Room temperature optically detected magnetic resonance of single spins in GaN

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Abstract

High contrast optically detected magnetic resonance (ODMR) is a valuable property for reading out the spin of isolated defect color centers at room temperature. Spin-active single defect centers have been studied in wide bandgap materials including diamond, SiC, and hBN; each with associated advantages for applications. We report the discovery of ODMR in two distinct species of bright, isolated defect centers hosted in GaN. In one group, we find negative ODMR of a few percent associated with a metastable electronic state, whereas in the other, we find positive ODMR of up to 30% associated with the ground and optically excited electronic states. We examine the spin symmetry axis of each defect species and we establish coherent control over a single defect's ground-state spin. Given the maturity of the semiconductor host, these results are promising for scalable and integrated quantum sensing applications.

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Optically detected magnetic resonance[1,2] (ODMR) is an efficient mechanism to readout the spin of solid-state color centers at room temperature, thus enabling spin-based quantum sensors of magnetic field,[3–6] electric field,[7] and temperature[6,8,9] with high sensitivity and broad commercial applicability. The mechanism of room temperature ODMR is based on spin-dependent relaxation between the optically excited states to the ground states, and thus it is an intrinsic property of a defect center. While the diamond nitrogen-vacancy (NV) center is the most prominent example,[10,11] room temperature ODMR has also been discovered in silicon vacancy centers[12] and divacancy centers[13] in SiC, and recently in boron vacancy center ensembles[14,15] and unidentified single defects[16,17]in hexagonal boron nitride (hBN). Of these material systems, diamond NV centers are the most technologically important owing to their large (20–30%) ODMR contrast, long spin coherence, high quantum efficiency, and high brightness.[18] Unfortunately, diamond as a substrate is far from being technologically mature. For example, diamond is unavailable with high crystalline quality in large-scale wafers and lacks hetero-epitaxial integration with semiconductors for integrated sensor technologies. Likewise, boron vacancy centers in hBN have large contrast (up to 20%),[15,19,20] however, they are available only as small flakes, have low quantum efficiency,[21,22] and lack a visible zero-phonon line at room temperature.[23] Silicon carbide is a technologically mature substrate with recent advances in scalable monolithic integration of color-center-based quantum light sources.[24] A recent work demonstrates high contrast ($\sim$30%) in coherent manipulation of a PL6 divacancy defect,[25] however, most SiC defects exhibit low contrast room temperature ODMR (under 2%).[12,13,26,27]

GaN has emerged as a semiconductor of choice for power electronics owing to its wide direct bandgap and high breakdown field.[28–31] Recently, it has also been found to host bright single photon emitters with spectrally narrow photoluminescence (PL) in the visible spectrum.[1,2] These defect centers have large room temperature saturation photon count rates on the order of 800 kCt/s in a solid-immersion lens at a saturation power of $\approx$ 1 mW, and they have zero phonon linewidths of a few meV at room temperature and less than 1 meV at cryogenic temperatures.[3,35,36] In contrast to NV centers in diamond, these defect centers in GaN have large Debye-Waller factors > 0.5. These excellent optical properties, combined with the engineerability of GaN make these single-photon emitting defects attractive for on-chip photonics and quantum technologies that require single-photon sources. The atomic structure of these defects has not yet been identified.

In this work, we report that bright single photon emitters in GaN possess spin $S \geq 1$ and exhibit magnetic field dependent PL and ODMR with up to ∼30% contrast at room temperature. Our study reveals at least two distinct groups of defects, each with a distinct ODMR spectrum as well as sign of ODMR contrast. Finally, we demonstrate coherent Rabi driving and extract a Rabi coherence time on the order of 100 ns. This is promising for sensing applications owing to the high ODMR contrast hosted by a mature semiconductor platform, and it is also promising for unraveling the atomic structure of these defect types by providing critical information about the defect orientation within the crystal, spin multiplicity, and sign of the ODMR response.

Magneto-photoluminescence of single defects in GaN

Figure 1(a-c) detail the typical room temperature optical properties of an isolated GaN defect used in our study. The defects are optically separated on the scale of a few micrometers, enabling photon correlation measurements to ensure we examine single defects. A solid-immersion lens aids in photon collection, with a typical rate of 80 kCounts/s into a 0.9 NA microscope objective when excited with a 532 nm laser with 20 μW power. Defect #2 emits most of its PL into a narrow linewidth centered near 667 nm. As noted previously,[1–3] not all GaN defects share the same emission energy. Additionally, while these defects are mainly photo-stable, like most solid-state single photon emitters, these defects suffer some instabilities, including occasional photo-bleaching. Additional information is detailed in Supplementary Note 1, Supplementary Table 1 and Supplementary Figures 1 and 2.

A simple method of screening a particular defect for spin-dependent optical properties is measuring its magnetic field dependent PL (magneto-PL).[37,38] Although the specific magneto-PL response depends on the angle of the magnetic field with respect to the defect spin quantization axis, we select the GaN c-axis as a potential direction of high symmetry. The result for seven individual defects is shown in Fig. 1(d). We immediately notice that the defects fall into two groups of behavior. In the first group, there is a ∼7% dip in PL at low magnetic fields, followed by an increase of PL to saturation (#1 and #5, which we label group I). In the second group, the PL falls monotonically with magnetic field, showing up to a 30% change in PL (#2 – #4, which we label group II).

We proceed under the initial assumption that these GaN defect groups have a spin-

dependent PL mechanism similar to that of the diamond nitrogen-vacancy center, in which a spin-state-dependent intersystem crossing can occur from the excited states to a metastable state (Fig. 1(e)), which ultimately creates spin-dependent PL contrast.[37] It is also possible to obtain spin-dependent PL even if the ground and excited states are spin singlets or doublets if there is spin-dependent relaxation from a $S \geq 1$ metastable states (Fig. 1(f)).[38,39] In both cases, the precise spin contrast results from a competition between radiative and non-radiative relaxation rates, branching ratios, and optical pumping rates. While the full characterization of the GaN defect optical cycle is beyond the scope of this work, we re-examine some details of the spin-dependent optical cycle below.

Regardless of the specific mechanism of spin-dependent PL contrast, magneto-PL originates from Zeeman-induced spin state mixing between spin states with different average PL rates. This mechanism is relevant to systems with electronic spin $S \geq 1$. The ground state Hamiltonian of a spin system with $S \geq 1$ in a magnetic field B is given by

$$\mathcal{H} = DS_z^2 + E(S_x^2 - S_y^2) + g\mu_B \vec{S} \cdot \vec{B}, \tag{1}$$

where S is the electronic spin operator, g the electronic g-factor, μ_B the Bohr magneton, D and E together the zero-field interaction parameters. Here we ignore coupling to nuclear spins for simplicity. An angle between the external field $\vec{B}$ and the spin quantization axis introduces off-diagonal large matrix elements between the spin eigenstates, mixing them. The spin eigenstates can also mix at low fields if $E \neq 0$.

Returning to the magneto-PL measurements of Fig. 1(d), the group-I magneto-PL response suggests a Zeeman-induced spin degeneracy at low magnetic fields, hinting $S \geq 1$ with a value of D under a few hundred megahertz for an electron spin with $g = 2$, depending somewhat on the alignment of the magnetic field to the spin quantization axis. Additionally, it suggests that optical pumping puts defects in this group into a state with higher PL, while spin mixing reduces the overall PL; a situation similar to diamond NV centers. While group-II defects also must have $S \geq 1$, in contrast to group-I defects, the magneto-PL is monotonically decreasing. This could be explained by two possibilities. The first is that the magnetic field is severely misaligned with respect to the defect symmetry axis, the transverse magnetic field mixes the spin states, and thus spin-mixing occurs for such a large range of magnetic fields. In this case, spin mixing would lower the PL. The second scenario is that the spin states are already mixed at low magnetic field and the magnetic field is reducing

their mixing. Here, optical pumping would be initializing the system into a state that emits less PL than the other states, and thus the PL would become lower at higher fields as the initialization state becomes less mixed. More inputs are required to distinguish between these two cases, which we will discuss below and further in Supplementary Note 2.

Spin quantization axes

Having confirmed that both groups of individual defects have spins with $S \geq 1$ and a spin-dependent optical cycle, we study the spin-resonant transitions and spin Hamiltonian by measuring continuous wave (cw-) ODMR. To study the spin resonance, we continuously drive a microwave magnetic field, optically pump the defect optical transition, and count the emitted PL. Figure 2 shows the resulting cw-ODMR traces at $B = 1$ kG for a group-I (#1) and a group-II (#2) defect. We immediately notice that the two groups have an opposite sign of ODMR contrast, as suggested by the magneto-PL, with group-I defects showing negative cw-ODMR contrast and group-II defects showing positive cw-ODMR contrast. We also notice that the group-I defect has a modest contrast of ∼2% at this driving power, while the group-II defect has a ∼30% contrast for one of the three resonance features, with smaller contrast for two other features.

A key input for establishing the identity of a new defect is its spin quantization axis. Having discovered a reliable cw-ODMR signal on multiple GaN single defects, we now make the assumption that the cw-ODMR contrast will be largest when we align the external magnetic field along the z-axis defined by Eqn. 1. A misaligned static field will mix the spin eigenstates, which will reduce the cw-ODMR contrast if the fluorescence contrast mechanism is tied to $|m_s|$ as it is for the diamond NV center. To test this we systematically vary the polar angle θ with respect to the c-axis of the crystal, and then the azimuthal angle ϕ, which is measured with respect to the a-lattice vector of GaN. Fig. 2(c) and (d) show the ODMR contrast for defect #1 and #2, respectively, as a function of θ, while the corresponding data as a function of ϕ is shown in Supplementary Figure 3 in Supplementary Note 3. We find that the spin quantization axis for the group-I defect #1 forms a ∼27-degree angle with the GaN crystal c-axis, with an in-plane component points along the a-axis. For the group-II defect #2 we find a spin quantization axis approximately 10 degrees away from the c-axis, and an in-plane component along the a-axis. Neither spin quantization axis matches

a vector between a lattice site and its nearest few neighbors, suggesting the involvement of interstitial atoms (Fig. 2(e)-(f)).

Optically detected magnetic resonance spectra

Now we study the Zeeman effect on the spin levels. First, we align our set-up so that $\vec{B}$ is parallel to the direction of the largest ODMR contrast discussed above and record ODMR as a function of B. Under these conditions we assume $B = B_z$ from Eqn. 1. Figure 3 shows the resulting cw-ODMR data from defect #1 (group I) and defect #2 (group II) from 100 G to 1500 G. The most visible spin resonances disperse with a g-factor $g = 2$, confirming that we study electronic spins.

First focusing on defect #1, we see two transitions of unequal contrast that appear at $B \gtrsim 250$ G. The lack of cw-ODMR contrast at low magnetic fields suggests a mixing between the spin eigenstates that leads to the suppression of spin contrast. If we assume a minimum spin multiplicity to explain the two transitions, $S = 1$, then this data can be described by Eqn. 1 with $D \approx E \approx 389$ MHz. An overlay of the fitted spin transitions is shown in Supplementary Figure 4(a) in Supplementary Note 4. Under these conditions at low magnetic fields, the zero-field spin eigenstates would indeed be strongly mixed, thus suppressing spin-dependent optical contrast. We note, however, that this scenario does not explain why the two transitions have unequal contrast, which may relate to dynamics of the optical cycle that have not been revealed by these measurements. Additionally, we find that the model deviates from the data at the lowest magnetic fields, which may point to other physics not contained in a toy model of a single electronic spin-1. For example, the Ga and N atoms that surround the defect all have a nonzero nuclear spin, which may interact very strongly with this defect and thus potentially explain a deviation from a simple electronic model. Additionally, we note that group-I defects are rare compared to those in group II. While we observed magneto-PL for two defects in this group, one of those stopped being optically active, and thus defect #1 is the only group-I defect that we have been able to record ODMR (See more in Supplementary Note 1).

Next, we examine the field-dependent cw-ODMR of defect #2, which has the same cw-ODMR spectrum as all of the group-II defects that we studied. Data for other defects can be found in Supplementary Figure 4. This defect shows three spin transitions that disperse

with $g = 2$, making spin $S = 3/2$ a minimal model assuming that there is an ODMR contrast mechanism for all $\Delta m_s = 1$ transitions. Again we note that the three transitions have unequal contrast. The strongest cw-ODMR feature extrapolates to zero frequency at zero field within experimental uncertainty, suggesting that it is due to a transition between $\left|m_s = -\frac{1}{2}\right\rangle$ and $\left|m_s = +\frac{1}{2}\right\rangle$ in the minimal model. In addition to the $g = 2$ resonance, we also see a 4th resonance that disperses with $g = 4$. This feature has a zero-frequency intercept at $B = 0$ and it appears to have an avoided-crossing with the highest frequency $g = 2$ spin resonance at $B \sim 300\,\mathrm{G}$ and $f_{\mathrm{mw}} = 1.5$ GHz. Although a $g = 4$ resonance can be explained by a $\Delta m_s = 2$ spin transition, that scenario does not give rise to an avoided-crossing or a zero intercept, suggesting that a toy electronic model based on Eqn. 1 is insufficient to describe this spin system if the magnetic field is aligned along the symmetry axis. If we ignore the $g = 4$ resonant line, these transitions are well-described for $B > 0.5$ kG by a $S = 3/2$ model with $D = 368$ MHz and $E = 0$ as shown in Supplementary Figure 4(b) (see further discussion in Supplementary Note 4). Lastly, this picture is consistent with the magneto-PL data shown in Fig. 1(d), because optical pumping initializes the system into a state emitting less PL when the magnetic field is large enough.

Identifying the electronic states associated with ODMR

We now return to the question of whether the spins associated with these defects are in the ground-state and excited-state manifold as in the case of the diamond NV center (Fig. 1(e)), or whether they are associated with a metastable state (Fig. 1(f)) as in the case of the diamond ST1 defect.[39] To clarify that assignment, we perform both pulsed ODMR and time-resolved single photon counting experiments with separate microwave spin manipulation and optical excitation. The pulse timings are detailed in Supplementary Figure 5. If we manipulate a ground-state spin, then we expect the pulsed ODMR scheme shown in Fig. 4(a) to result in a visible spin resonance. However, if there is no contrast, then we can assign the cw-ODMR response to a metastable spin state.

We start with pulsed ODMR of defect #1 from group I, shown in Fig. 4(b). We observe no spin resonance response, with the noise floor of our integration at the level of 0.2%. Comparing this figure to the $\sim$2% contrast that we observed for cw-ODMR, we conclude that this defect likely has a ground-state/excited-state singlet or a ground-state doublet with

no ODMR contrast. Thus, the $S \geq 1$ spin state that gives rise to cw-ODMR must reside in a metastable state. We confirm our conclusion that ground-state microwave preparation has no impact on the PL using direct time-resolved single photon counting (Fig. 4(c)). We see that after turning on the laser, defect #1 has a microsecond-timescale reduction in PL as a function of time, however, we note no difference between the curves generated by a laser pulse alone and a microwave pulse followed by a laser pulse. These data, along with measurements of $g^{(2)}$ of this defect that shows photon bunching (see Supplementary Figure 2), support the existence of a metastable state, and are consistent with the picture of a $S \geq 1$ metastable state. Further work will be necessary to pin down all the rates in the optical cycle; however, these measurements all point to an optical cycle like that schematically shown in Fig. 1(f).

Next we repeat this series of measurements on a member of group II, defect #2, and find the opposite result in Fig. 4(d). Here we find visible pulsed ODMR contrast, confirming that the cw-ODMR measurements are the result of a ground-state spin. Interestingly, while the pulsed ODMR contrast is lower than the cw-ODMR owing to different details of the measurement protocol, we see that the same ratio of contrast between the three $\Delta m_s = 1$ transitions is preserved (See Supplementary Note 5 for further discussion). This suggests that there are non-trivial spin-dependent intersystem crossing rates, and in particular they are not proportional to $|m_s|$ as in the case of diamond NV centers. We perform the time-resolved PL measurement of defect #2 as before, with the microwave pulse tuned to the largest-contrast resonance. As expected, we see a noticeably larger initial PL response when we manipulate the ground-state spin before the laser pulse than when we do not, with a contrast lasting for $\sim$2 μs. While this experiment does not establish all the details of the optical cycle and spin, it is consistent with a level diagram and dynamics as shown in Fig. 1(e).

Coherent control of a group II single spin

Finally, we demonstrate coherent Rabi oscillations of a group-II defect (#6). We follow a common pulsed timing scheme for Rabi measurements, shown in Fig. 5(a). We work at a magnetic field in which the spin transitions are well separated such that we probe each of the $g = 2$ transitions independently. Figures 5(b-d) show the resulting Rabi oscillations of the lower-, intermediate-, and higher-frequency spin transitions respectively. We attribute the

difference between the Rabi frequencies on each transition to the frequency dependence of the antenna's power coupling. The lower- and higher-frequency transitions have a Rabi spin coherence time of $T_{2,R} = 108 \pm 4$ ns and 113 ± 5 ns, respectively, whereas the intermediate-frequency resonance has $T_{2,R} = 41 \pm 4$ ns (see Supplementary Figure 6, Supplementary Table 2, and Supplementary Note 6 for additional details). These coherence times are comparable to those measured from single spins in h-BN, which also has a large abundance of nuclear spins in the lattice.[40]

The measured trend in Rabi coherence times is also roughly consistent with the cw-ODMR linewidths we observed in Fig. 2(b), which are 82, 234, and 60 MHz, respectively, for the lowest- to the highest-frequency spin resonances. The origin of this disparity in spin coherence is unclear based on the current experimental work. One potential explanation is that the intermediate-frequency spin transition, which corresponds to $\left|m_s = -\frac{1}{2}\right\rangle \leftrightarrow \left|m_s = +\frac{1}{2}\right\rangle$ for a spin-3/2 model, is resonant at all fields with the bath of $S = 1/2$ spins due to electron charge traps that are presumably abundant in the environment of the defect. This situation would enable energy-conserving spin flip-flops and thus increase the rate of spin relaxation out of these states relative to the other spin states. On the other hand, the transitions corresponding to $\left|m_s = -\frac{3}{2}\right\rangle \leftrightarrow \left|m_s = -\frac{1}{2}\right\rangle$ (the lower-frequency resonance) and $\left|m_s = \frac{1}{2}\right\rangle \leftrightarrow \left|m_s = \frac{3}{2}\right\rangle$ (the higher-frequency resonance) are not resonant with a $S = 1/2$ spin bath. Meanwhile, the naturally abundant isotopes of Ga and N have nuclear spins of $I = 3/2$ and $I = 1$, respectively. We speculate that hyperfine interactions with the nuclear spin bath dominate the coherence time of these transitions.

In conclusion, we report high-contrast optically detected spin resonance of GaN single defect spins at room temperature. We find two distinct defect groups that we categorize based on their magneto-PL and ODMR spectra. They display complex optical cycles and spin resonance behavior that will require further investigation to understand fully; however, this work establishes key facts about these defect groups. The first group has a small negative ODMR contrast, with spin at least $S = 1$ in its metastable state to explain the experimental results. The second group has a large (up to 30%) positive ODMR contrast, with a complicated ground-state spin Hamiltonian including at least $S = 3/2$. Additionally, through angle-dependent cw-ODMR measurements, we establish a spin quantization axis in terms of the magnetic field angle with the largest ODMR contrast. The spin quantization axes of both groups do not connect neighboring GaN lattice sites, suggesting the involvement

of interstitials. Finally, we show coherent spin manipulation in this system, enabling a new tool to study the properties of this system and its environment. Beyond providing critical new clues to help identify these high-performance single photon emitters, our findings are promising as the basis for magnetic sensing technologies using defect fluorescence based on a mature optoelectronic semiconductor platform.

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AUTHOR CONTRIBUTIONS

All authors conceived the experiment. J. L. and G. F. developed the experimental approach. J. L. and Y. G. prepared samples. J. L. made measurements. J. L. and G. F. analyzed the experiment. J. L. and G. F. wrote the manuscript text. All authors reviewed the manuscript.

COMPETING INTERESTS STATEMENT

The authors declare no competing interests.

FIG. 1. **Optical properties of GaN defects.** (a) Photoluminescence image of an isolated defect (#2) and its surroundings. The scale bar is $2\,\mu$m. (b) Optical spectrum of defect #2. Inset: scanning electron microscope image of a solid-immersion lens carved around the defect. The scale bar is $4\,\mu$m. (c) Second-order photon auto-correlation $g^{(2)}(\tau)$ of defect #2. The zero-delay autocorrelation $g^{(2)}(0) = 0.3 < 0.5$, which is consistent with a single photon emitter. (d) Magnetic field dependent PL measured with the magnetic field roughly aligned to the c-axis of the GaN crystal showing two groups of behavior, as discussed in the text. (e) Minimal level diagram that is consistent with a $S \geq 1$ ground/excited-state spin. The non-radiative intersystem crossing (ISC) rate $\gamma_{\mathrm{ISC}}(m_s)$ into a meta-stable state (M) is spin-dependent. (f) Minimal level diagram that is consistent with a $S \geq 1$ metastable state. The non-radiative intersystem crossing rate $\gamma_{\mathrm{ISC,\,g}}(m_s)$ from a metastable state $|\mathrm{M}, m_s\rangle$ to the ground state $|\mathrm{g}\rangle$ depends on the metastable state spin m_s whereas the radiative relaxation rate γ_{eg} is spin-independent.

FIG. 2. **Optically detected magnetic resonance.** (a) and (b) show ODMR signals of defects #1 and #2 at $B = 1\,\mathrm{kG}$ respectively. (c) and (d) show the dependence of the ODMR peak contrasts of defects #1 and #2 on the alignment between the magnetic field and the crystal c-axis ($\theta = 0$). The dashed lines are guides for the eye. The error bars in (c) and (d) represent the standard error from least-squared fitting. No error bar is given when no above-noise-floor ODMR signal can be seen. (e) and (f) visualize the spin quantization axes with respect to the lattice. The white and black spheres represent Ga and N atoms, respectively. The solid blue arrow represents defect #1 and the dashed red arrow represents defect #2. The green circles highlight a hypothetical starting point of the spin quantization axes to help visualize the relationship between the lattice and the spins.

FIG. 3. **cw-ODMR spectrum as a function of magnetic field.** The magnetic-field-dependent ODMR signals for defects (a) #1 and (b) #2. Two spin resonances can be seen on the group-I defect #1 and four can be seen on the group-II defect #2. Note that the three faint lines dispersing with $g = 1$ indicated by the arrows are harmonic replica artifacts due to the microwave power amplifier nonlinearity.

FIG. 4. **Spin-dependent optical dynamics** (a) Timing diagram of a single measurement cycle of the pulsed ODMR and time-resolved PL schemes. The microwave pulse (blue) is applied before turning on the laser (green). After the optical readout, the laser is turned off to relax all population out of the metastable state before the microwave pulse turns back on in the next cycle. The optical detection is via either integrated counting during the signal and normalization windows (red and orange, respectively), or time-resolved single photon counting. (b) Pulsed ODMR measurement of defect #1. (c) Time-resolved PL of defect #1, with and without a microwave pulse applied prior to the laser pulse. Note the two data sets sit right on top of each other. (d) Pulsed ODMR measurement of defect # 2. (e) Time-resolved PL of defect #2, with and without a microwave pulse applied prior to the laser pulse. The solid and dashed arrows in (a, c, e) represent the times when the laser turns on and off, respectively.

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FIG. 5. **Rabi oscillation of a group-II defect.** (a) Timing diagram of a single measurement cycle of Rabi oscillation. A long green laser pulse (30 μs) initializes the system into a spin-polarized state. A short readout window (5 μs) reads the PL intensity of this spin state. Then a microwave tuned to an individual spin resonance is turned on for a varying pulse duration τ_{mw}. Lastly, a short readout laser pulse is turned on to read the final state PL intensity. The signal is obtained by normalizing the integrated PL intensity during the signal readout window to that during the norm readout window. (b), (c), and (d) show the Rabi oscillations when the microwave is tuned to the lower-, middle-, and higher-frequency resonances respectively.

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METHODS

Sample preparation. We study a GaN sample commercially available from the Xiamen Powerway Advanced Material Co., Limited, China. A 4 μm-thick layer of GaN is grown on a 430 μm-thick sapphire wafer by hydride vapor phase epitaxy (HVPE). The GaN is Fe-doped to make it semi-insulating. We pre-select GaN defects using our home-built scanning laser confocal microscope. We check that the PL spectra of the defects are consistent with the ones previously reported[1–3] and verify that they are single photon emitters by measuring the photon auto-correlation $g^{(2)}$. GaN is a high-index material with $n \sim 2.4$, which leads to a low fraction of PL leaving the material. To enhance photon collection, we use focused-ion-beam milling to carve out a 4 μm-diameter hemisphere-shaped solid-immersion lens (SIL) on the pre-selected defects. We conduct all measurements at room temperature.

Magneto-PL. We use a 50.4 mm-diameter 50.4 mm-long cylindrical neodymium iron boron permanent magnet to apply magnetic fields to the sample. To adjust the magnetic field amplitude and direction, we move the magnet on a motorized translation stage, having calibrated the magnetic field against magnet position. The details of the magnet setup are described in Supplementary Figures 8 and 9, and Supplementary Note 9.

Continuous-wave ODMR (cw-ODMR). To drive spin resonance, a copper microwire is lithographically patterned near the SILs containing the defects of interest. The details of the microwave set-up are described in Supplementary Figure 8(b) and Supplementary Note 9. We drive about 20 dBm of microwave power to induce the spin resonances and excite the defects with an optical power of 15–20 μW.

Pulsed measurements. Figure 4(a) shows the pulse scheme in a measurement cycle for pulsed ODMR and time-resolved PL measurements. The details of the timing can be found in Supplementary Figure 5. In both schemes, we apply microwaves before we excite the defects for optical readouts, and we turn off the laser for a sufficient time before we apply microwaves again in the next cycle to allow relaxation from all populations to the ground state.

Supplementary Figure 5(a) shows the timings of a cycle of pulsed ODMR measurement. After the optical pulse has been off for 3 μs, the microwave pulse in the next cycle is turned on for 2 μs and off 65 ns before the laser excitation. We read the PL for 2 μs after the microwave turns off and normalize it to the PL registered after the laser has repolarized

the system for 8 μs. This sequence is designed to distinguish between a ground-state and a meta-stable state spin.

To measure time-resolved PL, we apply a microwave pulse tuned to the largest contrast resonance frequency for 1 μs before the laser turns on as depicted in Supplementary Figure 5(b), and we allow the system to relax for 1.5 μs before applying a microwave pulse again in the next cycle. The optical detection is done by a time-correlated single-photon-counting (TCSPC) module that is triggered by a synchronization pulse when the laser turns on at $t_{L,on}$ in each pulse cycle. This way, we record the photon arrival times relative to the laser excitation time. The histogram of photon arrival times gives the time-resolved PL.

Supplementary Figure 5(c) shows the timing of a cycle of the Rabi oscillation measurement. A long-duration laser pulse (40 μs) initializes the system, a microwave, with duration τ_{mw}, tuned to a spin resonance frequency rotates the spin, and a short readout laser pulse (5 μs) is applied after the spin manipulation. We count the PL intensity for the same readout duration (5 μs) at the end of the optical initialization pulse before the microwave turns on to normalize the signal readout.

DATA AVAILABILITY

All data are available on eCommons: Digital Repository at Cornell (URL available soon).

CODE AVAILABILITY

All analysis codes are available on eCommons: Digital Repository at Cornell (URL available soon).

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