Tunable multiphoton Rabi oscillations in an electronic spin system
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I. INTRODUCTION

Harmonic systems are a basic manifestation of quantum mechanics and appear in various forms, down to the nanoscale, as electromagnetic or mechanical oscillators. In the case of a finite number of excited states, multilevel systems are proposed to perform quantum algorithms in size-limited or scalable schemes by using microwave (MW) pulses to generate entangled states. The coherent manipulation of spins in a multilevel system is fundamental to the implementation of the Grover algorithm. At the same time, the quasiharmonic nature of the system can lead to interesting effects, where the system behavior is tunable between a multi-level and a two-level type of system.

Spin systems benefit from relatively large coherence and relaxation times, which makes them suitable as quantum-bit implementations or as a different type of quantum random access memory. In some studies, the spin qubit operation is demonstrated at temperatures up to ambient. Recent studies demonstrated that in diluted systems, spin-spin dipolar interactions are sufficiently low to allow coherent, quantum manipulations. Such situations are well exemplified by systems such as the nitrogen-vacancy color centers in diamonds, N atoms in C60, Er3+, Cr5+ (Ref. 16) ions and molecular magnets.

In a previous work we demonstrated multiphoton spin coherent manipulation in a multilevel system (Mn2+) diluted in MgO, a highly symmetric nonmagnetic matrix. In this work we report on the possibility of using a multilevel system to tune the Rabi oscillations using a combination of two parameters: microwave power and/or magnetic-field orientation. To this end, one needs a magnetic system with a well-defined anisotropy, but sufficiently small in size to be overcome by microwave amplitudes achievable in typical experimental conditions.

The 5/2 Mn2+ spin is our system of choice. As detailed in Sec. II, the crystalline anisotropy for this high cubic symmetry is orders of magnitude smaller than the magnetic (Zeeman) energy and therefore the multilevel system is quasiharmonic. This is essential for successful multiphoton spin manipulation and for state tunability by magnetic-field orientation. At the same time, the anisotropy remains smaller than or comparable to the microwave drive, an aspect that is essential for tunability by drive.

In this $S = 5/2$ system we demonstrate multiphoton Rabi oscillations and controlled tuning of the system in and out of a quasiharmonic level configuration. Using the rotating-frame approximation, the experiments are explained very well by both an analytical model and a generalized numerical model. The calculated multiphoton Rabi frequencies are in excellent agreement with the experimental data.

The article is structured as follows. In Sec. II we describe the spin Hamiltonian and its parameters. In Sec. III the experimental procedure and setup are detailed. In Sec. IV an analytical and a numerical model are given, describing the tunability of the multiphoton dynamics. The experimental findings are described in Sec. V.

II. QUASIHARMONIC SPIN HAMILTONIAN

The spins $S = 5/2$ of the Mn2+ ions are diluted in a MgO nonmagnetic matrix of cubic symmetry $Fm\overline{3}m$ (lattice constant 4.216 Å). The Mn2+ ions are located in substitutional positions of Mg2+ ions. The high degree of symmetry ensures that the spins experience an almost isotropic crystalline environment and thus the fourth-order magnetic anisotropy can be made much smaller than the Zeeman splittings. Interactions between Mn2+ ions and symmetry deformations are neglected. The spin Hamiltonian at resonance is given by

$$H = \frac{a}{\hbar} 2 \sum_{i} S_{i}^{+} S_{i}^{-} - S(S+1)(3S^{2}-1)/5$$

$$+ \gamma \vec{H}_{MW} \cdot \vec{S} \cos(2\pi ft),$$

where $\gamma = g \mu_{B}/\hbar$ is the gyromagnetic ratio, $g = 2.0014$ is the g factor, $\mu_{B}$ is Bohr’s magneton, and $h$ is Planck’s constant. $S_{x,y,z}$ are the spin projection operators, $\vec{S}$ is the total spin, $a = 55.7$ MHz is the anisotropy constant, $A = 244$ MHz is the hyperfine constant of $^{55}$Mn ($I = 5/2$), $h_{MW}$ and $f$ represent the microwave amplitude and frequency, respectively.
crystal field for a Zeeman splitting $\gamma H_0 = E_{1/2} - E_{-1/2}$. Arrows indicate one-photon (black), three-photon (red), and five-photon (blue) monochromatic transitions between spin projections $m$, as shown on the left-hand side. The dashed and dotted lines show equidistant virtual levels enhancing the multiphoton transitions.

The sample is a $3 \times 3 \times 1$-mm$^3$ single crystal of MgO doped by a small amount of Mn$^{2+}$. The static field ensures a Zeeman splitting of $H_0 \approx f = 9$ GHz, which is much stronger than all other interactions of Eq. (1). This implies that (i) $H_0$’s direction can be approximated as the quantization axis and (ii) coherent MW driving is confined between levels of the same nuclear spin projection $m_1$ (see also Refs. 5, 21, and 22).

The six-level system is consequently quasiharmonic, as shown by the level diagram in Fig. 1. The dashed and dotted lines show virtual levels of equidistant separation $g \mu_B H_0 = hf$, which strongly enhance the multiphoton transition probabilities. Since $a \ll f$, the distance between the real and virtual levels in Fig. 1 is exaggerated for clarity. The number of arrows for each monochromatic transition suggests the type of multiphoton coherent excitation (one, three, or five photons). A coherent Rabi superposition of the $m = -5/2, \ldots, 5/2$ states (as counted on the left-hand side of the figure) can thus be obtained, which is strongly dependent on the microwave drive power and nutation time.

III. EXPERIMENTAL PROCEDURE

Rabi oscillation measurements have been performed in a Bruker Elexsys 680-pulse electron paramagnetic resonance (EPR) spectrometer working at about $f = 9.6$ GHz ($X$ band). The sample is a $3 \times 3 \times 1$-mm$^3$ single crystal of MgO doped by a small amount of Mn$^{2+}$ (a few parts per 10$^6$). The orientation between the sample and the static field is controlled by a goniometer (with a precision to 1') with the rotation axis parallel to the MW magnetic-field direction $\vec{h}_{MW}$.

In the measurements presented here, $\vec{h}_{MW} || [-110]$ (see Fig. 2 inset). All measurements have been made at room temperature. Calibration of the microwave field $h_{MW}$ has been made using the 1,3-bisphenylene-2-phenylallyl (BDPA) standard:

$$\text{BDPA:} \quad |0\rangle \rightarrow |1\rangle \rightarrow |2\rangle \rightarrow |3\rangle \rightarrow |4\rangle \rightarrow |5\rangle \rightarrow |6\rangle \rightarrow |7\rangle \rightarrow |8\rangle \rightarrow |9\rangle$$

FIG. 1. (Color online) Level diagram of the Mn$^{2+}$ spin in a cubic crystal field for a Zeeman splitting $\gamma H_0 = E_{1/2} - E_{-1/2}$. The dashed and dotted lines show virtual levels of equidistant separation $g \mu_B H_0 = hf$, which strongly enhance the multiphoton transition probabilities. Since $a \ll f$, the distance between the real and virtual levels in Fig. 1 is exaggerated for clarity. The number of arrows for each monochromatic transition suggests the type of multiphoton coherent excitation (one, three, or five photons). A coherent Rabi superposition of the $m = -5/2, \ldots, 5/2$ states (as counted on the left-hand side of the figure) can thus be obtained, which is strongly dependent on the microwave drive power and nutation time.

A small amount of BDPA containing isotropic $S = 1/2$ spins gives a Rabi frequency of exactly $F_R = \mu_B h_{MW} / \hbar$.

By design, the detection is sensitive to frequency $f$ and therefore it can probe only transitions between consecutive spin projections, $m \leftrightarrow m+1$. As the transition $| -1/2 \rangle \leftrightarrow |1/2 \rangle$ is of highest probability, as ensured by Fermi’s golden rule, and is not sensitive to crystal strain effects we choose to use the levels $m = \pm 1/2$ as a probe of the six-level dynamics.

By definition, the magnetization is given by $\langle S_i \rangle \equiv \text{Tr}(\rho S_i)$, where $\rho$ is the density matrix and $S_i$ is the $\hat{S} = 5/2$ spin operator $i = x, y, z$. Since we probe only the transition $| -1/2 \rangle \leftrightarrow |1/2 \rangle$ we define the magnetization of the subset $\langle S_i \rangle_{1/2} \equiv \text{Tr}(\rho_{1/2} S_i)$, where $\rho_{1/2}$ is the central block matrix of $\rho$ corresponding to the base subset $| \pm 1/2 \rangle$.

The drive and detection of the multiphoton Rabi oscillations are implemented with the pulse sequence presented in Fig. 2. A drive pulse $P_R$ of duration $\tau_R$ and resonant to the $| -1/2 \rangle \leftrightarrow |1/2 \rangle$ transition is applied at $t = 0$. At the end of this pulse, the density of states has been coherently changed.

Right after the $P_R$ pulse, spin dephasing and line inhomogeneity induce a free induction decay (FID) signal. The first point of this FID ($t = \tau_R$) gives the transverse magnetization traced on the $| \pm 1/2 \rangle$ subset: $\langle S_i \rangle_{1/2} = \rho_{-1/2,1/2} + \rho_{1/2,-1/2}$. Unfortunately, the dead time $\tau_{DT}$ of the spectrometer ($\sim 80$ ns) prevents the measurement of this first point. If the entire inhomogeneous line $| -1/2 \rangle \leftrightarrow |1/2 \rangle$ of width $\Delta H_0$ is excited by a sufficiently short $P_R$ pulse (nonselective pulse, here $\tau_R^{-1} > \gamma \Delta H_0 / 2 \pi = 0.27$ MHz), the FID is simply the Fourier transform of the absorption line and an exponential decay in MgO:Mn$^{2+}$. Therefore, we integrate the detected FID and the transform of the absorption line and an exponential decay in MgO:Mn$^{2+}$.

FIG. 2. (Color online) Pulse sequence used in the multiphoton Rabi oscillation measurements, which starts with a strong excitation pulse $P_R$ inducing the multiphoton nutation. (a) Right after the pulse $P_R$, the free-induction-decay signal gives the transverse magnetization state between $-1/2$ and $1/2$. (b) After a time $\tau_R \gg T_2$, a $\pi/2$ pulse rotates the longitudinal magnetization so it can be measured by our setup (see the text for more details). The inset shows the static- and electromagnetic-field orientations with respect to the crystalline axes.
shown in Fig. 2(b). After the Rabi pulse $P_R$, one waits a time $\tau_p - \tau_R$, which is smaller than the system’s relaxation time $T_1$ but larger than the decoherence time $T_2$. Thus, at $t = \tau_p$, only the longitudinal magnetization is nonzero.26

A $\pi/2$ pulse follows, rotating the $|\pm 1/2\rangle$ population in a coherent mixture, which is located in the $xy$ plane (or the equatorial plane). The detection observes the FID toward zero in this mixture, giving essential information such as the initial total magnetization in the $xy$ plane, its FID decoherence time $T_2$, and a potential shift away from the resonance static field.

As indicated with arrows in Fig. 1, in a cubic symmetry the three- and five-photon transitions use the $|\pm 1/2\rangle$ states as intermediate ones. Therefore, the $\pi/2$ pulse can distinguish between the various multiphoton Rabi oscillations due to their different frequencies, as detailed in the following section.

IV. RABI ROTATIONS IN A TUNABLE MULTILEVEL SYSTEM: THEORETICAL TREATMENT

The aim of this section is to present a model that describes the coherent multiphoton Rabi oscillation in a quasiharmonic large spin system under monochromatic radiation. To compute the multiphoton Rabi frequencies, we used the density-matrix theory applied to the Mn$^{2+}$ ion ($S = 5/2, I = 5/2$) in the rotating-frame approximation.

A. Analytical calculation

Let us consider a quantum system with six states $|S_i\rangle$, $S_i = \{-5/2, -3/2, -1/2, 1/2, 3/2, 5/2\}$, irradiated by an electromagnetic field in resonance with the $-1/2$ and $1/2$ levels. We assume initially $I = 0$, but we will describe the effect of the nuclear spin at the end of this section. The Hamiltonian of the system is

$$\hat{H} = \hat{E} + \hat{V}(t) = \sum_{S_i=-5/2}^{5/2} E_{S_i}|S_i\rangle\langle S_i| + \hat{V}(t),$$

(2)

with $E_{S_i}$ the static energy levels, $\hat{V}(t) = \frac{\gamma}{2} h_{\text{MW}} (\hat{S}_z + \hat{S}_- \cos(2\pi f t))$, $S_+$ ($S_-$) the raising (lowering) operators, and $f = E_{1/2} - E_{-1/2}$.

In a cubic symmetry and first order in perturbation ($a \ll \gamma H_0$), the static energy levels are given by

$$E_{\pm 5/2} = (\pm 5/2)\gamma H_0 + (1/2)pa + O(a^2),$$

$$E_{\pm 3/2} = (\pm 3/2)\gamma H_0 + (3/2)pa + O(a^2),$$

$$E_{\pm 1/2} = (\pm 1/2)\gamma H_0 + pa + O(a^2),$$

(3)

where $p = 1 - 5(A^2B^2 + B^2C^2 + C^2A^2)/3$, with $A$, $B$, and $C$ representing the cosine directors of $\vec{H}_0$ with the crystalline axes. For our geometry, $p$ is given by

$$p = 1 - 5 \sin^2 \theta + \frac{15}{4} \sin^4 \theta,$$

(4)

with $\theta$ the angle between $\vec{H}_0$ and the $c$ axis [001]. Since $H_0 \gg h_{\text{MW}}$, we can use the rotating-wave approximation (RWA) to make Eq. (2) time independent. Since in a conventional pulsed spectrometer we have access to only the first harmonic detection $hf$, we define the unitary transformation $\hat{U}(t) = \exp(-i2\pi f \hat{S}_z t)$ and apply it to Eq. (2). In the rotating frame, the Hamiltonian in Eq. (2) becomes

$$\hat{H}_{\text{RWA}} = U\hat{H}U^\dagger + i\hbar \frac{\partial U}{\partial t} U^\dagger$$

$$= \begin{pmatrix}
\hbar^2 p a & \frac{\sqrt{3}}{2} V & 0 & 0 & 0 & 0 \\
\frac{\sqrt{3}}{2} V & -\frac{1}{2} p a & \sqrt{2} V & 0 & 0 & 0 \\
0 & \sqrt{2} V & pa & 3\frac{1}{2} V & 0 & 0 \\
0 & 0 & 3\frac{1}{2} V & pa & \sqrt{3} V & 0 \\
0 & 0 & 0 & -\frac{1}{2} p a & \frac{\sqrt{3}}{2} V & 0 \\
0 & 0 & 0 & 0 & \frac{1}{2} p a & 2\hbar^2 V \\
\end{pmatrix},$$

(5)

where $V = \gamma h_{\text{MW}}/2$. By diagonalization, the eigenenergies $E_{\alpha}/pa$ of the dressed states $|\Psi_{\alpha}\rangle$ are calculated as a function of $\hbar \equiv V / pa$, as illustrated in Fig. 3 for $p > 0$.

For high anharmonicity or low microwave power $\hbar \rightarrow 0$, in which case $|\Psi_1\rangle = (|1/2\rangle + |-1/2\rangle)/\sqrt{2}$ and $|\Psi_2\rangle = (|1/2\rangle - |-1/2\rangle)/\sqrt{2}$ are the coherent superposition of states $|\pm 1/2\rangle$. The other wave functions remain unchanged: $|\Psi_3\rangle = |5/2\rangle$, $|\Psi_4\rangle = |-5/2\rangle$, $|\Psi_5\rangle = |3/2\rangle$, and $|\Psi_6\rangle = |-3/2\rangle$. The splitting between $|\Psi_1\rangle$ and $|\Psi_2\rangle$ is detected as an oscillation of frequency $F_R = E_2 - E_1 = 3\frac{1}{2} \gamma h_{\text{MW}}$, in agreement with the general formula

$$F_R^{(1)} = \frac{\gamma}{2} h_{\text{MW}} \sqrt{S(S+1) - S_z(S_z + 1)}$$

(6)

for one-photon Rabi oscillations between consecutive states $S_z$ and $S_z + 1$.

When the microwave power $h_{\text{MW}}$ increases, the degeneracy of $|\Psi_1\rangle$ ($n = 3, 4, 5, 6$) is lifted, allowing coherent three- and five-photon transitions ($-3/2 \leftrightarrow 3/2$ and $-5/2 \leftrightarrow 5/2$, respectively). Note that two- and four-photon processes are out of resonance and therefore are not discussed here. In first-order perturbation in $\hbar$, only the one-photon process exists between states $|\Psi_1\rangle$ and $|\Psi_2\rangle$. In third-order perturbation, a three-photon process mixes the states $|\pm 3/2\rangle$, leading to a pure three-photon Rabi oscillation with frequency $F_R^{(3)} = \frac{21}{2} \hbar^2 pa$, as detailed in the Appendix. This three-photon...
process perturbs the aforementioned one-photon process, which now has the Rabi frequency \( F_R^{(1)} = F_R^{(1)} = \frac{\gamma H_0}{h} \).

In fifth-order perturbation, the five-photon process mixes the states \([\pm 5/2]/5/2\), leading to a Rabi frequency \( F_R^{(5)} = \frac{15 \gamma^2 H_0^2}{2^{11/2} h} \) (a pure five-photon process). Similarly, the five-photon process will change the dynamics of the three- and one-photon Rabi oscillations [see the Appendix, Eqs. (A3)–(A5)].

For \( h \rightarrow \infty \), the microwave field excites all transitions and the six levels become equidistant [see Eq. (A7)] while the dynamics is coherently driven by a five-photon process. This dynamics resembles that of a spin \( S = 1/2 \). The diagonal of the Hamiltonian in Eq. (5) becomes negligible and the diagonalization simply rotates the Hamiltonian along the \( h_{MW} \) field.

When the nuclear spin is included, the model becomes more complicated, but analytical solutions can still be found. The full spin Hamiltonian of \( ^{55}\)Mn\(^{2+}\) \((S = 5/2, I = 5/2)\) is a 36 \( \times \) 36 matrix. If we assume no forbidden nuclear transitions \( (\Delta m_I = 0) \), with \( m_I \) quantifying the \( I \) projections), the Hamiltonian in Eq. (1) can be separated into six 6 \( \times \) 6 matrices of the form in Eq. (2).

The static energies \( E_S \) now depend also on \( m_I \) and \( A \) and have been computed in first-order perturbation in \( A/H_0 \) by Low.\(^{19}\) Having the new \( E_S \), renormalized by the hyperfine field, we can use the same procedure as above to compute the eigenvalues of the dressed states and the Rabi splittings.

Finally, note that all other transitions can be computed by the method presented here by changing the value of \( f \) in Eq. (2). For instance, for \( f = (E_{-3/2} - E_{1/2})/2 \) one can study the two-photon Rabi oscillations in the transition \([\pm 3/2] \leftrightarrow [1/2])\).

### B. Numerical calculation

The analytical model presented above describes the multiphoton frequencies of the Rabi oscillations very well but fails to describe their intensities (the time evolution of spin populations). In order to describe the experimental results we developed a model using the Hamiltonian in Eq. (1), which shows the actual crystal-field parameters and includes the hyperfine coupling between the \( S = 5/2 \) electronic spin and the \( I = 5/2 \) nuclear spin of \( ^{55}\)Mn\(^{2+}\).

The anisotropy \( a \) and hyperfine \( A \) constants are two orders of magnitude smaller than the electron Zeeman interaction. Therefore, it is appropriate to assume that the orientation of the static field imposes the quantization axis \( \hat{z} \). On this basis, the Hamiltonian in Eq. (1) is rewritten as in Eq. (2), where \( E_S \) is obtained by exact diagonalization and reflects the hyperfine coupling.

Once we get the form of Eq. (2) for \( \mathcal{H} \), the transformation into the rotating frame become obvious. Experimentally, the spectrometer signal represents the transient magnetic resonant signal in the rotating frame, obtained by mixing the signal reflected by the cavity \([S(t) \cos(2\pi f t)]\) and the reference arm \([\cos(2\pi f t)]\). This leads to a frequency-independent signal \( S(t) \) and a double frequency signal (which is filtered out). The unitary transformation is fixed by the microwave frequency \( f \), with \( U(t) = \exp(-i2\pi f \hat{S}_z t) \), and using Eq. (5) one gets

\[
\mathcal{H}_{RWA} = \frac{\gamma^2}{2} h_{MW} \hat{S}_z + \hat{E} - f \hat{S}_z.
\]

For a fixed \( m_I \), the static field \( H_0 \) is chosen to satisfy the condition \( E_{1/2} - E_{-1/2} + \Delta = f \), where \( \Delta \) represents the detuning of the static field, away from resonance. As an example, the eigenvalues of the Hamiltonian in Eq. (7), for \( H_0 = 0 \) and nuclear projection \( m_I = 1/2 \), are shown in Fig. 4 as a function of field detuning \( \Delta \). In these simulations, \( h_{MW} = 0.8 \) mT, which is sufficiently strong to induce Rabi splitting with three and five photons, as shown in insets.

The dynamics of the Hamiltonian in Eq. (7) is described by the time evolution of the 6 \( \times \) 6 density matrix \( \rho(t) \):

\[
\frac{d\rho}{dt} = [\mathcal{H}_{RWA}, \rho].
\]

In the rotating frame (Dirac or interaction picture), the solution is \( \rho(t) = U_\rho(t) \rho_0 U_\rho^\dagger(t) \), with \( \rho_0 \) the matrix density at thermal equilibrium,

\[
\rho_0 = \frac{\exp(-\hat{E}/kT)}{\text{Tr}[\exp(-\hat{E}/kT)]},
\]

and \( U_\rho(t) \) is the propagator operator \( U_\rho(t) = \exp(-i2\pi \mathcal{H}_{RWA} t) \).

When the Rabi pulse \( P_R \) is applied, the spin population is coherently manipulated. At the end of the sequence a weak \( \pi/2 \) pulse will selectively probe the difference of population between states \(-1/2 \) and \( 1/2 \). The signal output of the spectrometer is \( S(t) = \sigma_{-1/2}(t) - \sigma_{1/2}(t) \). Note that for a spin \( S = 1/2 \), \( S(t) \equiv \langle \hat{S}_z \rangle(t) \).

With the method presented here, the time evolution of \( S(t) \) can be computed for a fixed value of the static \( H_0 \) and microwave \( h_{MW} \) fields. To extract the Rabi frequencies, we perform a fast Fourier transform (FFT) on the computed \( S(t) \). Since the model does not take into account the decoherence or decay effects of the Rabi oscillations, we multiply the simulated \( S(t) \) by a phenomenological exponential decay, with a characteristic time (or FFT width) similar to that of the experimentally observed oscillations.

### V. Experimental Results

A typical MW absorption spectrum of MgO:Mn\(^{2+}\) as a function of \( H_0 \) shows six sets (one for each \( m_I \) projection)
A. Tunable multiphoton Rabi oscillations by changing the orientation of the static field

The static-field orientation is described by the $p$ parameter [see Eq. (4)] and therefore can tune the reduced microwave field $\tilde{h} = V/pa$. We measured Rabi oscillations and corresponding FFTs at constant microwave field ($h_{MW}=1.48$ mT) for three different orientations ($p$ values) of $H_0$, as shown in Fig. 5. One could see a highly anisotropic behavior of the coherent dynamics depending on the relative orientation between $H_0$ and the crystal axes.

For $H_0||[001]$ ($p = 1$, maximum value), only one frequency is observed, but its value (57 MHz) is smaller than the one expected for the one-photon Rabi frequency in the transition $1/2 \leftrightarrow 1/2$ in $S = 5/2$ [62.5 MHz; see Eq. (6)] and the red dashed line in Fig. 5. Although the multiphoton Rabi oscillation is not clearly resolved, it is indirectly slowing down the expected one-photon dynamics, as described in Sec. IV A by the difference $F_R^{(1)} - F_R^{(3)}$.

For $H_0||[110]$ ($p = -1/4$), in addition to the high-frequency one-photon process, a low-frequency three-photon Rabi oscillation becomes observable due to a lower-$p$ value and thus a larger $h$. Finally, for $\theta = \theta_{CA}$ [the compensation angle of the cubic anisotropy: $\rho(\theta_{CA}) = 0$] and hence $\tilde{h} \rightarrow \infty$, only one Rabi frequency is observed (Fig. 5, top). Its value is the same as the one expected for an isotropic spin (dash-dotted line in Fig. 5). As explained in Sec. IV A and the Appendix, a coherently driven five-photon process (and only this process) is induced, leading to a spin dynamics similar to that of a spin 1/2.

A detailed $\theta$ dependence of detected Rabi frequencies, defined as FFT peaks of coherent oscillations (as introduced in

B. Tunable multiphoton Rabi oscillations by changing the MW field

Aside from the tuning method based on static-field orientation, described in the preceding section, an alternative method is based on the MW drive intensity. To demonstrate the method, we apply a static field not far from the compensation angle $\theta_{CA}$ described above such that the spin system is slightly anisotropic. We choose an angle $\theta = 31 \pm 1^\circ$ and record coherent Rabi oscillations as a function of the microwave field $h_{MW}$. The FFT peaks (Rabi frequencies) are shown in Fig. 7 by points and the error bars represent the FFT linewidths. The dashed blue line is the MW field dependence of Rabi frequency for a pure one-photon transition between levels $|-1/2\rangle \leftrightarrow |1/2\rangle$ of the $S = 5/2$ system [$F_R = 3\gamma h_{MW}$; see Eq. (6)]. The dotted black line is the MW field dependence of Rabi frequency in the case of a pure isotropic spin [$F_R = \frac{\gamma}{2} h_{MW}$; see Eq. (A7)].

Indeed, at low MW power, the system has the dynamics of an anisotropic $S = 5/2$ system, as shown by the very
A quantitative numerical description is quite difficult because of the extreme nonlinearity of $\hbar$ near $\theta_{CA}$. At the compensation angle ($\theta_{CA} = 29.67^\circ$), $\hbar \rightarrow \infty$, but a small change in $\theta$ induces significant changes in $\hbar$ (e.g., at $\theta = 30^\circ$, $\hbar = 16.3 \hbar_{MW}$; at $\theta = 31^\circ$, $\hbar = 4.1 \hbar_{MW}$; and at $\theta = 32^\circ$, $\hbar = 2.4 \hbar_{MW}$). As the accuracy of our goniometer is $1^\circ$, a numerical quantitative description around $\theta_{CA}$ is not relevant. Qualitatively, one notes that for $\hbar_{MW} > 0.5$ mT, the one-photon Rabi frequency deviates from $F_R^{(1)}$ of Eq. (6) due to the formation of multiphoton processes. For high microwave powers ($\hbar_{MW} > 1$ mT), the one-photon FFT peak is barely visible (shown by open up-pointing triangles) while the five-photon process entirely dominates the dynamics. This is indicated by the good agreement between the five-photon FFT peaks (solid squares) and the dotted line resulting from an isotropic $S = 5/2$ model (or a spin 1/2), as explained by Eq. (A7).

Our experiment demonstrates that a sufficiently intense microwave field can tune the spin dynamics from that of an anisotropic (unequidistant) system to that of an equidistant, quasiharmonic five-level system (equivalent to a two-level system).

C. Five-photon Rabi oscillations at the compensation angle

When the static field is applied along a direction for which $\theta = \theta_{CA}$ (that is, $\theta = 29.67^\circ$), the cubic anisotropy is geometrically compensated for and therefore $p = 0$ and $\hbar \rightarrow \infty$. As explained in Sec. IV A and the Appendix, one expects a perfectly isotropic $S = 5/2$ spin system.

We performed room-temperature Rabi oscillations for this field orientation. The FFT of the coherent oscillations are single peaked, as shown in Fig. 8, that is, the size of the symbol). The color plot is calculated using the numerical procedure described in Sec. IV B and the dashed line indicates the analytical result of Eq. (A7). There is excellent agreement between the analytical and numerical models and the experimental data points (no fit parameter has been used).

During a Rabi rotation for this isotropic case, the spin density gradually moves upward on the equidistant six-level ladder and when it reaches the highest level ($S_z = 5/2$), the dynamics is reversed. In contrast to a one-photon dynamics, here we have a collective five-photon process, ensuring a continuous nutation within the $S_z = -5/2, \ldots, 5/2$ space.
VI. DISCUSSION

The experimental data and the model presented here use a monochromatic source to both induce and probe the multiphoton Rabi oscillations. Therefore, we restrict the types of multiphoton transitions that are measurable to those that exist at the same resonant static field as the one-photon transition (used to induce and probe). In order to probe directly all types of multiphoton transitions, two tunable frequencies will be needed: one to induce the multiphoton oscillations, i.e., to induce a two-photon Rabi oscillation \( \left( f_1 = (E_{1/2} - E_{-1/2})/2 \right) \), and one to probe it \( f_2 = E_{1/2} - E_{-1/2} \). Note that the experiments described here, with a single or two tunable frequencies, have the appeal of being applicable to on-chip studies, using detectors placed in strong magnetic fields.29-31

VII. CONCLUSION

We report on single-photon and multiphoton coherent rotations in a \( S = 1/2 \) spin system featuring a cubic anisotropy. The anisotropy is much smaller than the Zeeman splittings, i.e., the six-level scheme shows only a small deviation from an equidistant diagram. This allows us to tune the spin dynamics by compensating for the cubic anisotropy with either a precise static-field orientation or a microwave-field intensity. In both cases, we can transition the system between an anisotropic to an isotropic situation, the latter case showing a spin dynamics governed by equidistant diagram. This allows us to tune the spin dynamics to the desired frequency: \( \hbar \gamma_{MW} \) with solutions \( E_{\pm} = |\hbar| \pm \frac{12\hbar^2}{25} \). We found the classical result that during the resonance \( |1/2\rangle \) and \( |-1/2\rangle \) are mixed by the electromagnetic wave and the other states stay unperturbed.

When \( \hbar \) is increased, we computed the solutions of Eq. (A1) up to the fifth-order perturbation in \( \hbar \). The Rabi frequencies \( F_R = (E_+ - E_-)/|p|\hbar \) are

\[
F_R^1 = p \left( 3\hbar - \frac{24\hbar^3}{25} - \frac{768\hbar^5}{125} \right),
\]

\[
F_R^3 = p \left( \frac{24\hbar^3}{25} - \frac{339\hbar^5}{250} \right),
\]

\[
F_R^5 = p \left( \frac{15\hbar^5}{2} \right).
\]

These equations are used to simulate the solid lines in Fig. 9, which are compared to the numerical solutions of Eq. (A1) (discrete data points). The inset shows a close-up view at low microwave powers and Rabi frequencies. One could conclude that for powers up to \( \hbar \sim 1/2 \), the perturbative analysis works very well.

When \( \hbar \to \infty \), Eq. (A1) becomes

\[
P_{\pm} \approx \pm \frac{15\hbar^3}{2} - 26\hbar^2\hbar \mp 12\hbar^2 \pm 8\hbar^3 = 0.
\]

with solutions \( E_{\pm} = \hbar \pm \frac{15\hbar^3}{2} \pm \frac{12\hbar^2}{2} \), which represent dressed-state energies of an isotropic spin 1/2 with Rabi frequency:

\[
F_{R/2}^{\text{iso}} = F_{1/2} = \frac{\gamma_{MW}}{2}.
\]
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