Quantum Information Processing with Large Nuclear Spins in GaAs Semiconductors

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We propose an implementation for quantum information processing based on coherent manipulations of nuclear spins $I = 3/2$ in GaAs semiconductors. We describe theoretically an NMR method which involves multiphoton transitions and which exploits the nonequidistance of nuclear spin levels due to quadrupolar splittings. Starting from known spin anisotropies we derive effective Hamiltonians in a generalized rotating frame, valid for arbitrary I, which allow us to describe the nonperturbative time evolution of spin states generated by magnetic rf fields. We identify an experimentally observable regime for multiphoton Rabi oscillations. In the nonlinear regime, we find Berry phase interference.

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Recent advances in spintronics [1] have shown that the coherent control of electron and nuclear spins in semiconductors is experimentally feasible, enabling, in particular, an all-optical NMR in GaAs, based on the hyperfine interaction between electrons and nuclei [2,3]. Such a control of nuclear spins can also be achieved via electrical gates as recently demonstrated for GaAs heterostructures in the quantum Hall regime [4], or even via conventional NMR directly accessing the nuclei [5]. In the present work, we show that such advances in coherent spin control have opened up the possibility to manipulate the nuclear spins I for the purpose of quantum information processing, thereby presenting a scheme that is based on ensembles of large spins $I > 1/2$ instead of qubits. Nuclear spins are ideal candidates for this purpose due to their long decoherence times.

An implementation of the Grover algorithm [6] has recently been proposed for molecular magnets [7], based on a perturbative approach to the unitary Grover operations which encode and decode the information stored in the phases of *small* amplitudes a_m [8]. An alternative version of Grover's algorithm was presented in Refs. [9,10] that is described by a Hamiltonian that lets a completely delocalized state of the form $|\psi\rangle = \sum_{m=-I}^{I} a_m|m\rangle$, in some basis states $|m\rangle$ with equal occupation probabilities $|a_m|^2$, evolve into a wanted localized state $|M\rangle$, where $|\psi\rangle$ and $|M\rangle$ are degenerate and have a finite overlap. The information is encoded in the energies of $|m\rangle$. In order to produce $|\psi\rangle$, we propose here a novel NMR scheme that allows us to generate any desired distribution of amplitudes a_m, being not restricted to small values. For this we specifically exploit the properties of GaAs nuclei where quadrupolar spin splitting results in spin anisotropies and thus in nonequidistant energy levels—being a necessary condition for our scheme. The theoretical problem then is to find one magnetic rf pulse—inducing a unitary time evolution of the spins—that produces the desired spin state $|\psi\rangle$ and a second rf pulse that lets $|\psi\rangle$ evolve into $|M\rangle$, given certain spin anisotropies and adjustable magnetic fields (see below). In a nonperturbative approach, we find an analytic solution to this problem, valid for arbitrary spin I. For the special case of GaAs with $I = 3/2$, we have confirmed our analytical results by exact numerics. In contrast to previous work [7] our method also holds for vanishing detuning energies, which turns out to be essential to perform nonperturbative unitary operations, i.e., quantum computations (QCs). Once the control over $2I$ magnetic fields is established, the scheme proposed here allows for QC and quantum storage with a *single* pulse, provided that there is sufficient signal amplification due to the spin ensemble [11].

As a first step towards this goal, it is useful to generate and monitor multiphoton Rabi oscillations, as we describe in detail below. Finally, we show that oscillating quadratic transverse spin terms, which can be generated by optical pulses in GaAs [3,12], give rise to Berry phase oscillations [13] in the transition probabilities.

In the following we mainly focus on a nuclear spin of length $I = 3/2$, as appropriate for GaAs, but indicate its generalization to arbitray I. Our nuclear spin system is described by the Hamiltonian $\mathcal{H}_0 = \mathcal{H}_Z + \mathcal{H}_Q$, consisting of the nuclear Zeeman energy $\mathcal{H}_Z = -g_N\mu_N H_z I_z$, $g_N = 1.3$ [2], and the quadrupolar splitting [5] $\mathcal{H}_Q = A[3I_z^2 - I(I+1)]$. The quadrupolar constant is $A = 7 \times 10^{-7}$ K for ^{69}Ga, $A = 3 \times 10^{-7}$ K for ^{71}Ga, and $A = 2 \times 10^{-6}$ K for ^{75}As nuclei[3]. For the purpose of QC we need to achieve complete control over unitary state evolutions, i.e., control over amplitudes a_m to form a desired superposition $|\psi\rangle = \sum_{m=-I}^{I} a_m|m\rangle$ of the nuclear basis states $|m\rangle$ (eigenstates of $\mathcal{H}_0$). Our goal is now to show that such a control over a_m is indeed feasible under experimentally attainable conditions.

We start from a configuration where mainly the ground state $|3/2\rangle$ is populated; see Fig. 1. This can be achieved by the Overhauser effect [14]. The next goal is to coherently populate all or a part of the excited states $|m\rangle$, $m \neq 3/2$, by means of $\Delta m =$ one-, two-, and three-photon transitions. Figure 1 shows the two transition schemes, QC and RO, which will turn out to be appropriate for quantum computation and multiphoton Rabi oscillations, respectively. In the QC scheme the frequencies ω_k of the external transverse magnetic fields, $H_{x,k}(t) = \tilde{H}_k(t)\cos(\omega_k t + \Phi_k)$, $k = 1, 2, 3$, are blue ($\delta_k < 0$) and red ($\delta_k > 0$) detuned with respect to the energy level separations $\hbar\omega_{m,m'}$. In the RO scheme, the transverse fields $H'_{x,k}(t) = \tilde{H}'_k(t)\cos(\omega'_k t + \Phi'_k)$, $k = 1, 2, 3$, oscillate at frequencies $\omega'_{\Delta m} = \omega_{3/2-\Delta m,3/2}/\Delta m$, which are blue detuned by $3A$ ($6A$) for the two(three)-photon transition. For GaAs, $\omega_k, \omega'_k \sim 10$ MHz with $\delta_k \sim 1$ kHz, and a longitudinal magnetic field $H_z \sim 1$ T is appropriate. It is desirable to make H_z sufficiently large to accommodate many spin precessions before the spins dephase. We note that in contrast to the fields $H_{x,k}(t)$, the fields $H'_{x,k}(t)$ lead to transitions governed by noncommuting operators, with the important consequence that the RO scheme suffers from strong interferences between the transitions if two or more fields $H'_{x,k}(t)$ are nonzero, leading to a quick loss of amplitude control. Indeed, the RO scheme allows control of a_m's only for times $t \ll 2\hbar(V'_k + V'_{k'})/V'_k V'_{k'}$, which we estimate from the Baker-Campbell-Hausdorff formula and which we confirmed by exact numerical calculations. Here, $V'_k = 2[(g_N\mu_N H'_k)^k p_{3/2-k,3/2}]/\prod_{j=1}^{k-1}\hbar\omega_{3/2-j,3/2}$ (see below). Although the RO scheme is suited only for QCs using perturbative approaches, it has its usefulness for testing the coherence of the spin system (see below).

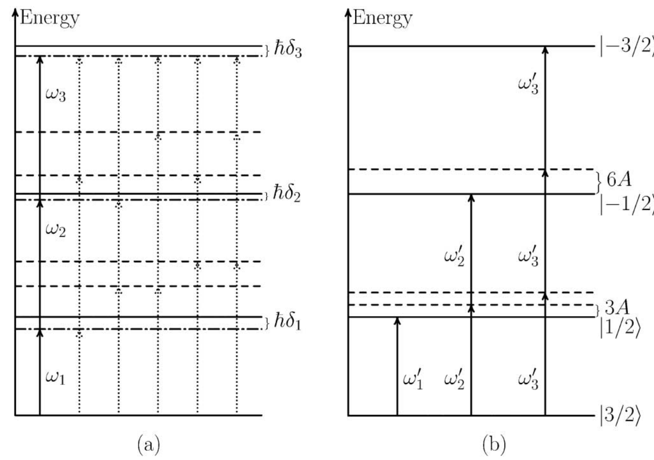

FIG. 1. Multiphoton transition schemes for the coherent population of the I_z eigenstates $|m\rangle$ of a nuclear spin $I = 3/2$. (a) Quantum computation (QC) scheme: the frequencies ω_k of the fields H_k are red (- ·) and blue (- -) detuned. Diagrams containing blue detunings are negligible for large quadrupolar splitting, i.e., $A \gg \hbar\delta_k \geq 0$. (b) Rabi oscillation (RO) scheme: the magnetic fields $H'_k\cos(\omega'_k t + \Phi'_k)$, $k = 1, 2, 3$, give rise to k-photon RO.

Now we proceed with demonstrating the existence of the desired spin transitions in the QC scheme. For this we evaluate the transition amplitudes for the diagrams of Fig. 1(a) in high-order perturbation theory which allows us then to obtain an appropriate nonperturbative Hamiltonian (see below). The three transverse fields $H_{x,k}(t)$ complete the Hamiltonian $\mathcal{H} = \mathcal{H}_0 + V(t)$, where $V(t) = \sum_{k=1}^{3} g_N\mu_N\tilde{H}_k(t)\cos(\omega_k t + \Phi_k)I_x$, with $I_x = (I_+ + I_-)/2$, and phases Φ_k (see below). Then we expand the S operator $S = \sum_{j=0}^{\infty} S^{(j)}$ in powers of $V(t)$. We use rectangular pulse shapes of duration T for all fields, i.e., $\tilde{H}_k(t) = H_k$ for $-T/2 < t < T/2$, and 0 otherwise. Then we obtain

$$\tilde{S}^{(3)}_{-\frac{3}{2}\frac{3}{2}} = \prod_{k=1}^{3} H_k e^{-i\Phi_k}\left[\frac{1}{\delta_1\delta_2} - \frac{1}{\delta_1(\frac{6A}{\hbar} - \delta_1 + \delta_2)} - \frac{1}{\frac{6A}{\hbar} + \delta_1 - \delta_2}\left(\frac{1}{\delta_2} - \frac{1}{\frac{12A}{\hbar} + \delta_1}\right) + \frac{1}{\frac{12A}{\hbar} + \delta_2}\left(\frac{1}{\frac{6A}{\hbar} - \delta_1 + \delta_2} + \frac{1}{\frac{12A}{\hbar} + \delta_1}\right)\right] \tag{1}$$

for $\delta_3 = 0$, $\tilde{S}^{(2)}_{-\frac{1}{2}\frac{3}{2}} = \prod_{k=1}^{2} H_k e^{-i\Phi_k}(-\frac{1}{\delta_1} + \frac{1}{\frac{6A}{\hbar} + \delta_1})$ for $\delta_2 = 0$ and $H_3 = 0$, and $\tilde{S}^{(1)}_{\frac{1}{2}\frac{3}{2}} = H_1 e^{-i\Phi_1}$ for $\delta_1 = 0$ and $H_2 = H_3 = 0$, where $S^{(j)}_{\frac{3}{2}-j,\frac{3}{2}} = \frac{2\pi}{i}(\frac{g_N\mu_N}{4\hbar})^j \tilde{S}^{(j)}_{\frac{3}{2}-j,\frac{3}{2}} p_{\frac{3}{2}-j,\frac{3}{2}}\delta^{(T)}(\omega_{\frac{3}{2}-j,\frac{3}{2}} - \sum_{k=1}^{j}\omega_k)$, $p_{m,m'} = \prod_{k=m}^{m'}\langle k|I_-|k+1\rangle$, and $\delta^{(T)}(\omega) = \frac{1}{2\pi}\int_{-T/2}^{+T/2} e^{i\omega t}dt = \frac{\sin(\omega T/2)}{\pi\omega}$ is the delta function of width $1/T$. The energy is conserved for $\omega T \gg 1$. Also, the duration T of the rf pulses must not exceed the dephasing time τ_ϕ of the spin states. Interestingly, $\lim_{A\to 0} S^{(3)}_{-\frac{3}{2}\frac{3}{2}} = \lim_{A\to 0} S^{(2)}_{-\frac{1}{2}\frac{3}{2}} = 0$; i.e., destructive interference is maximal. However, if $A \gg \hbar|\delta_k|$, $k = 1, 2, 3$, destructive interference is negligible.

Now we are in the position to extract an effective Hamiltonian, which governs the desired unitary evolutions[9,10]. For the QC scheme we use the Hamiltonian $\mathcal{H}$. After applying the rotating wave approximation [5] we keep only the most left diagram of Fig. 1(a), which gives the dominant contribution to the transition amplitudes for $\hbar|\delta_k| \ll |A|$. This is a direct consequence of the nonequidistance of the energy levels $|m\rangle$ due to the quadrupolar splitting. It is now possible to eliminate the time dependence of $\mathcal{H}$ by a unitary operation $U(t)$, the matrix elements of which can be determined by solving $2I$ linear equations. This is a transformation to a generalized rotating frame. Then, for a spin I we obtain an effective

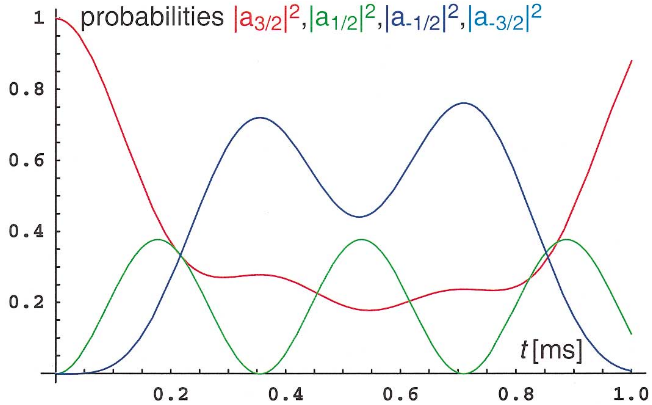

FIG. 2 (color). Preparation of $|s\rangle = (1/\sqrt{3})\sum_{m=-1/2}^{3/2}|m\rangle$ by means of Eq. (2) for ^{71}Ga nuclei in the QC scheme, which takes about 0.2 ms for $H_1 = H_2 = 1$ G, $H_3 = 0$, $\delta_1 = 6083\ \mathrm{s}^{-1}$, and $\delta_2 = 0$. The duration of the QC operation is $\leq 1/2\nu_{\mathrm{Rabi}}^{(2)}$. The analytical result is confirmed by numerics. Note that $a_{-3/2} = 0$.

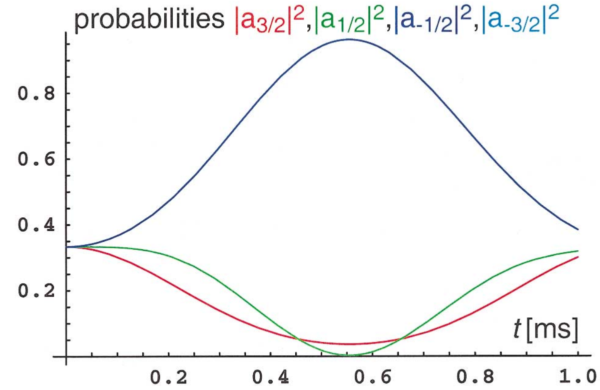

FIG. 3 (color). Grover algorithm calculated by means of Eq. (2) in the QC scheme (numerically confirmed), where $|s\rangle = (1/\sqrt{3})\sum_{m=-1/2}^{3/2}|m\rangle$ is concentrated mainly into $|-1/2\rangle$ after 0.55 ms for $H_2 = \hbar\delta_2/2g_N\mu_N = 1$ G, $h_1 = h_2$, $h_3 = 0$, $\delta_1 = 0$. The duration of the QC is $\leq 1/2\nu_{\mathrm{Rabi}}^{(2)}$. Note that $a_{-3/2} = 0$.

time-independent Hamiltonian

$$\mathcal{H}_{\mathrm{rot}}^{(2I)} = \begin{bmatrix} 0 & h_1 & 0 & \cdots & 0 \\ h_1 & \hbar\delta_1 & h_2 & \ddots & \vdots \\ 0 & h_2 & \hbar\delta_2 & \ddots & 0 \\ \vdots & \ddots & \ddots & \ddots & h_{2I} \\ 0 & \cdots & 0 & h_{2I} & \hbar\delta_{2I} \end{bmatrix}, \qquad (2)$$

where $h_k = g_N\mu_N H_k\sqrt{k(2I+1-k)}/2$ $(k = 1, \ldots, 2I)$. Focusing on $I = 3/2$, we obtain, e.g., for $H_3 = 0$ approximately $\mathcal{H}^{(2)} = \mathcal{H}_0 + h_1 e^{i(\omega_1 t + \Phi_1)}|3/2\rangle\langle 1/2| + h_2 e^{i(\omega_2 t+\Phi_2)}|1/2\rangle\langle -1/2| + \mathrm{H.c.}$ Applying $U(t) = e^{-i[(\omega_1+\omega_2)t+(\Phi_1+\Phi_2)]/2}|3/2\rangle\langle 3/2| + e^{i[(\omega_1-\omega_2)t+(\Phi_1-\Phi_2)]/2}|1/2\rangle\langle 1/2| + e^{i[(\omega_1+\omega_2)t+(\Phi_1+\Phi_2)]/2} \times |-1/2\rangle\langle -1/2|$ yields $\mathcal{H}_{\mathrm{rot}}^{(2)}$. Note that the Hamiltonian in Eq. (2) remains valid even in the limit $\delta_k \to 0$, where perturbation expansions such as in Eq. (1) break down. However, we must require that $|g_N\mu_N H_k| \ll |A|$, which means that the larger the $|A|$, the faster the QCs. Propagators of the form $U^\dagger(t)e^{-i\mathcal{H}_{\mathrm{rot}}^{(2I)}t/\hbar}$ have $2I$ phases Φ_k and $2I$ detunings $\hbar\delta_k$, which determine the $2I$ phases and the $2I$ moduli of a_m [15].

For Grover's algorithm [9,10] we must first produce $|s\rangle = (1/\sqrt{n})\sum_m |m\rangle$ (see Fig. 2), n being the number of basis states involved in the search. Then we make use of the degeneracy between $|s\rangle$ and $|M\rangle$, which yields the resonance condition $h_k = \hbar\delta_{I-M}/2 \neq 0\ \forall\ k$, if $\delta_k = 0\ \forall\ k \neq I - M$. In contrast to [9,10], $\mathcal{H}_{\mathrm{rot}}^{(2I)}$ has only nearest-neighbor coupling, which results in a decreasing amplification of $|M\rangle$ with increasing I or $|M|$. However, even for the largest nuclear spin $I = 9/2$, we find that the resolution for identifying $|M\rangle$ is still sufficient ($\gtrsim 10\%$). In Fig. 3 $|M = -1/2\rangle$ is found out of the three states $|m\rangle$ $m = 3/2, 1/2, -1/2$, for $I = 3/2$.

As a first test for the proposed schemes, it would be useful to measure generalized ROs involving multiphoton absorptions. They can be thought of as nutation of the large spin I between spin states $|m\rangle$. First, we consider the QC scheme. For the two-photon RO, with frequency $\nu_{\mathrm{Rabi}}^{(2)}$, to become observable, we need $|g_N\mu_N H_k| \ll \hbar\delta_1$, $k = 1, 2$, so that the one-photon transitions are completely suppressed. To obtain $\nu_{\mathrm{Rabi}}^{(2)}$, it is useful to think of (2) as describing the dynamics of a (fictitious) particle in a triple well with nearest-neighbor tunnel coupling h_k. The independent control of the tunnel couplings h_k and the biases $\hbar\delta_k$ between the wells are ensured by a large value of A. Then, the energy ("tunnel") splitting [16] between $|3/2\rangle$ and $|-1/2\rangle$ reads for $\delta_2 = 0$ as follows:

$$\Delta_{\mathrm{Rabi}}^{(2)} = \sqrt{3}(g_N\mu_N)^2 H_1 H_2/\delta_1, \qquad (3)$$

which gives $\nu_{\mathrm{Rabi}}^{(2)} = \Delta_{\mathrm{Rabi}}^{(2)}/2\pi\hbar$. In order to obtain large Rabi frequencies $\nu_{\mathrm{Rabi}}^{(2)}$, the external fields H_1, H_2 and the detuning $\hbar\delta_1$ are to be maximized under the conditions $|H_1|, |H_2| \ll \hbar|\delta_1|/g_N\mu_N \ll |A|/g_N\mu_N$ [17], i.e., the larger the $|A|$ the larger $\nu_{\mathrm{Rabi}}^{(2)}$ can be achieved. We note

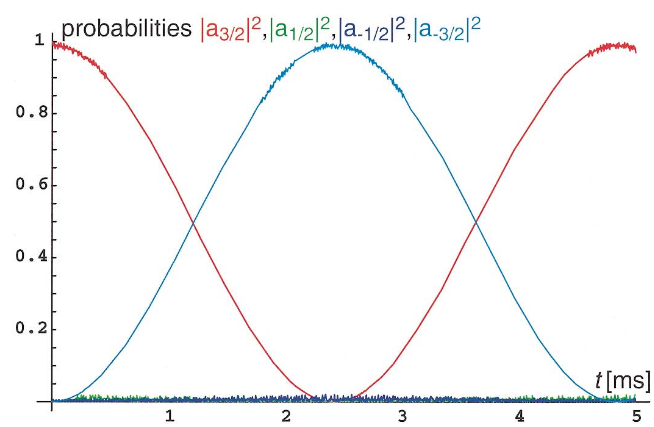

FIG. 4 (color). Numerical solution for the three-photon ROs of ^{75}As nuclei between $|3/2\rangle$ and $|-3/2\rangle$ driven by $H_3' = 20$ G with the RO scheme (b) in cw mode. $H_1' = H_2' = 0$.

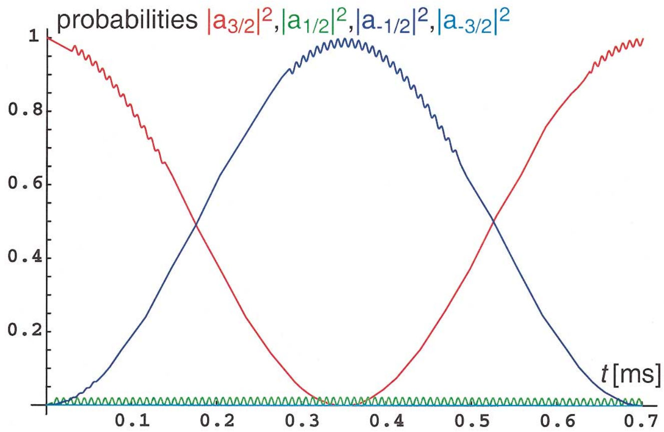

FIG. 5 (color). Numerical solution for the two-photon ROs of ^{75}As nuclei between $|3/2\rangle$ and $|-1/2\rangle$, driven by $H_2' = 10$ G according to the RO scheme in cw mode, and $H_1' = H_3' = 0$.

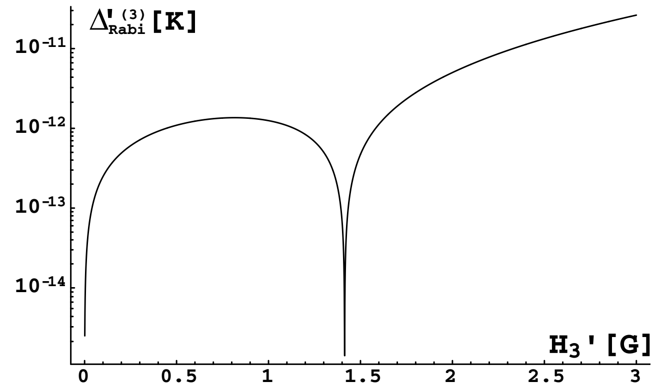

FIG. 6. Berry phase oscillation. The three-photon transition probability vanishes where $\Delta'^{(3)}_{\rm Rabi}$ is zero.

that $|A|$ could, e.g., be enhanced by optical laser pumping [3] or by modulated electric field gradients [12].

Next we turn to the RO scheme. Here it is sufficient to apply only one single field $H'_{x,k}(t)$ in order to see the multiphoton ROs shown in Figs. 4 and 5. We now also allow for oscillating quadratic transverse anisotropies which can be externally generated by modulating the electric field gradient felt by the nuclei [3,12]. For this we adopt the most general Hamiltonian [5]

$$\begin{aligned}\mathcal{H}' = A[3I_z^2 - I(I+1)] - g_N\mu_N H_z I_z \\ + e^{i\omega_k' t I_z}[-h_k' I_x + B(I_x I_z + I_z I_x) \\ + C(I_x^2 - I_y^2)]e^{-i\omega_k' t I_z},\end{aligned} \quad (4)$$

where $h_k' = g_N\mu_N H_k'$ ($k = 1$, 2, or 3). Next we transform $\mathcal{H}'$ to the rotating frame, which yields $\mathcal{H}'_{\rm rot} = A[3I_z^2 - I(I+1)] - (g_N\mu_N H_z I_z - \hbar\omega_k')I_z + B(I_x I_z + I_z I_x) + C(I_x^2 - I_y^2) + h_k' I_x$. Then the time evolution takes the simple form $|\psi(t)\rangle = e^{i\omega_k' t I_z} e^{-i\mathcal{H}'_{\rm rot}t/\hbar}|I\rangle$. Although the transverse quadratic term C is not in resonance with any transition energy, it leads to a time-independent transverse quadratic anisotropy in the rotating frame. For the 3-photon transition in the RO scheme we obtain the following Hamiltonian in the rotating frame:

$$\mathcal{H}'^{(3)}_{\rm rot} = \begin{bmatrix} 3A & \frac{\sqrt{3}}{2}h_3' & \sqrt{3}C & 0 \\ \frac{\sqrt{3}}{2}h_3' & -3A & h_3' & \sqrt{3}C \\ \sqrt{3}C & h_3' & -3A & \frac{\sqrt{3}}{2}h_3' \\ 0 & \sqrt{3}C & \frac{\sqrt{3}}{2}h_3' & 3A \end{bmatrix}, \quad (5)$$

where we have neglected the B term since we choose $B \ll h_3'$. Inserting a typical value $C = -10^{-10}$ K [3], we obtain oscillations of the splitting $\Delta'^{(3)}_{\rm Rabi}$ between $|3/2\rangle$ and $|-3/2\rangle$ as a function of H_3'; see Fig. 6. These oscillations are due to the Berry phase in a biaxial spin system as shown in [13]. Note that C must be negative for the Berry phase interference to occur [13]. Also, the Berry phase interference is present only for Δm-photon transitions with $\Delta m \geq 2$. In Figs. 4 and 5 the population-probabilities $|a_m(t)|^2$ are shown for $C = 0$. The corresponding normalized magnetization reads $M(t) = \sum_m m|a_m|^2$.

In conclusion, we have shown that via multiphoton transitions a controlled superposition of spin states can be achieved by appropriate field pulses. Our results can be extended to arbitrary spin I and to any single-particle quantum system with nonequidistant energy levels.

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