Spin-echo dynamics of a heavy hole in a quantum dot

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We develop a theory for the spin-echo dynamics of a heavy hole in a quantum dot, accounting for both hyperfine- and electric-field-induced fluctuations. We show that a moderate applied magnetic field can drive this system to a motional-averaging regime, making the hyperfine interaction ineffective as a decoherence source. Furthermore, we show that decay of the spin-echo envelope is highly sensitive to the geometry. In particular, we find a specific choice of initialization and π-pulse axes which can be used to study intrinsic hyperfine-induced hole-spin dynamics, even in systems with substantial electric-field-induced dephasing. These results point the way to designed hole-spin qubits as a robust and long-lived alternative to electron spins.

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Electron spins in solid-state systems provide a versatile and potentially scalable platform for quantum information processing [1, 2]. This versatility often comes at the expense of complex environmental interactions, which can destroy quantum states through decoherence. Many theoretical and experimental studies have now established that the coherence times of electron spins in quantum dots [3-5], bound to donor impurities [6, 7], and at defect centers [8] are typically limited by the strong hyperfine interaction with surrounding nuclear spins [2-10]. Heavy-hole spin states in III-V semiconductor quantum dots have emerged as a platform that could mitigate the negative effects of the hyperfine interaction. Due to the p-like nature of the valence band in III-V materials, the contact interaction vanishes for hole spins, leaving only the weaker anisotropic hyperfine coupling [11, 12]. Moreover, the anisotropy of this interaction in two-dimensional systems should allow for substantially longer dephasing times in a magnetic field applied transverse to the quantum-dot growth direction [11, 11].

Recent experiments have measured hyperfine coupling constants for holes [13-15], as well as spin-relaxation ($T_1$) [16] and free-induction decay times, $T_2$, through indirect (frequency-domain) [17] and direct (time-domain) studies [18]. Coherent optical control has now been demonstrated for hole spins in single [19, 20] and double quantum dots [21]. This technique has been used to implement a Hahn spin-echo sequence [21] giving an associated spin-echo decay time, $T_2^\ast \sim 1 \mu s$. The $T_2^\ast$ value reported in Ref. [18] has been attributed to device-dependent electric-field fluctuations, rather than the intrinsic hyperfine interaction. Motivated by these recent experiments, here we present a theoretical study of heavy-hole spin-echo dynamics with an emphasis on identifying the optimal conditions for extending coherence times. In particular, we show that dephasing due to electric-field fluctuations, rather than the intrinsic hyperfine interaction, are highly sensitive to the geometry. In particular, we find a specific choice of initialization and π-pulse axes which can be used to study intrinsic hyperfine-induced hole-spin dynamics, even in systems with substantial electric-field-induced dephasing. These results point the way to designed hole-spin qubits as a robust and long-lived alternative to electron spins.

Figure 1. (Color online) (a) Quantum-dot geometry, with nuclear field $h_z$ and magnetic field $B = B\hat{z}$. For unstrained and flat quantum dots ($d \ll L$), $\gamma_H = g_H \mu_B \approx 0$ and $h_{x,y} \approx 0$ [11, 22]. (b) Hahn echo sequence. Two π-rotations ($U_x$ and $U'_x$, taken here about the $x$-axis) are applied at $t = \tau$ and $2\tau$, to refocus the HH spin.

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where $S = \sigma / 2$ is a pseudospin-1/2 operator in the two-dimensional ($J^z = \pm 3/2$) HH subspace and $I_k$ the nuclear spin at site $k$. $H_Z$ gives the hole- and nuclear-Zeeman interactions for an in-plane magnetic field $B = B\hat{z}$ [see Fig. 1(a)]. $\gamma_{ik}$ is the gyromagnetic ratio of isotope $k$ at site $k$ having total spin $I_k$. The hole gyromagnetic ratio is $\gamma_H = g_H \mu_B$, with $g_H$ the in-plane $g$-factor for a dot with growth axis along [001] and $\mu_B$ the Bohr magneton. The hyperfine interaction [10, 11] is expressed

$$H = H_Z + h_z S_z, \quad H_Z = -\gamma_H BS_x - \sum_k \gamma_{ik} B I^Z_k,$$

$$\gamma_{ik} = \frac{I^Z_k}{2I_k + 1}.$$
in terms of the Overhauser operator, \( \mathbf{h} = \sum_k A_k \mathbf{I}_k \). The coupling constants, \( A_k \), are given by \( A_k = A^iv_0|\psi(r_k)|^2 \), with \( A^i \) the hyperfine constant for isotope \( i \), \( v_0 \) the volume occupied by a single nuclear spin, and \( \psi(r_k) \) the HH envelope wavefunction. When the isotopes are distributed uniformly across the dot, we define the average \( A = \sum_k A_k \approx \sum_i \nu_i A^i \), with \( \nu_i \) the isotopic abundance. In this case, and for a Gaussian envelope function in two dimensions, \( A_k \approx (A/N)e^{-k/N} \) with \( N = 10^4 - 10^6 \) a typical number of nuclear spins within a quantum-dot Bohr radius. The ratio of \( |A| \) to the strength of the hyperfine coupling of electrons, \( |A^{(e)}| \), has been estimated theoretically \(^{11}\) in GaAs and confirmed experimentally \(^{13,15}\) in InGaAs and InP/GaInP to be \( |A/A^{(e)}| \sim 0.1 \). For simplicity, we will evaluate numerical estimates with a single averaged value \( |A^{(e)}| \sim 13 \mu eV \) \(^{11}\) and \( \nu_i, \gamma_i \) appropriate for In\(_{0.5}\)Ga\(_{0.5}\)As.

**Spin echo.** Under the action of Eq. (1), spin dephasing results from fluctuations in \( h_z \). Provided these fluctuations remain static on the timescale of decay of the hole spin, this source of decay can be removed via a Hahn echo [see Fig. 1(b)]. The process is better analyzed in the interaction picture with respect to \( H_z \),

\[
\hat{H}(t) = \hat{h}(t)\hat{S}_z(t),
\]

where, for any \( O \), \( \hat{O}(t) = e^{iHzt}Oe^{-iHzt} \). In particular, \( \hat{h}(t) = \sum_k A_k[I_k^z\cos(\gamma_{ik}Bt) - I_k^y\sin(\gamma_{ik}Bt)] \) and \( \hat{S}_z(t) = [\hat{S}_z\cos(\gamma_HBt) - \hat{S}_y\sin(\gamma_HBt)] \). The time-evolution operator after a time \( 2\tau \) is then given by

\[
\hat{U}(2\tau) = Te^{-i\hat{I}_k^z\tau}d\hat{h}(t),
\]

Here, the modified echo Hamiltonian,

\[
\hat{H}_e(t) = \begin{cases} \hat{H}(t) & 0 \leq t \leq \tau, \\ \sigma_\alpha \hat{H}(t)\sigma_\alpha & \tau \leq t \leq 2\tau, \end{cases}
\]

takes into account \( \pi_\alpha \)-pulses (\( \pi \)-rotations about \( \alpha = x, y, z \)). As seen in Eq. (11), \( \pi_\alpha \)-pulses (but in general not \( \pi_y, \pi_z \)) have the beneficial effect of inverting the sign of the Hamiltonian, \( \hat{H}(t) \to -\hat{H}(t) \), in the interval \( \tau \leq t \leq 2\tau \). Provided \( \hat{H}(t) \) is approximately static over the interval \( 0 < t < 2\tau \), this will induce time-reversed dynamics for \( \tau \leq t \leq 2\tau \), refocusing decay at the time \( 2\tau \). For this reason, unless otherwise specified, we will focus in the following discussion on a geometry with the magnetic field along \( \hat{z} \) and \( \pi_\alpha \)-pulses. We will contrast this analysis later with an alternate geometry relevant to recent experiments.

**Vanishing \( g_L \).** We first consider the limit \( \gamma_H = g_L \mu_B = 0 \) in Eq. (1). The dynamics we find in this limit will be a good description whenever \( \gamma_H < \gamma_i \), corresponding to \( g_L < 10^{-3} \) and \( g_L < 5 \times 10^{-3} \) has been reported in 2D wells \(^{25}\)). This limit considerably simplifies the theoretical analysis and allows for an exact solution: \( \hat{H} \) becomes block diagonal in the eigenbasis of \( \hat{S}_z \) and, in each block, the eigenstates are obtained after rotating \( I_k^z \)-eigenstates by an angle \( \theta_k = \pm \arctan(2\gamma_{ik}/A) \) about \( \hat{y} \). Representative results of the exact evolution of \( \langle S_x(2\tau) \rangle \) are shown in Fig. 2. The spin-echo signal has a remarkable dependence on the magnetic field: there is a clear transition from a low-field regime, where the decay time decreases with increasing \( B \), to a high-field regime, where there is no decay, only modulations of the echo envelope.

To give physical insight, we have developed an analytical approximation scheme based on the Magnus expansion. The Magnus expansion is an average-Hamiltonian theory typically applied to periodic and rapidly oscillating systems \(^{20}\). This scheme is suggested by the oscillating terms in Eq. (2), and will allow us to analyze the more general problem with \( \gamma_H \neq 0 \). In the Magnus expansion, we assume the evolution operator, Eq. (3), can be written as \( \hat{U}(2\tau) = e^{-iH_M(2\tau)} = e^{-i\sum_{\alpha}H^{(\alpha)}(2\tau)} \). The \( \alpha \)-th order term, \( \hat{H}^{(\alpha)}(t) \), is found using standard methods \(^{20}\). Each higher-order term in the Magnus expansion contains one additional integral over time. Oscillating terms are therefore suppressed by a factor of order \( ||\hat{H}||/\omega \), with \( \omega \) the typical oscillation frequency. The leading-order term is \( \hat{H}^{(0)}(t) = \hat{H}(t) \), where \( \hat{H}(t) \) is simply the average of \( \hat{H}_e(t) \) over an interval \( t \). The spin components \( S_\alpha \) (\( \alpha = x, y, z \)) are then given by

\[
\langle S_\alpha(2\tau) \rangle = \langle \hat{U}(2\tau)\hat{S}_\alpha(2\tau)\hat{U}(2\tau) \rangle = \langle e^{iL_M(2\tau)}\hat{S}_\alpha(2\tau) \rangle,
\]

where \( L_M(t) \) is defined by \( L_M(2\tau)\hat{O} = [H_M(2\tau), \hat{O}] \) and \( \langle \hat{O} \rangle = \text{Tr}[\hat{O}\rho] \). The initial state \( \rho = \rho_S \otimes \rho_I \) is assumed to describe a product of the hole-spin \( \rho_S \) and nuclear-spin \( \rho_I \) density matrices, where the nuclear spins are in an infinite-temperature thermal state. For \( N \gg 1 \) uncorrelated nuclear spins, the central-limit theorem gives...
nearly Gaussian fluctuations, resulting in

$$\langle e^{iL_M(2\tau)}\hat{S}_z \rangle \simeq \left\langle \exp\left(-\frac{1}{2}(L_M^2(2\tau))_I\right)\hat{S}_z \right\rangle_S,$$

(6)

where we define $\langle L_M^2(t)\rangle_O = \text{Tr}_t\{[L_M^2(t)O]_I\}$ and $\langle O\rangle_S = \text{Tr}_S\{O\}_S$.

At high $B$, rapid oscillations in $\hat{H}(t)$ allow us to keep only the leading term: $L_M^2(2\tau)O_S \simeq L^0(2\tau)O_S = [H^0(2\tau),O_S]$. Setting $\gamma_H = 0$, as appropriate for Fig. 4 and with the help of $\sum_i A_i^2 \simeq \nu_i A^2/(2N)$ (where the prime restricts the sum to nuclei of isotopic species $i$), we obtain:

$$\frac{\langle S_x(2\tau) \rangle}{\langle S_x(0) \rangle} \simeq \exp\left[-\sum_i \frac{4\nu_i A^2 I_i(I_i + 1)}{3N(\gamma_i B)^2} \sin^4\left(\frac{\gamma_i B\tau}{2}\right)\right].$$

(7)

As seen in Fig. 2, Eq. (7) (markers) reproduces the exact dynamics very well. The precise conditions for the validity of the Magnus expansion will be given below.

The simple form of Eq. (7) enables us to understand why the behavior of $\langle S_x(2\tau) \rangle$ changes as $B$ is increased. For $B \ll A/(\gamma_\text{rms}\sqrt{N})$ (gray solid line in Fig. 2), a short-time expansion of Eq. (7) gives $\langle S_x(2\tau) \rangle \simeq \langle S_x(0) \rangle (1 - (\tau/\tau_o)^4)$, with

$$\tau_o \simeq \frac{1}{\sqrt{B}} \left[\sum_i \frac{\nu_i (\gamma_i A)^2 I_i(I_i + 1)}{4N} \right]^{-1/4}.$$  

(8)

Surprisingly, when $B$ is increased, $\tau_o$ decreases. This behavior is opposite to the situation for electron spins, in which the echo decay time increases for increasing $B$. This decrease is due to rapid fluctuations in $h_z$ from nuclear spins precessing at frequencies $\sim \gamma_i B$. The Hahn echo can no longer refose these dynamical fluctuations at finite $B$, although Eq. (7) does predict partial reocurrences (dash-dotted line in Fig. 2) due to the finite number of discrete precession frequencies $\sim \gamma_i B$.

In contrast, at large magnetic field, $B \gtrsim A/(\gamma_i\sqrt{N})$, the system enters a motional-averaging regime in which the decay of $\langle S_x(2\tau) \rangle$ is bounded by $\langle A/B\gamma_i\sqrt{N}\rangle^2$, giving rise to beating (black solid curve in Fig. 2). This beating has the same physical origin as electron-spin-echo envelope modulation (ESEEM) [28], although the extreme anisotropy of the hole hyperfine interaction allows uniquely for its complete suppression. Fig. 3 shows the $1/e$ decay time, $\tau_o$, as $B$ is increased, leading to a discontinuity when $\gamma_i B\sqrt{N}/A \gtrsim 1$, at which point $\langle S_x(2\tau) \rangle$ always remains close to its initial value [see Eq. (7)].

Finite $g_\perp$. Although there are definite advantages to making flat unstrained dots leading to $g_\perp \simeq 0$ and $h_{x,y} \simeq 0$, current experiments are performed on hole systems with finite (albeit small) $g_\perp$ [18, 24]. For this general case, with $\gamma_H = g_\perp\mu_B \neq 0$, we have no closed-form exact solution for the dynamics, but our analysis can still be applied for a certain range of $\tau, B$.

We neglect subordinate oscillating terms in the Magnus expansion when $\|H^0(2\tau)\| \gg \|H^0(2\tau)\|_2, \|H^{(1)}\|^2$. More specifically, if the relevant fast oscillation frequency is $\omega \sim \gamma_i B$, each precessing nuclear spin experiences a typical hyperfine field $\delta \omega_{\text{rms}} \sim A/N$ from the hole. Otherwise, if the fast frequency is $\omega \sim \gamma_H B$, the hyperfine field acting on the precessing hole is of order $\delta \omega_{\text{rms}} \sim A/\sqrt{N}$, averaging over the nuclear configurations. As a consequence, the parameter $\delta \omega_{\text{rms}}/\omega < 1$ controls the expansion with $\delta \omega_{\text{rms}} = A/\sqrt{N}(A/N)$ and $\omega = \gamma_H B(\gamma_i B)$ for $\gamma_H \gg \gamma_i$. In addition to bounded oscillating terms, the Magnus expansion generates terms that grow with $\tau$. These terms approach $1$ at $\tau \sim \tau_{\max}$, beyond which a finite-order Magnus expansion fails. For $B \to \infty$, we find that progressively higher-order terms become relevant at a progressively shorter time, saturating at $\tau_{\max} \sim \omega/\delta \omega_{\text{rms}}^2$. However, for the special case $\gamma_H < \gamma_i$ there is an intermediate regime, $\delta \omega_{\text{rms}} < \gamma_i B \ll \sqrt{N}\delta \omega_{\text{rms}}$, in which low-order subleading terms dominate, giving $\tau_{\max} \sim (1/A)(\omega/\delta \omega_{\text{rms}})^2$. There is no such intermediate regime when $\gamma_H \gg \gamma_i$.

FIG. 3. (Color online) Decay time, $\tau_o$, vs. $B$. Here, $\gamma = \sum_i \gamma_i$ and parameters are as given in the caption of Fig. 2. Insets: typical $\langle S_x(2\tau) \rangle$ in each of the three regions: $\gamma B\sqrt{N}/A \ll 1$, $\gamma B\sqrt{N}/A \sim 1$, and $\gamma B\sqrt{N}/A \gg 1$.

The upshot of this analysis is that $\omega > \delta \omega_{\text{rms}}$ (i.e. $\gamma_i B > A/N$) can be easily satisfied for $\gamma_H \gg \gamma_i$, even outside of the motional-averaging regime (defined by $\gamma_i B > A/\sqrt{N}$), giving the good agreement shown in Fig. 2. In contrast, when $\gamma_H \gg \gamma_i$, the Magnus expansion is only strictly valid in the motional-averaging regime, $\gamma_H B > A/\sqrt{N}$ (Fig. 3). Nevertheless, in this regime the Magnus expansion will provide an accurate description whenever $\delta \omega_{\text{rms}}/\omega < 1$ and for $\tau \lesssim \tau_{\max}$.

In practice, the value $g_\perp = 0.04$ measured in [29] suggests that $\gamma_H \gg \gamma_i$ in many current experiments. For $g_\perp = 0.04$, the condition $B > A/(\gamma_H\sqrt{N})$ is already satisfied above a rather small value, $B \gtrsim 100$ mT for $N \sim 10^4$. In Fig. 4 we plot representative curves in this
The observed exponential decay is consistent with Gaussian white noise of the form \( \langle \delta \omega(t) \delta \omega(t') \rangle_{\delta \omega} = \frac{\gamma}{\Delta} \delta(t-t') \) (and \( \langle \delta \omega(t) \rangle_{\delta \omega} = 0 \)), where \( \langle \ldots \rangle_{\delta \omega} \) indicates averaging with respect to realizations of \( \delta \omega(t) \). We have included this additional dephasing mechanism in the evaluation of Eq. (7) for the \( \pi_x \)-pulse echo sequence examined previously and obtained a power-law decay at \( \tau \gg T_2 \):

\[
\frac{\langle S_y(2\tau) \rangle}{\langle S_y(0) \rangle} \approx \langle \exp \left[ \frac{\hbar^2}{2} \sum_{i=x,y,z} f_i^2(t) \right] \rangle_{\delta \omega} \approx \frac{1}{1 + \tau/\tau_D}.
\]

where \( f_i(t) = \int_{t}^{t+\tau} \phi(t) \delta \omega(t) dt \), \( f_i(t') = \int_{t'}^{t+\tau} \phi(t') dt' \), and

\[
\tau_D = \frac{1 + \gamma_h B T_2}{2 \hbar^2 I T_2}.
\]

This decay time scale is exceedingly long (\( \tau_D \approx 20 \) s) for the experimental value \( \gamma_h B \approx 2 \times 10^{11} \text{ s}^{-1} \) and using \( \langle h_2^2 \rangle I \approx 10^{13} \text{ s}^{-2} \), which demonstrates the negligible effect of spectral diffusion on the previous discussion (e.g., Figs. 2, 3, and 4). For simplicity, we have derived Eqs. (9) and (10) with static nuclear-field fluctuations \( \langle h_2^2 \rangle I \). This corresponds to a worst-case scenario for the present model. At the high magnetic field of Ref. [19] (\( B \approx 8 \) T), motional averaging would likely inhibit decay even further.

**Conclusion.** We have calculated the spin–echo dynamics of a single heavy-hole spin in a flat unstrained quantum dot. The relevant dynamics are highly anisotropic in the spin components and \( \pi \)-rotation axes. When \( \gamma_h \ll \gamma_i \), we predict an initial decrease of the coherence time with increasing \( B \), followed by a complete refocusing of the HH-spin signal and motional averaging when \( B \gtrsim B_c \) (\( B_c \approx A/\gamma_i \sqrt{N} \approx 3 \) T for \( N = 10^4 \)). The motional-averaging regime is also realized when \( \gamma_h \gg \gamma_i \), relevant to current experiments. In this regime, decay due to the hyperfine coupling can only occur for \( \tau \gtrsim \tau_{\text{max}} \propto B \), and can therefore be completely suppressed. We have further shown that device-dependent electric-field noise becomes negligible for a specific geometry, allowing for a measurement of the limiting intrinsic decoherence due to nuclear spins.

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In Eq. (1), we neglect terms $\sim h_{x,y}$, and nuclear quadrupole coupling. This is valid for a flat quantum dot with weak strain [11, 21, 22, 31]. The nuclear dipolar interaction may also influence the Hahn-echo decay of an electron spin on a timescale $\tau_{dd} \sim 10 \mu s \propto 1/\sqrt{A(e)}$ [32]. Since $|A| < |A(e)|$, we expect $\tau_{dd}$ to be still longer for holes, beyond the times considered here.