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Electric Field Tunability of Nuclear and Electronic Spin Dynamics due to the Hyperfine Interaction in Semiconductor Nanostructures

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We present formulas for the nuclear and electronic spin relaxation times due to the hyperfine interaction for nanostructed systems and show that the times depend on the square of the local density of electronic states at the nuclear position. A drastic sensitivity (orders of magnitude) of the electronic and nuclear spin coherence times to small electric fields is predicted for both uniformly distributed nuclear spins and δ-doped layers of specific nuclei. This sensitivity is robust to nuclear spin diffusion.

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Traditional semiconductor electronic devices are based on precise control of the electronic charge distribution using electric fields, ignoring the spin degrees of freedom of the electrons. Similar control over an electron's spin may lead to the development of new electronic devices with improved performance and new functionality [1,2]. Electronic spin coherence times exceed 100 ns at low temperatures in GaAs [3], and nuclear spin coherence times can exceed 1 s in GaAs quantum wells (QW) [4–6]. Because of these long coherence times, nuclear spins are also candidates for spin-based devices [7].

A natural way to control both electronic and nuclear spins would rely on magnetic fields. However, high magnetic fields are difficult both to achieve and to change rapidly. Furthermore, detection of nuclear magnetic resonance (NMR) signals from samples of reduced dimensionality is limited by the low nuclear polarization achievable with standard techniques [5]. Studies in semiconductor quantum wells [5,8–10] show that a strong local magnetic field and high nuclear polarization emerge as a consequence of optically induced dynamical nuclear polarization (DNP) [11] via hyperfine interaction. Kawakami *et al.* [12] have further demonstrated "imprinting" of nuclear spin polarization from adjacent ferromagnetic layers. Smet *et al.* [6] have manipulated nuclear spins by electrically tuning the electron density in a QW across a quantum Hall ferromagnet transition; the electric field tunes the nuclear spin relaxation time by changing the spectrum of collective mode excitations. Polarization of nuclei has also been predicted to alter electronic decoherence dynamics in quantum dots [13]. Hence, the electronic-nuclear spin interaction is of major interest, with implications for both electronic and nuclear spin lifetimes [14].

Here we derive general formulas applicable to nanostructures for the nuclear and electronic spin relaxation and decoherence times, T_1 and T_2, from the hyperfine interaction. The central physical quantity is the electronic local density of states (ELDOS) at the nuclei. We reanalyze the measurements of Ref. [5] using these formulas to obtain new values of the hyperfine coupling in GaAs QW's. We predict that the dominant process for nuclear T_1 in these QW's (and T_2 in others) can be tuned with an electric field by modifying the ELDOS at particular locations. For a parabolic QW electric-field tuning of nuclear spin relaxation by many orders of magnitude is possible, at temperatures considerably higher than in Ref. [6] and despite nuclear spin diffusion. The calculations of nuclear spin diffusion properly consider the ELDOS and inhomogeneous nuclear magnetization and indicate nonexponential long-time nuclear dynamics.

We assume nuclei are polarized through DNP and most of our calculations are performed at 30 K, where DNP is very efficient with typical laboratory magnetic fields (although tunability of T_1 and T_2, in principle, extends to much higher temperatures). In GaAs QW's the nuclear T_1 is dominated by the hyperfine interaction; however, nuclear dipolar interactions limit T_2 to 10^{-4} s. The electronic T_1 and T_2 in GaAs QW's are dominated by other processes. Therefore our specific predictions focus on control of the nuclear T_1. The general equations, however, are valid for describing the tuning of nuclear T_2 and electronic T_1 and T_2 in situations where the hyperfine interaction dominates those times. At the end of this Letter we propose several such situations.

For GaAs QW's we propose two different experimental configurations to demonstrate the electric-field tunability of the nuclear T_1. The same approaches can be used to tune nuclear T_2 and electronic spin decoherence in other material systems. In the first configuration, the T_1 of Ga and As nuclei in the nanostructure depends on the occupancy of conduction subbands, decreasing stepwise as the number of occupied conduction subbands (and hence the density of states) increases. Manipulation of the QW density, and implicitly the number of occupied subbands, can be accomplished with a gate voltage, permitting the manipulation of T_1. In the second configuration, a single δ-doped layer of a different material (such as In) is inserted at a specific position. The tunability of T_1 of these nuclei comes from the change in the electronic wave functions due to the applied electric field.

Our analysis of the electronic and nuclear spin relaxation times due to the hyperfine interaction in low dimensional systems follows in spirit the calculation by Overhauser [14] for bulk metals but now includes new effects due to the nanostructure. The interaction Hamiltonian can be written as

$$H = \frac{8\pi}{3}\beta_e\beta_n(\vec{\sigma}_n \cdot \vec{\sigma}_e)\delta(\mathbf{r} - \mathbf{r}_n), \tag{1}$$

where β_n and β_e are, respectively, the nuclear and electron magnetic moments, and $\vec{\sigma}_n$ and $\vec{\sigma}_e$ are the Pauli spin operators for the nucleus and electron, respectively. The argument of the delta function, $\mathbf{r} - \mathbf{r}_n$, represents the relative distance between the nuclear and electronic spins. The main effect of this Hamiltonian is a spin-flip process involving both the electronic and nuclear spins, which we evaluate using Fermi's golden rule.

The time dependence of the electronic magnetization is

$$\frac{dD}{dt} = \frac{D_0 - D}{T_{1e}} + G\frac{\Delta_0 - \Delta}{T_{1n}}, \tag{2}$$

where D and Δ are the electronic and nuclear magnetization with D_0 and Δ_0 their equilibrium values, and $G = 2I(I+1)(2I+1)/3$ (I represents the nuclear spin magnetic number). The electronic (T_{1e}) and nuclear (T_{1n}) relaxation times for general nanostructures and weak spin polarization are thus

$$T_{1e}^{-1} = \frac{1}{V}\sum_n \frac{1024\pi^3\beta_e^2\beta_n^2 \int d\varepsilon A_e^2(\mathbf{r}_n, \varepsilon) f'_{\mathrm{FD}}(\varepsilon)}{9\hbar(2I+1)\int d\mathbf{r}\, d\varepsilon A_e(\mathbf{r}, \varepsilon) f'_{\mathrm{FD}}(\varepsilon)} \tag{3}$$

and

$$T_{1n}^{-1}(\mathbf{r}_n) = \frac{512\pi^3\beta_e^2\beta_n^2 k_B T \int d\varepsilon A_e^2(\mathbf{r}_n, \varepsilon) f'_{\mathrm{FD}}(\varepsilon)}{3\hbar I(I+1)(2I+1)}, \tag{4}$$

where

$$A_e(\mathbf{r}_n, \varepsilon) = \sum_m |\psi_m(\mathbf{r}_n)|^2 \delta(\varepsilon - E_m). \tag{5}$$

Here $A_e(\mathbf{r}_n, \varepsilon)$ is the ELDOS (m labels the state, and $\psi_m(\mathbf{r}_n)$ the electron wave function of that state at the nucleus), $f_{\mathrm{FD}}(\varepsilon)$ is the Fermi-Dirac distribution function, and T is the temperature. If there is no energy bottleneck for the electron (e.g., there is none in QW's), the transverse spin decoherence rate T_2^{-1} from this mechanism is equal to T_1^{-1}. According to Eqs. (3)–(5), the electronic and nuclear spin relaxation times will depend on the position of the nuclei. T_{1e} is temperature independent, suggesting that it is possible for the hyperfine interaction to dominate T_{1e} at low temperatures, for the relaxation times corresponding to other electronic mechanisms increase as the temperature decreases [14,15].

For a QW the system's dispersion relations are quasi-two-dimensional; therefore, the electronic wave functions can be written as a product between an envelope function, $\phi(z)$, and a Bloch function, $u(\mathbf{r})$, such that $\psi_{j\mathbf{K}}(\mathbf{r}_n) = \exp[i\mathbf{K}\cdot\mathbf{R}]\phi_j(z)u(\mathbf{r}_n)$. For this situation $A_e(\mathbf{r}_n, \varepsilon) = \sum_j |\phi_j(z_n)|^2 N_{2\mathrm{D}}\Theta(\varepsilon - E_{j(\mathbf{K}=0)})$, where $N_{2\mathrm{D}}$ is the density of states for a two-dimensional electron gas and Θ is the Heaviside step function. Available experimental data for the nuclear spin relaxation time of a GaAs/$\mathrm{Al_{0.1}Ga_{0.9}As}$ QW [5] allow us to extract the value of the conduction band Bloch function, $|u(\mathbf{r}_n)|^2 = 5.2 \times 10^{25}\ \mathrm{cm}^{-3}$. This value compares well with $|u(\mathbf{r}_n)|^2 = 5.8 \times 10^{25}\ \mathrm{cm}^{-3}$ extracted from bulk GaAs in Ref. [16]. $\phi_j(z)$ is evaluated using a 14-band $\mathbf{k}\cdot\mathbf{p}$ calculation [17]. We consider now two different systems: a square GaAs QW ($L = 75$ Å) confined within two barriers of $\mathrm{Al_{0.4}Ga_{0.6}As}$ and a parabolic $\mathrm{Al}_x\mathrm{Ga}_{1-x}\mathrm{As}$ QW ($L = 1000$ Å) confined within two barriers of $\mathrm{Al_{0.4}Ga_{0.6}As}$. The parabolic QW is obtained by gradually varying the Al concentration, x, of $\mathrm{Al}_x\mathrm{Ga}_{1-x}\mathrm{As}$ layers from 0.4 in the two barriers to 0.07 in the center of the QW.

In Fig. 1 we present the position dependence of the relaxation times for the square GaAs [Fig. 1(a)] and parabolic $\mathrm{Al}_x\mathrm{Ga}_{1-x}\mathrm{As}$ [Fig. 1(b)] QW's for different conduction band occupancy. The shape of the curves describing $T_{1n}(z)$ are similar for the two considered situations. An initial nuclear polarization obtained by DNP will be inhomogeneous, and for short times will be proportional to $T_{1n}^{-1}(z)$, so for one occupied subband the initial nuclear magnetization $m(z, t=0) \propto |\phi(z)|^4$. The *initial* T_{1n} for the *total* Ga and As nuclear magnetization

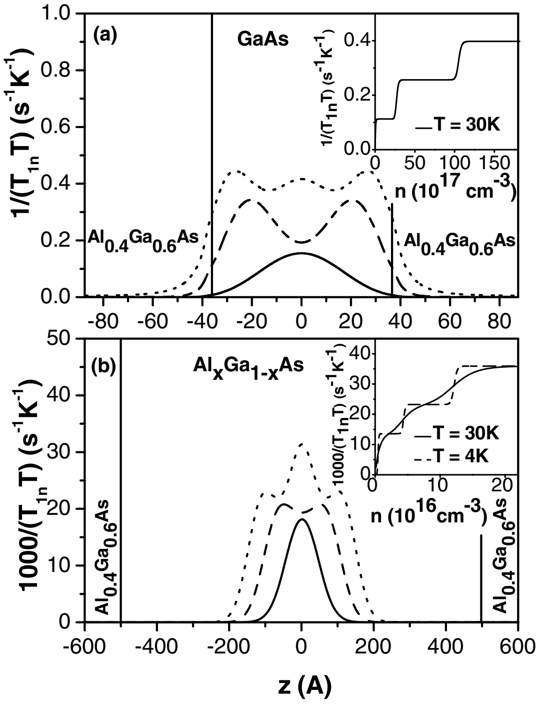

FIG. 1. The nuclear spin relaxation rate as a function of the position in the QW for different conduction subband occupancy at $T = 30$ K (full line: single subband occupancy, dashed line: double subband occupancy, and dotted line: triple subband occupancy). Inset: initial nuclear spin relaxation rate for different subband occupancy. (a) Square GaAs QW. (b) Parabolic $\mathrm{Al}_x\mathrm{Ga}_{1-x}\mathrm{As}$ QW.

initialized this way is plotted in the insets of Fig. 1 as a function of electron density. Note that as the electron density in the QW increases, the number of occupied conduction subbands will increase, and as a consequence the T_{1n} will decrease stepwise even for these uniformly distributed Ga and As nuclei. For the parabolic QW, where the energy difference between the minimum of two consecutive conduction subbands is about 15 meV, thermal smearing of the Fermi function at $T = 30$ K (solid line) will suppress the stepwise shape of T_{1n}. However, at $T = 4$ K, where the Fermi function is sharper (dashed line), the stepwise dependence of T_{1n} is observable. Application of an electric field across a QW can also tilt the confining potential. The direct dependence of both electron and nuclear spin coherence and relaxation times on the electronic envelope function suggests that control of spin relaxation times can thus be achieved by using such an external electric field $\mathbf{E}$. Salis *et al.* [18] suggested that the wave function shift with $\mathbf{E}$, and hence the electrical control of spin coherence, is particularly effective in a shallow parabolic QW.

We now consider the effects of shifting the electronic envelope wave functions to overlap different parts of the initial polarized nuclear population (different positions), *and nuclear spin diffusion,* by tracking the polarizations at the different positions as a function of time and then summing them to track the time dependence of the total nuclear polarization. For this and all subsequent calculations we consider electron densities where only the first subband is occupied. $m(z, t)$ can be obtained by solving

$$\frac{dm(z,t)}{dt} = D\frac{\partial^2 m(z,t)}{\partial z^2} - \frac{m(z,t)}{T_{1n}(z)}, \qquad (6)$$

where D represents the diffusion constant, whose value is of the order of 10^3 A^2/s for GaAs systems [19]. Our results indicate that the longer-time dynamics of the magnetization will be nonexponential.

In Fig. 2 we plot, for Ga and As nuclei which have been polarized via DNP at $\mathbf{E} = 0$, the time dependence of the total QW's nuclear magnetization for different values of the applied electric fields in the presence and the absence of spin diffusion. The inset shows the field dependence of the total initial nuclear spin relaxation time extracted as the first derivative of the magnetization at $t = 0$ s. The diminished overlap of the electron envelope function with the region of polarized nuclei reduces the relaxation rates [shown in Figs. 3(a) and 3(c)]. Magnetization decay in the square QW [Fig. 2(a)] is almost unaffected by the electric field, whereas for the parabolic QW [Fig. 2(b)] a large increase of the relaxation time is obtained even in small electric fields. In the presence of nuclear spin diffusion the effect of the electric field is reduced; in the parabolic QW, however, one can still see a significant difference between relaxation times at different applied electric fields. Recent measurements of nuclear spin diffusion in AlGaAs barriers indicated diffusion constants an order of magnitude smaller than in GaAs [20]. This suggests the tunability in the parabolic QW may be even more robust to diffusion than shown in Fig. 2.

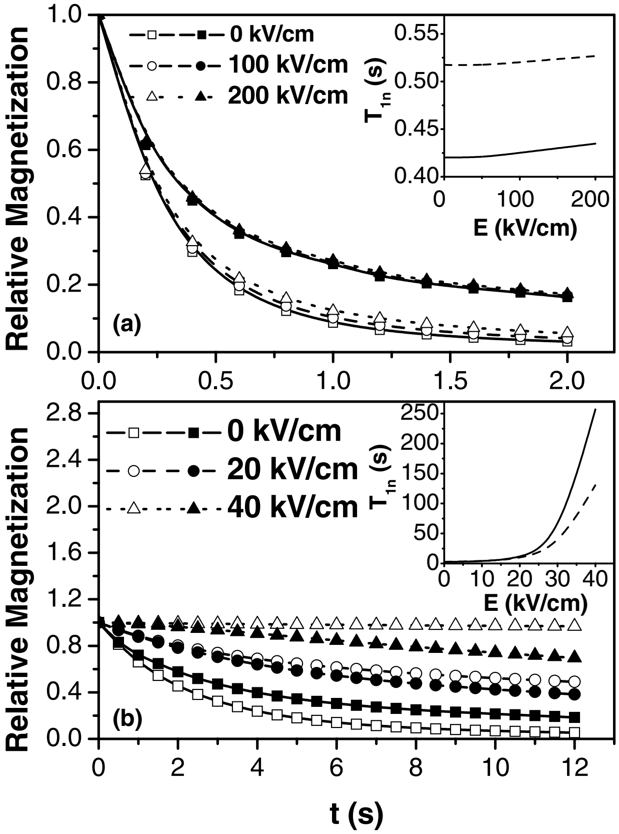

FIG. 2. The total relative nuclear magnetization as a function of time for different values of the applied electric field at $T = 30$ K in the presence (full symbols) and the absence (open symbols) of diffusion. Inset: total nuclear spin relaxation time as a function of the electric field in the presence (full line) and the absence (dashed line) of diffusion. (a) Square GaAs QW. (b) Parabolic Al$_x$Ga$_{1-x}$As QW.

An even more precise level of electric-field control is possible in structures which have been intentionally δ doped with a layer of different nuclei, such as In. For such a structure T_{1n}^{-1} depends on the position of the δ-doped layer according to Fig. 3(a), assuming the Bloch function on In is the same as that on Ga. Although T_{1n} for this layer could vary considerably, in a GaAs host T_{2n} would not because of transverse spin diffusion to or from the host nuclei. In Fig. 3(b) we plot the ratio of the spin relaxation times in the presence and the absence of an applied electric field as a function of the position along the growth direction for the square QW. We can see that the effect of the electric field is strongest within the two barriers. The effect of the electric field is far greater for the parabolic QW; the spin relaxation times increase 4 orders of magnitude for an electric field as low as 10 kV/cm [Fig. 3(d)], and this increase occurs in regions of large initial nuclear polarization. If the T_{1n} from the hyperfine interaction is made sufficiently long, eventually

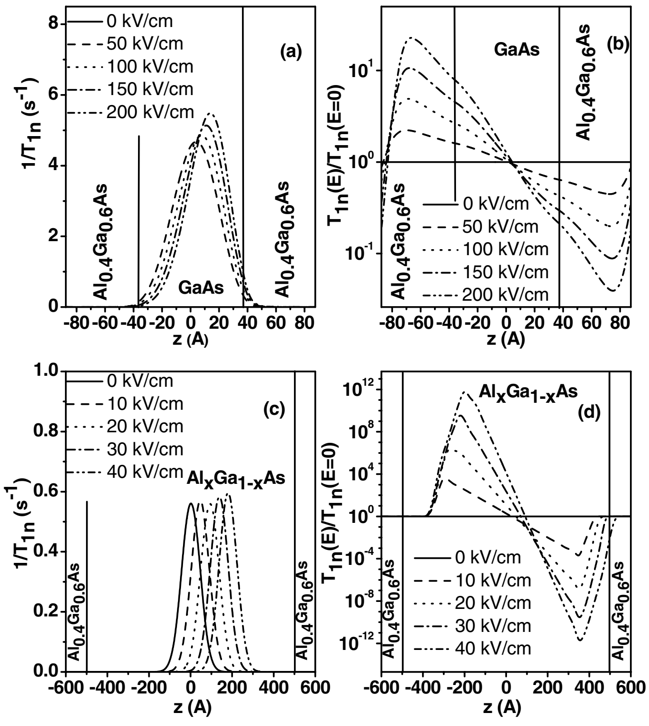

FIG. 3. The nuclear spin relaxation rate and the ratio of the relaxation times in the presence and the absence of the electric field as a function of the position in the QW for different values of the applied electric field at $T = 30$ K. (a),(b) Square GaAs QW. (c),(d) Parabolic $Al_xGa_{1-x}As$ QW.

the total T_{1n} will come to be dominated by the ~10 min T_{1n} time [21] from spin-phonon interactions.

From Eqs. (3)–(5) and T_{1n} from Ref. [5] we estimate the spin relaxation time via the hyperfine interaction for the *electron* in both the square and parabolic QW's of Fig. 1. For both structures we obtain $T_{1e} \approx 10^{-5}$ s for a single occupied subband. For uniformly distributed nuclei the electric-field dependence of T_{1e} is small, except for the density dependence (which is similar to that shown in the Fig. 1 inset for T_{1n}). The electronic relaxation time is $T_{1e} \sim 10^{-7}$ s from other processes; however, if these other processes could be suppressed times of 10^{-5} s might be observable. The influence of a δ-doped layer of nuclei on T_{1e} could also be electric-field tuned, with the same behavior as T_{1n} in Fig. 3.

We conclude by describing how to reduce competing processes for both the nuclear and electron spin coherence times. We have considered In as the δ-doped layer of nuclei in the GaAs QW. Although the different resonant frequency will limit the effect on the In T_{1n} of spin diffusion to the Ga and As nuclei, the host nuclei could significantly reduce the T_{2n} through dipole-dipole coupling. Another choice of QW, ZnCdSe/ZnSe, can be grown entirely from spin-0 nuclei; hence a δ-doped Mn layer in this structure should have a T_{2n} dominated by the tunable hyperfine interaction.

Electrons in either GaAs or ZnCdSe QW's may have T_{1e}'s limited by spin-orbit interaction. Si QW's in SiC (or SiO_2) [22], however, may have both spin-0 nuclei and weak spin-orbit interaction. Thin Si layers in these QW structures can have a direct band gap, so these layers could be probed or pumped optically. The electron spin coherence times could then be dominated by interactions with the δ-doped nuclei. In these QW's a good choice for a δ-doped nucleus would be the spin-1/2 Si nucleus.

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