Quantum Information Processing with Large Nuclear Spins in GaAs Semiconductors

Michael N. Leuenberger and Daniel Loss

Department of Physics and Astronomy, University of Basel, Klingelbergstrasse 82, 4056 Basel, Switzerland

M. Poggio and D. D. Awschalom
Department of Physics, University of California, Santa Barbara, California 93106-9530

(Received 16 April 2002; published 28 October 2002)

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.

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 operation 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 \( |\psi\rangle = \sum a_m |m\rangle \), 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 2\( I \) 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 arbitrary \( I \). Our nuclear spin system is described by the Hamiltonian \( \mathcal{H}_0 = \mathcal{H}_Z + \mathcal{H}_Q \), consisting of the nuclear Zeeman energy \( \mathcal{H}_Z = -g_N \mu_N H \mathcal{I}_Z \), \( g_N = 1.3 \) [2], and the quadrupolar splitting [5] \( \mathcal{H}_Q = A(3\mathcal{I}_Z^2 - \mathcal{I}_+\mathcal{I}_- + 1) \). The quadrupolar constant is \( A = 7 \times 10^{-7} \) K for \( ^{69}\text{Ga} \), \( A = 3 \times 10^{-7} \) K for \( ^{71}\text{Ga} \), and \( A = 2 \times 10^{-6} \) K for \( ^{75}\text{As} \) 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 \( |\psi\rangle = \sum a_m |m\rangle \) 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 $\Delta m = 1$, 2, 3, and a longitudinal magnetic field $H_z \sim 1$ T. Interestingly, $|m\rangle$ is populated; see Fig. 1. This can be achieved only for times $t < \hbar \delta_k \approx 0$ and blue detunings are negligible for large quadrupolar splitting, i.e., $A \gg \hbar \delta_k \approx 0$. (b) Rabi oscillation (RO) scheme: the magnetic fields $H_k(t)$ of the fields $H_k(t)$ are red (- - -) and blue (- - -) detuned. Diagrams containing blue detunings are negligible for large quadrupolar splitting, i.e., $A \gg \hbar \delta_k \approx 0$. We note that in contrast to the fields $H_k(t)$ of the fields $H_k(t)$, the fields $H_k(t)$ lead to transitions governed by noncommuting operators, with the important consequence that the RO scheme suffers from strong interferences between the two or more fields $H_k(t)$ are nonzero, leading to a quick loss of amplitude control. Indeed, the RO scheme allows control of $a_m$'s only for times $t < 2\hbar (V_1^2 + V_1^3)/V_1^3$, which we estimate from the Baker-Campbell-Hausdorff formula and which we confirmed by exact numerical calculations. Here, $V_1^3 = 2[(g_N \mu_N H_i)^j p_{3/2-k,3/2}] / \prod_{j=1}^3 \hbar \omega_{3/2-j,3/2}$ (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).

$$\hat{S}^{(3)}_{-1/2} = \prod_{k=1}^3 H_k e^{-i\Phi_k} \left[ \frac{1}{\delta_1 + \delta_2} - \frac{1}{\delta_1 + \delta_2 + \Delta_2} \right]$$

for $\delta_4 = 0$, $\hat{S}^{(2)}_{-1/2} = \prod_{k=1}^3 H_k e^{-i\Phi_k} (- \frac{1}{\delta_1 + \delta_2} + \frac{1}{\Delta_2})$ for $\delta_2 = 0$ and $H_3 = 0$, and $\hat{S}^{(1)}_{-1/2} = H_1 e^{-i\Phi_1}$ for $\delta_1 = 0$ and $H_2 = H_3 = 0$, where $S^{(i)}_{j-1/2} = 2\pi (2\pi)^{i+j}/\prod_{j=1}^{i+j} (\omega_{j-1/2} - \omega_k) p_{m,m} = \prod_{k=1}^{i+j} (k |I| k + 1)$, and $S^{(i,j)}(\omega) = \frac{1}{2\pi} \int_{-\pi/2}^{+\pi/2} e^{i\omega t} d\omega = \sin(\omega T/2)/\pi\omega$ 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 $\tau_\phi$ of the states. Interestingly, $\lim_{A \to 0} S^{(3)}_{-1/2} \approx \lim_{A \to 0} S^{(2)}_{-1/2} \approx 0$; i.e., destructive interference is maximal. However, if $A \gg \hbar |\delta_k|$, $k = 1, 2, 3$, destructive interference is negligible.

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 $H_k(t)$ complete the Hamiltonian $H = H_0 + V(t)$, where $V(t) = \sum_{k=1}^{3} g_N \mu_N H_k(t) \cos(\omega_k t + \Phi_k) I_k$, with $I_k = (I_x + I_y)/2$, and phases $\Phi_k$ (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., $H_k(t) = H_k \cos(\omega_k t + \Phi_k)$ for $-T/2 < t < T/2$, and 0 otherwise. Then we obtain

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 $\omega_k$ of the fields $H_k$ are red (- - -) and blue (- - -) detuned. The frequencies $\omega_k$ of the fields $H_k$ are red (- - -) and blue (- - -) detuned.

Now we are in the position to extract an effective Hamiltonian, which governs the desired unitary evolution[9,10]. For the QC scheme we use the Hamiltonian $H$. After applying the rotating wave approximation [5] we keep only the 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 $H$ 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
time-independent Hamiltonian

$$H_{\text{rot}}^{(2)} = \begin{bmatrix} 0 & h_1 & 0 & \cdots & 0 \\ h_1 & 0 & h_2 & \cdots & 0 \\ 0 & h_2 & 0 & \cdots & 0 \\ \vdots & \vdots & \ddots & \ddots & \vdots \\ 0 & \cdots & 0 & h_{2J} & 0 \end{bmatrix}, \quad (2)$$

where

$$h_k = g_N \mu_N H^N \delta \left(2I + 1 - k\right)/2 \quad (k = 1, \ldots, 2I).$$

Focusing on $I = 3/2$, we obtain, e.g., for $H_1 = 0$ approximately

$$H_{\text{rot}}^{(2)} = H_0 + \frac{1}{2} \left( h_1 e^{i(\omega_1 + \Phi_1)J/2} - h_2 e^{i(\omega_2 + \Phi_2)J/2} \right) \left(1/2\right) + \text{H.c.}$$

Applying

$$U(t) = e^{-i\left( h_1 e^{i(\omega_1 + \Phi_1)J/2} + h_2 e^{i(\omega_2 + \Phi_2)J/2} \right)J/2}\left(1/2\right)} \quad (3)$$

which gives $\nu_{\text{Rabi}}^{(2)} = \Delta_{\text{Rabi}}^{(2)} / 2\pi h$. In order to obtain large Rabi frequencies $\nu_{\text{Rabi}}^{(2)}$, the external fields $H_1$, $H_2$ and the detuning $h\delta_1$ are to be maximized under the conditions $H_1^2, H_2^2 \ll h|\delta_1|^2 g N \mu_N \ll |A|/g N \mu_N$ [17], i.e., the larger the $|A|$ the larger $\nu_{\text{Rabi}}^{(2)}$ can be achieved. We note 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 $\nu_{\text{Rabi}}^{(2)}$, to become observable, we need $|g_N \mu_N H_1| \ll h\delta_1$, $k = 1, 2$, so that the one-photon transitions are completely suppressed. To obtain $\nu_{\text{Rabi}}^{(2)}$, 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 $h\delta_k$ between the wells are ensured by a large value of $A$. Then, the energy ("tunnel") splitting [16] between $[3/2]$ and $[-1/2]$ reads for $\delta_2 = 0$ as follows:

$$\Delta_{\text{Rabi}}^{(2)} = \sqrt{3} \left( g_N \mu_N \right) H_1 H_2 / \delta_1, \quad (3)$$

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

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 $h_k = h\delta_{k-M}/2 \neq 0 \forall k$, if $\delta_k = 0 \forall k \neq M$. In contrast to [9,10], $H_{\text{rot}}^{(2)}$ has only nearest-neighbor coupling, which results in a decreasing amplification of $|M\rangle$ with increasing $I$ or $|M\rangle$. However, even for the largest nuclear spin $I = 9/2$, we find that the resolution for identifying $|M\rangle$ is still sufficient ($\approx 10\%$). 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 $\nu_{\text{Rabi}}^{(2)}$, to become observable, we need $|g_N \mu_N H_1| \ll h\delta_1$, $k = 1, 2$, so that the one-photon transitions are completely suppressed. To obtain $\nu_{\text{Rabi}}^{(2)}$, 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 $h\delta_k$ between the wells are ensured by a large value of $A$. Then, the energy ("tunnel") splitting [16] between $[3/2]$ and $[-1/2]$ reads for $\delta_2 = 0$ as follows:

$$\Delta_{\text{Rabi}}^{(2)} = \sqrt{3} \left( g_N \mu_N \right) H_1 H_2 / \delta_1, \quad (3)$$

which gives $\nu_{\text{Rabi}}^{(2)} = \Delta_{\text{Rabi}}^{(2)} / 2\pi h$. In order to obtain large Rabi frequencies $\nu_{\text{Rabi}}^{(2)}$, the external fields $H_1$, $H_2$ and the detuning $h\delta_1$ are to be maximized under the conditions $H_1^2, H_2^2 \ll h|\delta_1|^2 g N \mu_N \ll |A|/g N \mu_N$ [17], i.e., the larger the $|A|$ the larger $\nu_{\text{Rabi}}^{(2)}$ can be achieved. We note
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 $H'_{ik}(t)$ in order to see the multiphoton ROs shown in Figs. 4 and 5. We now also allow for oscillating 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]

$$H' = A[3I_z^2 - (I(I + 1))] - g_N \mu_N H I_z + e^{i\omega t}[-h[I_x + B(I_x I_z + I_y I_y)] + C(I_x^2 - I_y^2)]e^{-i\omega t},$$

(4)

where $h' = g_N \mu_N H' (k = 1, 2, \text{ or } 3)$. Next we transform $H'$ to the rotating frame, which yields $H'_{rot} = H'[3I_z^2 - I(I + 1)] - (g_N \mu_N H I_z - h' I_z) + B(I_x I_z + I_y I_y) + C(I_x^2 - I_y^2) + h'I_x$. Then the time evolution takes the simple form $|\psi(t)| = e^{i\omega t}[e^{-iH'_{rot}t/|C|}]$. 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:

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

(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 $\Delta_{\text{Rabi}}^{(3)}$ 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 $\Delta m = 2$. 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 $J$ and to any single-particle quantum system with nonequidistant energy levels.

We thank A. J. Leggett and G. Salis for useful discussions. We acknowledge the Swiss NSF, NCCR Nanoscience, Molnanomag, and the U.S. NSF and DARPA for financial support.

[11] Like all implementations based on a “unary” representation, the present scheme is not scalable.
[15] We subtracted two degrees of freedom: the global phase and the normalization condition, respectively.
[17] This condition must also be satisfied by the numerical values in Ref. [7].