad (2)$$

where n_s is the number of the NV[−] center that contributed to the Faraday effect, T_2^* is the intrinsic spin coherence time, and t is the measurement time. The intrinsic spin coherence time of our diamond is $T_2^* = 21$ ns. Substituting n_s , T_2^* , and $t = 0.5$ s in Eq. (2), we get $\Delta B = 1.7 \times 10^{-12}$ T. This is significantly better than what we have measured. When we set the frequency of our microwaves away from the resonance, the noise floor remains the same, indicating that we are limited by nonmagnetic noise. When we separately measure the laser noise, the noise floor is significantly lower. The temperature of our diamond increases by 30 K from room temperature at the microwave and laser power used—measured using the shift in the ODMR frequency [10] (see Appendix). This is unlikely to cause a significant issue. A potential noise source is the green laser that gets trapped inside the diamond due to diamond's high refractive index [7]. After spending some time inside the diamond, the trapped light with an arbitrary phase eventually exits the diamond, travels with the retroreflected beam, and interferes, causing a white noiselike background.

The Faraday effect is nonreciprocal, meaning that each time light interacts with the spins of NV[−] centers, its plane of polarization gets rotated by another ϕ [19,38]. By using a separate off-resonant, e.g., 650 nm probe laser and a cavity around the diamond, the interaction between the probe light and the spins of the NV[−] centers and thus the readout fidelity can be significantly enhanced. The cavity could be formed using two independent mirrors or by coating the diamond surfaces. Cavities formed by directly coating the surfaces of the diamond might be more stable due to their monolithic nature. Such cavities might also be preferable for applications requiring high spatial resolutions, such as MEG, since the dimensions of such a cavity will be the size of the diamond itself. The number of times the probe light bounces back and forth inside the cavity is determined by the cavity finesse. Loaded cavity finesse of $> 10^4$ has been demonstrated [39,40]. The maximum time for which light can be confined within the cavity to enhance sensitivity is $\leq T_2^*$ time of the nitrogen vacancy centers.

In our experiment, we continuously pump the NV[−] centers using a green laser and resonant microwaves. This creates a competition between the spin polarization to the $m_s = 0$ state by the green laser and the microwave excitation to the

$m_s = -1$ state. This reduces the interaction time between the probe light and the spin. This can be avoided by implementing pulsed operation in which a green laser pulse is used to initialize NV^- centers to the $m_s = 0$ state, and subsequently, a microwave π pulse is applied to excite the spins to the $m_s = -1$ state. This will allow probe light to interact with the spins more efficiently and thus enhance our ability to read spin states.

The sensitivity of our magnetometer can be improved in several ways. The laser power immediately before the PBS [Fig. 1(b)] is 100 mW. This is significantly higher than our existing balanced detector can take. So we use a neutral density filter to attenuate the power. By using photodiodes capable of taking extra laser power [7], the sensitivity can be improved straightforwardly. Another major area of improvement is to use chemical vapour deposition (CVD) diamonds, which have superior T_2^* time (17.5 μ s) [41]. The sensitivity can also be improved either by addressing more NV^- centers using extra green laser power or by increasing the density of the nitrogen vacancy centers [42]. The performance of our magnetometer can also be improved by driving the hyperfine transitions [7,42]. Taking $n_t = 1 \times 10^4$, $T_2^* = 0.5 \mu$ s, and $n_s = 1 \times 10^{12}$ (10 ppm NV^- centers), the smallest magnetic field that can be detected is 8×10^{-16} T—sufficient for MEG. It also seems viable that a single NV^- center can be read out using the Faraday effect at room temperature with the inclusion of a cavity.

V. CONCLUSIONS

We have demonstrated that the spin states of NV^- centers can be read out using the Faraday effect at room temperature. Using the absorption-detected magnetic resonance, the helicity of light, and the optical selection rules, we have shown that optical transitions from the ground state to $A_{1,2}$ in the excited state play roles in the Faraday effect. Our results also indicate that the spin-orbit coupling survives thermal averaging [20]. Using the readout technique of the NV^- spin states, we have developed a magnetic field sensor that has the potential to reach femtotesla-level sensitivity. Finally, our Faraday-effect-based magnetometer is amenable to different techniques, e.g., gradiometry used in optically pumped magnetometers (OPMs)[26,43]. The benefit of a solid-state sensor combined with the techniques developed for OPMs can be used for developing technological platforms benefiting biomedical imaging, magnetic navigation, and fundamental physics.

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

APPENDIX A: EXPERIMENTAL SETUP

Figure 5 shows the optical layout of our experiment. Various components used in the experiment have been labeled in green. In the top left corner, we show individual images of the antenna and the diamond used. The size of the diamond is $2 \times 2 \times 2$ mm³. We have characterized our diamond using EPR and found that it contains 0.220 ppm of NV^- centers and 50 ppm of substitutional nitrogen. In Fig. 6, we characterize our quarter-waveplate. In this case, the quarter-waveplate is placed between the pickoff mirror and the half waveplate located immediately after the polarizer (GL10-A, Thorlabs). The polarization of the output light of the polarizer is parallel to the table, which we call x polarized. We take the x axis as our reference for measuring angles (α). The total input power before the PBS in front of the balance detector (see Fig. 1) was set to 100 mW. Figure 6 shows the transmitted power as a function of the quarter-waveplate angle. In this case, no microwave was used. The maximum transmitted power was 93 mW at $\alpha = 12^\circ$ and the minimum transmitted power was at 57° . Ideally, the minimum of the transmission should be reached at $\alpha = 45^\circ$, indicating an offset of 12° . This offset is added by the steering mirrors, the diamond, and other imperfections in the system.

APPENDIX B: NV^- CENTER AND ITS ENERGY LEVELS

The nitrogen vacancy center in diamond consists of a substitutional nitrogen atom and an adjacent vacancy in the diamond lattice. Figure 1(a) shows an NV^- center oriented along the [111] crystallographic axis. The diamond used in our experiment is a single-crystal {111} cut $2 \times 2 \times 2$ mm³ Type 1b HPHT single-crystal synthetic diamond, and contains 0.220 ppm NV^- centers and 54 ppm substitutional nitrogen. For magnetometry, we apply a homogeneous dc magnetic field (B) parallel to the [111] axis of the diamond. The ground-state manifold ($^3A_2$) of an NV^- center is an orbital-singlet spin-triplet ($S = 1$, $m_s = 0, \pm 1$) state with zero orbital angular momentum, while the excited-state manifold is an orbital doublet and has orbital angular momenta of ± 1 [15,21]. As we use an ensemble of NV^- centers, some strain is inevitable [1,21–24], which splits the excited state into two branches: ^3E_x (A_1, A_2, E_x) and ^3E_y (E_1, E_2, E_y). At relatively low strain, the $A_{1,2}$ and $E_{1,2}$ states are spin-orbital entangled states, e.g., $A_{1,2} = |E_- \rangle \otimes | + 1 \rangle \mp |E_+ \rangle \otimes | - 1 \rangle$ and $E_{1,2} = |E_- \rangle \otimes | - 1 \rangle \mp |E_+ \rangle \otimes | + 1 \rangle$ [15,21], where $E_{\pm}$ is the excited state orbital angular momentum and $| \pm 1 \rangle$ represent the spin states

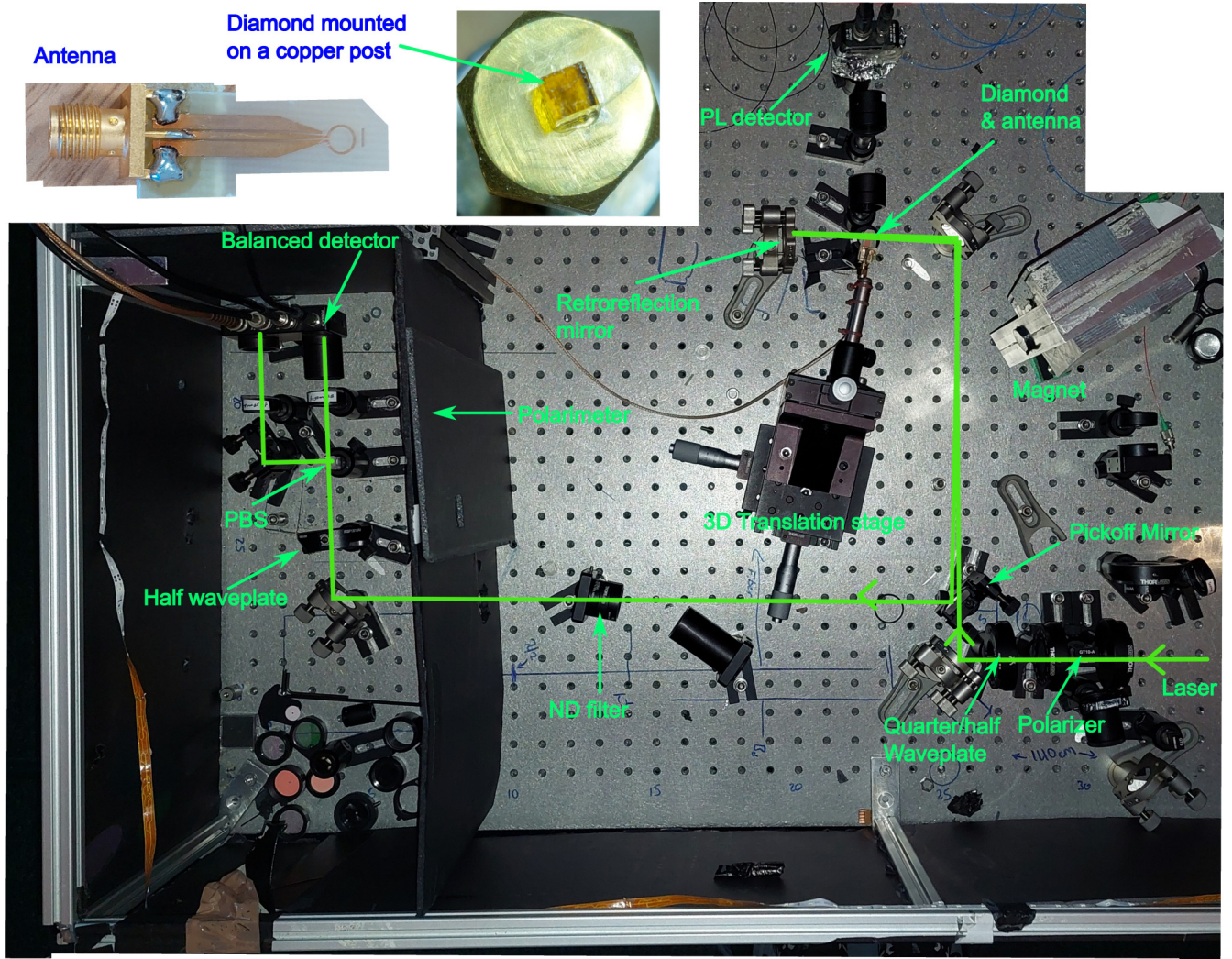


FIG. 5. The top view of our experimental setup. In the top left corner, we show the antenna and the diamond used in our experiment.

for $m_s = \pm 1$. $E_{x,y}$ states only couple to the $m_s = 0$ state of the ground-state manifold by absorbing linearly polarized light,

while the other four states can be excited from the $m_s = \pm 1$ states via circularly polarized light.

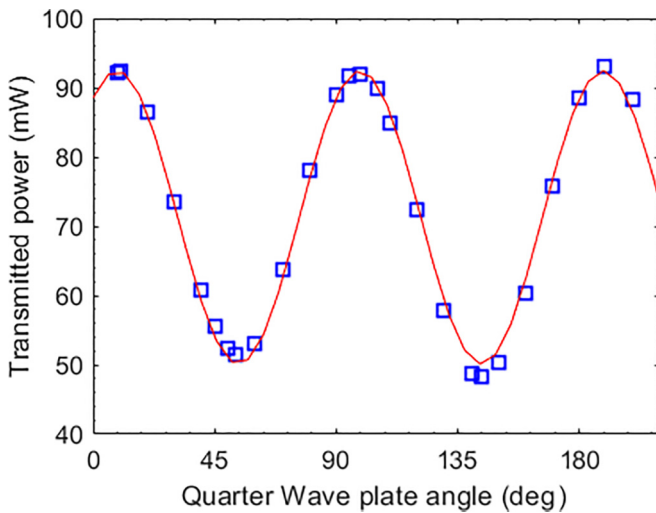


FIG. 6. Characterization of the quarter-waveplate used in our experiment.

APPENDIX C: DIPOLE RADIATION AND THE ORIENTATIONS OF THE NV^- DIPOLES

There are four different orientations of the nitrogen vacancy centers in diamond. One group is aligned along the $[111]$ crystallographic direction, while the other three make an angle 109° with the $[111]$ direction. Each of the nitrogen vacancy centers has two dipoles perpendicular to its axis. In the ideal case, the dipoles are perpendicular to each other as well. In our experiment, the $[111]$ direction is parallel to the z axis. As a result, the dipoles belonging to the NV^- center oriented along $[111]$ are in the x - y plane. Let the NV^- center orientated along the $[111]$ direction be denoted by $\text{NV}_1 = [0\ 0\ 1]$ and its two dipoles by $\mathbf{d}_{11} = [-\sin\phi_1\ \cos\phi_1\ 0]$ and $\mathbf{d}_{12} = [-\cos\phi_1\ \sin\phi_1\ 0]$, where μ is the dipole moment of the NV^- center [28,44]. In our experiment, the laser propagates along the z axis with a wave vector $\mathbf{k}$. The polarization of the laser is denoted by $\mathbf{E}_l = [\sin\beta\ \cos\alpha\ \sin\beta\ \sin\alpha\ \cos\beta]$, while our photoluminescence detector, placed along the x axis, is denoted by $\mathbf{D}_0 = [\sin\theta_5\ \cos\phi_5\ \sin\theta_5\ \sin\phi_5\ \cos\theta_5]$.

The dipoles of the remaining NV^- centers are located on planes that make an angle 71° with the x - y plane. We denote these NV^- centers and their dipoles [28] by

$$\begin{aligned}
 NV_2 &= [\sin \theta_2 \cos \phi_2 \ \sin \theta_2 \sin \phi_2 \ \cos \theta_2], \\
 \mathbf{d}_{21} &= \mu[-\sin \phi_2 \ \cos \phi_2 \ 0], \\
 \mathbf{d}_{22} &= \mu[\cos \theta_2 \cos \phi_2 \ \cos \theta_2 \sin \phi_2 \ -\sin \theta_2], \\
 NV_3 &= [\sin \theta_3 \cos \phi_3 \ \sin \theta_3 \sin \phi_3 \ \cos \theta_3], \\
 \mathbf{d}_{31} &= \mu[-\sin \phi_3 \ \cos \phi_3 \ 0], \\
 \mathbf{d}_{32} &= \mu[\cos \theta_3 \cos \phi_3 \ \cos \theta_3 \sin \phi_3 \ -\sin \theta_3], \\
 NV_4 &= [\sin \theta_4 \cos \phi_4 \ \sin \theta_4 \sin \phi_4 \ \cos \theta_4], \\
 \mathbf{d}_{41} &= \mu[-\sin \phi_4 \ \cos \phi_4 \ 0], \\
 \mathbf{d}_{42} &= \mu[\cos \theta_4 \cos \phi_4 \ \cos \theta_4 \sin \phi_4 \ -\sin \theta_4],
 \end{aligned} \tag{C1}$$

where $\theta_2 = \theta_3 = \theta_4 = 109^\circ$, $\phi_3 = \phi_2 + 2\pi/3$, and $\phi_4 = \phi_2 + 4\pi/3$. The angle between an NV^- dipole, say $\mathbf{d}_{11}$, and the detector is $\gamma_{11} = \cos^{-1}[\mathbf{d}_{11} \cdot \mathbf{D}_0]$. The intensity of photoluminescence radiated by a general dipole $\mathbf{d}$ is given by [28,45]

$$I = C_0 |\mathbf{E}_l \cdot \mathbf{d}|^2 \sin^2 \gamma, \tag{C2}$$

where γ is the angle between the dipole and the point of observation, and C_0 is a constant.

In our experiment, we use an ensemble of NV^- centers spread across the diamond. For our simple model, we assume that NV^- centers emit photoluminescence incoherently and that they, as a whole, act as a point source. In this simplified model, the intensity of PL is

$$\begin{aligned}
 I &= NC_0 \int_{\frac{\pi}{2}-\delta}^{\frac{\pi}{2}+\delta} \int_{-\delta}^{\delta} \sum_{i=1}^4 \sum_{j=1}^2 (\mathbf{E}_l \cdot \mathbf{d}_{ij})^2 \sin^2(\gamma_{ij}) d\phi_5 d\theta_5 \\
 &= NC_0 \int_{\frac{\pi}{2}-\delta}^{\frac{\pi}{2}+\delta} \int_{-\delta}^{\delta} \sum_{i=1}^4 \sum_{j=1}^2 (\mathbf{E}_l \cdot \mathbf{d}_{ij})^2 \sin^2(\cos^{-1}[\mathbf{d}_{ij} \cdot \mathbf{D}_0]) \\
 &\quad \times d\phi_5 d\theta_5,
 \end{aligned} \tag{C3}$$

where N is the total number of radiating NV^- centers and δ is an angle determined by the numerical aperture of the lens placed before our PL detector. The focal length of this lens is 35 mm and has an effective numerical aperture of ≈ 0.15 or $\delta \approx 9^\circ$. Note that since our PL detector is placed along the x axis, $\theta_5 = \pi/2 \pm \delta$ and $\phi_5 = \pm\delta$, since we take the x axis as reference. On performing integration and keeping terms only in second order of δ and taking $\beta = \pi/2$, we have

$$I \approx \frac{NC_0 \delta^2}{16} [85 - 40 \cos 2\alpha - 16 \cos (2\alpha - 4\phi_1)].$$

Notice that I does not depend on ϕ_2 , ϕ_3 , and ϕ_4 , the angles related to the dipoles belonging to the NV^- centers along all crystallographic axes except the [111] direction. In the main text, to determine the orientation of the dipoles belonging to the [111] axis, we fit a function of the form $A - B \cos 2\alpha - C \cos (2\alpha - 4\phi_1)$ with our experimental data and retrieve ϕ_1 .

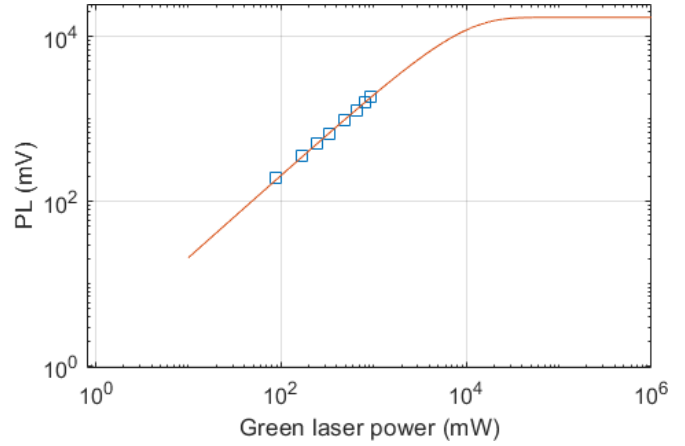


FIG. 7. Green laser power vs PL with a fit of the form $I = I_0(1 - e^{-\Gamma P})$, where P is the laser power and Γ is a constant.

APPENDIX D: SATURATION GREEN LASER POWER AND THE NUMBER OF NV^- CENTERS EXCITED

The amount of green laser power required to excite all the NV^- centers can be found by fitting a function of the form $I = I_0(1 - e^{-\Gamma P})$ to the measured photoluminescence (Fig. 7), where Γ is a constant and P is the laser power. In this case, the green laser was sent through the diamond once. From the fit, we find that the required power to excite all NV^- centers in the path of the green laser beam is ≈ 100 W. In our experiment, the maximum green laser power, after deducting the reflection (20%) at the entrance surface of the diamond, is 0.91 W. This leads us to conclude that we are interrogating about 1% of all the NV^- centers available in our diamond. This has been used in the calculation of the NV^- number density in the main text.

APPENDIX E: ROTATION ANGLE: A PHENOMENOLOGICAL ANALYSIS

When linearly polarized probe light, here the green laser, propagates through a spin-polarized medium, its $\hat{\sigma}_+$ and $\hat{\sigma}_-$ polarized components encounter different indices of refraction, which in turn rotate the plane of polarization of the green laser. The complex indices of refraction n_+ and n_- of the left- and right-circularly polarized light are [17,46]

$$n_{\pm} = n_0 + \zeta(1 \pm \langle S_z \rangle \hat{z} \cdot \hat{k}), \tag{E1}$$

where n_0 is the refractive index of the diamond with no nitrogen vacancy center is present, and $\zeta = \frac{\rho_n r_e f_s c^2 L(\nu)}{4\pi \nu}$ with ρ_n being the number density of NV^- centers, c being the speed of light, and r_e being the classical electron radius. f_s and $L(\nu) = 1/(\nu - \nu_0 + i\Gamma)$ are the oscillator strength and the absorption spectral profile of an NV^- center with ν being the laser frequency, ν_0 being the optical resonance frequency of the NV^- center, and Γ being the linewidth of the optical resonance. $\langle S_z \rangle = \sum_s m_s P_s$ is the average spin polarization of NV^- centers along the z axis with P_s being the probability of an NV^- center in the s state. The relative phase $\phi = n_t k L \Re n_+ - n_-$ acquired by the $\hat{\sigma}_+$ and $\hat{\sigma}_-$ polarized light after traveling n_t times through the diamond of length L is

$$\phi = 2n_t k L \Re \zeta \langle S_z \rangle \hat{z} \cdot \hat{k}, \tag{E2}$$

where $k = 2\pi\nu/c$. For $\langle S_z \rangle = \pm 1$, ϕ is greater/less than zero, giving rise to the opposite polarities of the FDMR peaks shown in the main text. When the Faraday rotated light is impinging on the polarizing beam splitter of the balanced polarimeter, the output of the balanced detector [26] is

$$\Delta I \approx 2I\phi, \quad (\text{E3})$$

where $I = P/\pi r^2$ with P being the input power to the polarimeter and r is the radius of the laser beam. We have also used $|\phi| \ll \pi/4$, which is valid in our experiment. The output of the balanced detector is fed to an LIA for low-noise detection [42].

APPENDIX F: CALIBRATION OF THE ROTATION ANGLE

When the Faraday effect is large, the rotation angle ϕ can be found by directly measuring the input laser power to the polarimeter and the difference in power between the two outputs of the PBS (see Fig. 1). Since in our case ϕ is small, it is impacted by laser noise and mechanical vibrations. Consequently, we use the balanced polarimeter output in combination with an LIA to improve the signal to noise ratio (SNR). The output of the lock-in amplifier is in volts. As a result, a calibration constant is needed to convert voltage signals into angles. Moreover, the lock-in amplifier output is in the form of a derivative of the actual signal. Consequently, an integration is required to retrieve the desired signal.

To calibrate our lock-in amplifier, we use the raw photoluminescence as a training signal, as we can measure it directly. The PL signal is subjected to the same frequency modulation of the microwave associated with the lock-in detection as the FDMRs. We first record the raw PL signal as a function of microwave frequency. In the next experiment, we input the PL signal to the lock-in amplifier and record the output of the LIA. As the first step, to find the calibration factor C , we perform cumulative numerical integration on the output ($S_I(f)$) of the lock-in amplifier using the following equation:

$$PL(f) = A + C \int_{f-\Delta f}^{f+\Delta f} S_I(f) df, \quad (\text{F1})$$

where A is equal to the PL when the microwave is off-resonant. We match the result of the integration with the raw PL recorded before and thus find the calibration factor. We use this calibration factor to convert the LIA output corresponding to the FDMRs and find the rotation angle ϕ shown in the main text [Fig. 4(b)].

APPENDIX G: FARADAY EFFECT AT DIFFERENT MICROWAVE AND GREEN LASER POWER

Figure 8 shows FDMRs for different microwave powers at a fixed green laser power of 1150 mW in the retroreflection geometry. As the microwave power increases (bottom figure, red squares are data points), the FDMR approaches a maximum value. This is expected. When there are no microwaves, about 80% of the spin population is in the $m_s = 0$ state in the ground-state manifold. As the microwave power increases, more and more spin population is transferred to the $m_s = -1$ state, increasing the Faraday effect. As the microwave power continues to rise, an equilibrium is established between the

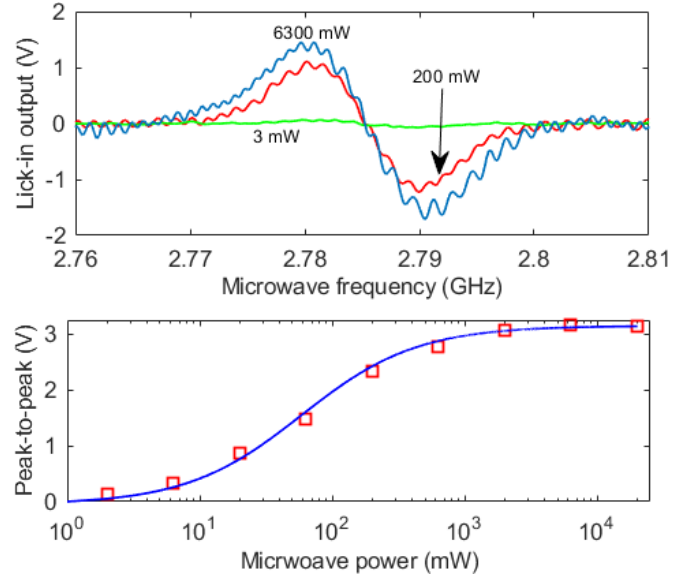


FIG. 8. Effect of microwave power on FDMRs when the $m_s = 0 \rightarrow -1$ transition is pumped. The top figure shows FDMRs for three different microwave powers. The bottom figure shows the peak-to-peak height of the FDMR peaks for different microwave powers: Red squares are experimental data, while the solid blue line is the rate equation-based fit.

green laser pumping rate to the excited-state manifold (^3E) and the microwave pumping rate within the ground-state manifold. At the highest microwave power (20 W or 43 dBm) used for this experiment, the FDMR peaks get distorted and the linewidth of the FDMR/ODMR increases by about 6%. To avoid a significant heating induced by microwaves, we operate our main experiment at 33 dBm (2 W), and all data presented in the main text were collected at this power.

In Fig. 9, we show FDMRs as we change the green laser power inputted to the diamond while keeping the laser

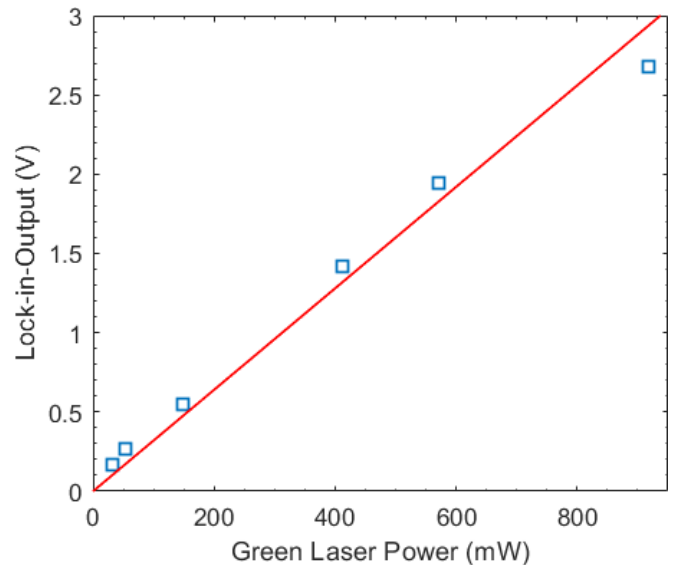


FIG. 9. Green laser power vs FDMRs for a fixed microwave power of 33 dBm.

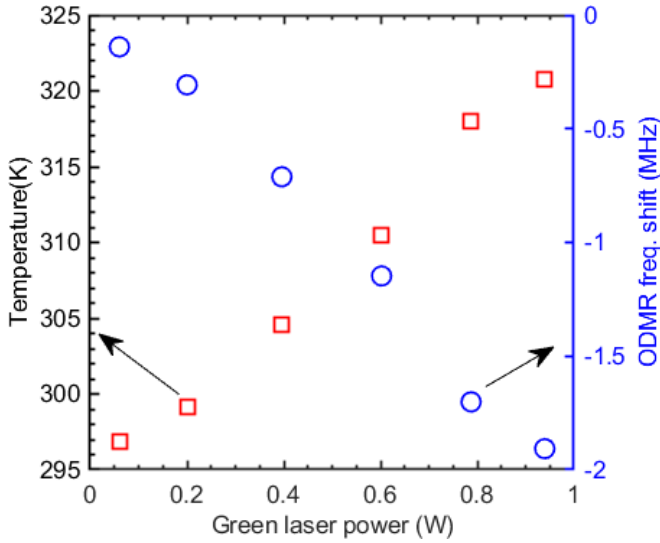


FIG. 10. Laser- and microwave-induced heating of diamond measured using the photoluminescence-based ODMR shifts assuming the diamond is at room temperature (295 K) when the microwave and laser powers are 18 dBm and 40 mW, respectively. All data points were collected at a fixed microwave power of 33 dbm.

polarization and the microwave power fixed (33 dBm). For this experiment, the input power to the balanced polarimeter [see Fig. 1(b)] was kept fixed at 4 mW using an adjustable neutral density filter. Within the maximum laser power available, we do not see any saturation of the FDMR. This is consistent with the measured photoluminescence as a function of the green laser power shown in Fig. 7, which does not saturate either.

APPENDIX H: LASER- AND MICROWAVE-INDUCED HEATING

In Fig. 10, we show the microwave- and laser-induced heating [10]. We find the temperature of the diamond by measuring the shifts of the photoluminescence-based ODMR resonance frequency [10]. We assume that the diamond is at room temperature when the microwave and laser powers are

set to 18 dBm (about 30 times smaller than the power used in the main text) and 40 mW (25 times smaller than the usual green laser power used), respectively. Subsequently, we fixed the microwave power to 33 dBm and varied the green laser power. At the maximum laser power used, the temperature of the diamond rises by 30 K.

APPENDIX I: NV⁻ CENTER POPULATION DISTRIBUTION—A RATE EQUATION-BASED ANALYSIS

To understand the population dynamics of the NV⁻ center at different laser and microwave powers, we performed a rate equation-based simulation. Although a ten-level model is ideal, a seven-level model captures the same physics [23]. In the seven-level model, there are three energy levels in the ground state: $x(m_s = -1)$, $y(m_s = +1)$, $z(m_s = 0)$, three energy levels in the excited state: $X(m_s = -1)$, $Y(m_s = +1)$, $Z(m_s = 0)$, and one singlet level S . Various transition rates are denoted by k_{xx} , k_{yy} , k_{zz} , etc. For example, k_{zz} denotes the transition rate from the z state in the ground-state manifold to the Z state in the excited-state manifold. Transition rates ($1/\tau$) vary in the literature [47,52], where τ is the excited lifetime. The transition rates and lifetimes used in our calculation are shown in Table I and are within the accepted values in the literature [47,48,52,53]. Rates that have not been listed in Table I have been set to zero. For the optical pumping rate, we estimate that, in the double-pass geometry, there is about 1.3 W of laser power inside the diamond at any point in time. We also estimate that the green laser beam diameter is ≈ 1 mm.

Using the transition rates listed in Table I, we calculate the photoluminescence $I(q) = \hbar\omega \sum_i \sum_j n_i k_{ij}$ with $i \in (X, Y, Z)$, $j \in (x, y, z)$, n_i denotes the NV⁻ population in state i , q is the microwave pumping strength, and ω is the frequency of the photon emitted by an NV⁻ center. For our model, we take all photons emitted to be of a single frequency. We define PL contrast as

$$C_q = \frac{I(q=1) - I(q)}{I(q=1)}. \quad (11)$$

TABLE I. Parameters for the rate equation-based calculation.

Parameters	Value
k_{zz}	80×10^6 Hz, lifetime 12.5 ns [47]
k_{xx}, k_{yy}, k_{zz}	125×10^6 Hz, lifetime 8 ns [47]
$k_{xz}, k_{yz}, k_{zx}, k_{zy}$	1.5×10^6 Hz [23]
k_{xs}, k_{ys}	50.7×10^6 Hz [48]
k_{zs}	6.4×10^6 Hz [48]
k_{sx}, k_{sy}	1×10^6 Hz [48]
k_{sz}	4.6×10^6 Hz, lifetime 219 ns [49]
Optical pumping, e.g., $k_{xx} = k_0 k_{xx}, k_{xz} = k_0 k_{xz}, \dots$	$k_0 = R_0/k_{zz}$, $R_0 = \frac{\sigma I}{\hbar\omega}$ with $\sigma = 3.1 \times 10^{-21}$ m ² being the absorption cross section [27] of the NV ⁻ center and I is the laser intensity
Transitions within triplets e.g., $k_{zy}, k_{zx} \dots$	$1/2T_1$ [50] with $T_1 = 3.0$ ms [51]
Microwave pumping: $k_{zx} = k_{xz} = q/2T_1$ [48]	q is a free parameter

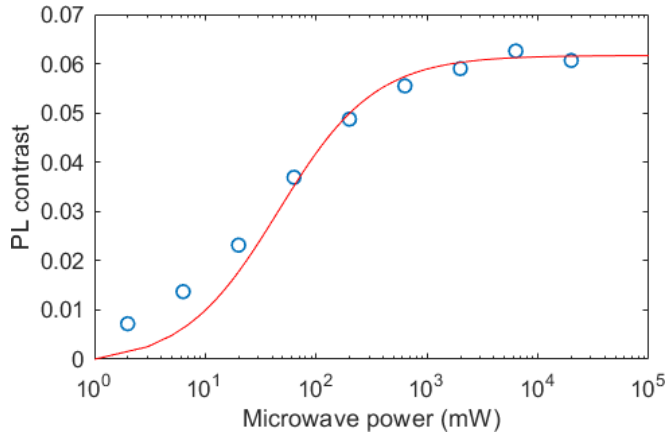


FIG. 11. Photoluminescence contrast as a function of the microwave power for the $m_s = 0 \rightarrow -1$ transition. Circles are experimental data points, while the red solid line is the rate equation-based fit.

We vary q and find C_q that matches our experimentally measured PL contrast shown in Fig. 11 (blue circles). The match between the experiment and the simulation is reasonably good. The data presented in Fig. 4 are at a microwave power of 33 dBm (2 W). At this microwave power, we find that about 43% (13%) of the NV^- centers are in the $m_s = -1$ ($m_s = +1$) states. The net spin polarization is then 30%, which is used in calculating the NV^- number density in the main text.

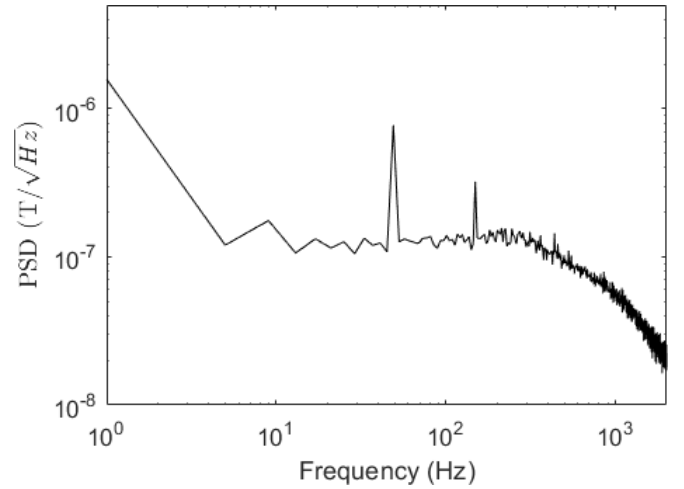


FIG. 12. Magnetic field sensitivity when the photoluminescence-based ODMR is used.

APPENDIX J: MAGNETIC FIELD SENSITIVITY WITH THE PHOTOLUMINESCENCE-BASED ODMR

Although demonstrating that the spin states of the NV center can be read out using the Faraday effect at room temperature is our main goal in this proof-of-principle experiment, for completeness, in Fig. 12, we show the magnetic field sensitivity when we use the traditional ODMR-based detection scheme for reading out the spin states of the NV center. In this case, we get a sensitivity of 120 nT, slightly better than the sensitivity we have found using the FDMR-based detection scheme.

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