Electron spin synchronization induced by optical nuclear magnetic resonance feedback

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We predict a new physical mechanism to explain the electron spin precession frequency focusing effect recently observed in singly charged quantum dots exposed to a periodic train of resonant circularly polarized short optical pulses [A. Greilich *et al.*, Science **317**, 1896 (2007)]. We show that electron spin precession in an external magnetic field and a field of nuclei creates a Knight field oscillating at the frequency of the nuclear spin resonance. This field drives the projection of the nuclear spin onto the magnetic field to the value that makes the electron spin precession frequency a multiple of the train cyclic repetition frequency, the condition at which the Knight field vanishes.

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An electron spin localized in a single quantum dot (QD) is a natural qubit candidate for solid state quantum information processing.^{2–4} However, various optical or electrical control operations on an electron spin in a QD affect the nuclear spin polarization (NSP), which was observed as the Overhauser shift of the electron spin precession frequency in a magnetic field using various pump-probe techniques.⁵⁻⁸ The NSP is changed by electron-nuclear spin flip-flop processes resulting from Fermi contact hyperfine interactions.^{9,10} Such processes, however, are suppressed in a strong magnetic field because of an approximately three orders of magnitude mismatch in energy between the electron and nuclear Zeeman splittings. The NSP could be preserved on the time scale of hours,¹¹ unless special energy-conserving conditions for electron spinflip are reached.¹² Consequently, various manipulations with an electron spin by optical or electrical means become a main source of nuclear spin pumping, because during the action of these time-dependent perturbations, spin-flip processes can occur without energy conservation.^{1,8,13–17}

One of the most remarkable demonstrations of such a phenomenon is the nuclear induced frequency focusing (NIFF) effect that was discovered in an ensemble of singly charged QDs under excitation by a periodic train of short resonant pulses of circularly polarized light.¹ This experiment showed that the nuclei change their polarization to values that allowed precession frequencies of all electrons in the ensemble to satisfy the phase synchronization conditions (PSC). These are the frequencies at which the Larmor precession period of electron spin is equal to an integer fraction of the pump pulse repetition period.¹⁸ Why does the NSP, which changes randomly under light excitation, reach the value allowing electrons to satisfy the PSC? The authors of Ref. 1 suggested a connection with suppression of nuclear spin dynamics in such dots. Indeed, the train synchronizes the spin precession of electrons satisfying PSC and makes them optically passive at the moment of pulse arrival. This significantly slows down the light-stimulated random dynamics of the NSP in these QDs, leading to the accumulation of electron spins satisfying the PSC.

In this Communication, we demonstrate that the NIFF could be the result of the Knight field feedback-stimulated nuclear magnetic resonance (NMR). Our calculations treat the electron spin and NSP as classical vectors precessing around each other and an external magnetic field, and show that the NSP increases its projection onto the magnetic field monotonically with time therefore modifying the electron spin precession frequency. When the electron spin precession frequency has reached the PSC, the time-averaged electron spin polarization generated by the train and the corresponding Knight field causing the NSP modifications vanishes. The suggested mechanism should result in much faster frequency focusing than that connected with random fluctuations of the NSP.^{1,13,17}

We consider a singly negatively charged QD exposed to the train of circularly polarized pump pulses propagating along the structure growth axis z, arriving at the QD with the repetition period T_R , and also to a transverse magnetic field $B \parallel e_x$, where e_x is the unit vector along x axis [see inset in Fig. 1(a)]. It is assumed that the optical transition involves the excitation of a singlet X^{-} trion with the hole spin projection on the growth axis being $\pm 3/2$ for σ^+ and σ^- pump pulses, respectively. The pulse duration τ_p is short as compared with the spin precession period in the external magnetic field and as compared with the photocreated trion lifetime. The optical selection rules are therefore the same as in the absence of a magnetic field. In the interval between the optical pulses, the electron spin interacts with the NSP, $m = \sum_{i=1}^{n} I_i$, where I_i are the nuclear spins and the sum is over a mesoscopic number ($N \sim 10^5$) of nuclear spins. At equilibrium, in the studied magnetic fields, nuclei are practically unpolarized: they are randomly oriented and the NSP magnitude is controlled by random fluctuations of nuclear spin directions $|m| \sim \sqrt{N} \sim 3 \times 10^2$. To describe this electron-nuclei interaction, we treat the electron spin polarization, S, and m as classical vectors¹⁹ and adopt the box model²⁰⁻²² in which the interaction between electrons and nuclear spins does not depend on their positions. These approximations lead to the following equations for S and m in the interval between the optical pulses, $(n-1)T_R \leq t < nT_R$, where *n* is the pulse number:²³

$$\frac{\mathrm{d}S}{\mathrm{d}t} = \{ [\mathbf{\Omega} + \alpha \mathbf{m}(t)] \times \mathbf{S}(t) \},\tag{1a}$$

$$\frac{\mathrm{d}\boldsymbol{m}}{\mathrm{d}t} = \{ [\boldsymbol{\omega} + \alpha \boldsymbol{S}(t)] \times \boldsymbol{m}(t) \}. \tag{1b}$$



FIG. 1. (Color online) Time dependence of z component of the electron spin polarization S_z calculated after the train initiation (a), and after ~4000 repetition periods of the train (b). Panel (c) shows electron spin precession frequency calculated numerically (magenta) and analytically from Eq. (8) (black solid) and Eq. (11) (black dashed) curves. Inset to panel (a) illustrates geometry of a single QD excitation and shows a pump pulse and an electron (red) and nuclear (black) spins. Inset to panel (c) shows the absolute value of electron spin as a function of time. Calculations were conducted for $\alpha = 0.4, m = 23.5$, which corresponds to approximately 2200 nuclei with spin I = 1/2, $\Theta = 2\pi/3$, $\Omega T_R/(2\pi) = 8.5$, and $\omega = \Omega/500$.

Here, $\Omega = \Omega e_x$ and $\omega = \omega e_x$ are the electron and nuclear spin precession frequencies in an external field, respectively, and α is the hyperfine coupling constant between the electron and nuclear spins in the QD. The difference of electron and nuclear magnetic moments gives $\omega/\Omega \sim 10^{-3}$. The electron and nuclear spins in Eq. (1a) are coupled via an Overhauser field of fluctuation acting on the electron, αm , and a Knight field of the electron spin acting on the nuclei, αS . We neglect completely a slow nuclear spin relaxation connected with dipole-dipole interactions between nuclei in Eq. (1b).

The dynamics of the electron and nuclear spins in the QD have several very different time scales. Under experimental conditions¹ following inequalities hold

$$rac{2\pi}{\Omega}\llrac{2\pi}{lpha m}\lesssim T_R\llrac{2\pi}{\omega}\llrac{2\pi}{lpha}$$

These inequalities mean that (i) electron spin dynamics in the interval between pulses occurs in the permanent field of the frozen fluctuation of NSP and (ii) the dynamics of NSP is controlled only by the electron spin polarization averaged over the pulse repetition period:

$$S_0 = \frac{1}{T_R} \int_{(n-1)T_R}^{nT_R} S(t) dt.$$
 (2)

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Straightforward calculation shows that

$$S_{0} = \boldsymbol{n}(\boldsymbol{n} \cdot \boldsymbol{S}^{(a)}) + \frac{\boldsymbol{S}^{(a)} - \boldsymbol{n}(\boldsymbol{n} \cdot \boldsymbol{S}^{(a)})}{\Omega_{\text{eff}} T_{R}} \sin\left(\Omega_{\text{eff}} T_{R}\right) + \frac{[\boldsymbol{S}^{(a)} - \boldsymbol{n}(\boldsymbol{n} \cdot \boldsymbol{S}^{(a)})] \times \boldsymbol{n}}{\Omega_{\text{eff}} T_{R}} [1 - \cos\left(\Omega_{\text{eff}} T_{R}\right)], \quad (3)$$

where $S^{(a)}$ is the electron spin polarization right after the excitation pulse, $\mathbf{n} = (\mathbf{\Omega} + \alpha \mathbf{m}) / \Omega_{\text{eff}}$ is a unit vector along the effective field, and $\Omega_{\text{eff}} = |\mathbf{\Omega} + \alpha \mathbf{m}| \approx \Omega + \alpha m_x$. One can see from Eq. (3) that the average electron spin polarization S_0 transverse to \mathbf{n} vanishes when Ω_{eff} satisfies the PSC: $\Omega_{\text{eff}} T_R = 2\pi K$ with K = 1, 2, ... The longitudinal component does not vanish at the PSC due to a small deviation of \mathbf{n} from the magnetic field direction caused by the nuclear field. As we show below, the transverse components of an electron spin are required for the NSP modification. As a result, the electron spin does not affect the nuclei if the PSC is fulfilled. If the PSC is not satisfied, however, the weak Knight field, αS_0 , modifies the NSP and drives its projection, $m_x(t)$, to the value that allows Ω_{eff} to satisfy the PSC.

To describe this effect, we need to complement Eq. (1a), which describes the electron-nuclear spin dynamics in the interval between pulses, by the relationship between the electron spin polarization before, $S^{(b)}$, and after, $S^{(a)}$, the pump pulse, which for resonant σ^+ excitation reads²⁴

$$S_{z}^{(a)} = \frac{Q^{2} - 1}{4} + S_{z}^{(b)} \frac{Q^{2} + 1}{2}, \quad S_{x}^{(a)} = Q S_{x}^{(b)}, \quad S_{y}^{(a)} = Q S_{y}^{(b)},$$
(4)

where $Q = \cos \Theta/2$ and $1 - Q^2$ is the probability of trion creation by the short circularly polarized pulse with area Θ . Numerical integration of Eqs. (1), which uses Eq. (4), clearly demonstrates the NIFF effect as one can see in Fig. 1. Calculations were conducted for the electron spin precession frequency, which does not satisfy the PSC: $\Omega T_R/(2\pi) = 8.5$, and an initial condition for the NSP, which was selected as $m \parallel e_z$. We exaggerated the value of α and reduced the number of nuclei from a typical value in a QD $N \sim 10^5$ down to $N \sim$ 2×10^3 to conduct numerically accurate calculations within reasonable computational time. Otherwise, the difference in the characteristic times of electron and nuclei spin dynamics requires carrying out calculations on a time scale covering nine orders of magnitude.

Figure 1(a) shows the temporal dynamics of the electron spin z component for the 6th and 7th repetition periods where the electron spin dynamics is already stationary¹⁸ but the nuclear effects have not come into play. Panel (b) shows those dynamics for the 3998th and 3999th periods when the nuclear spin precession had already taken place. One can see that the slow nuclear spin dynamics changes qualitatively the character of electron spin precession in this time interval: the amplitude of electron spin polarization is strongly enhanced and reaches its maximum value 1/2, see inset in Fig. 1(c). The effect is connected with the temporal dynamics of the electron effective spin precession frequency shown in Fig. 1(c). Apart from the oscillations of frequency ω related to the NSP precession, $\Omega_{\rm eff}$ initially grows linearly in time and then saturates at the multiple of $2\pi/T_R$. The periodic train of short pulses synchronizes the electron spin precession in the QD where Ω_{eff} satisfies the



FIG. 2. (Color online) (a) Schematic illustration of electron spin precession in quasistatic field $\Omega + \alpha m(t)$ (top) and temporal dependence of S_x (bottom). (b) Geometry of NMR induced by steady-state $\alpha \overline{S}_{y,0}$, and alternating, $\alpha S_x(t)$, Knight fields. Bottom panel shows static and oscillating fields in the (xy) plane.

PSC, leading to complete polarization of electron as seen in Fig. 1(b).

To understand the physical mechanism responsible for NIFF demonstrated in Fig. 1, let us first consider the effect of the nuclear spin precession on the electron spin dynamics. Since nuclear spin precession is slow as compared with electron spin precession and pump pulse repetition periods, one can treat the electron spin dynamics in the interval between pulses as a precession in the static field $\Omega + \alpha m$ [see Fig. 2(a)]. Using the procedure from Ref. 24 and Eqs. (3) and (4), we derive a steady state (on the timescale of T_R) value of S_x exposed to the train of optical pulses:

$$S_x \equiv S_{x,0}(t) = \alpha m_z(t) C_x / \Omega,$$

$$C_x = -\frac{2Q \sin^2 (\Omega_{\text{eff}} T_R / 2) + (Q - 1)^2 / 2}{(Q - 1)^2 + 2(Q + 1) \sin^2 (\Omega_{\text{eff}} T_R / 2)},$$
(5)

One can see from Eq. (5) that $S_x(t)$ oscillates slowly with the NSP precession frequency ω . This occurs because the electron spin precession axis [see Fig. 2(a)] is tilted from the *x* axis in the (*xz*) plane by the small angle $\alpha m_z(t)/\Omega$. The precession leads to a nonzero S_x projection of the electron spin, which value is proportional to the tilt angle oscillating at the frequency ω . The same geometrical arguments show that $S_{0,y}$ and $S_{0,z}$ are the sum of the time-independent terms $\overline{S}_{0,y}$ and $\overline{S}_{0,z}$ and small terms oscillating at 2ω that can be neglected. As a result, the NSP, which precesses around the static field $\omega + \alpha \overline{S}_0$ (the latter is slightly tilted from the *x* axis) experiences the alternating Knight field $\alpha S_x(t)$ [see Fig. 2(b)]. Since $S_x(t)$ oscillates with ω , it drives the NMR and leads to slow modification of m_x , as shown below.

To describe the time dependence of $m_x(t)$, we need to take into account that the NMR driving field, $\alpha S_x(t)$, is almost parallel to the static field. At first, it creates a time-independent shift of the NSP, \bar{m}_z , along the *z* axis. Indeed, in the first approximation on α :

$$m_z(t) = m_\perp \cos\left[\omega t + \int_0^t \alpha S_x(t') dt'\right],\tag{6}$$

where $m_{\perp} = \sqrt{m^2 - m_x^2}$ is the perpendicular component of the NSP. In the same approximation on α , Eq. (6) can be rewritten as $m_z(t) \approx m_{\perp} \cos \omega t + \bar{m}_z$, where $\bar{m}_z = -\alpha^2 m_{\perp}^2 C_x/(2\omega\Omega)$. The analogous calculation shows that $\bar{m}_y = 0$.

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Secondly, the NMR is caused only by the component of the alternating field perpendicular to the NSP precession axis, which is equal to $\alpha(\alpha S_0/\omega)S_x(t)$. Averaging the *x* component of Eq. (1b), $dm_x/dt = \alpha(S_ym_z - S_zm_y)$, over a sufficiently long temporal interval $\Delta T \gg 1/\omega \gg 1/\Omega$, we obtain the standard NMR expression:

$$\frac{\mathrm{d}m_x}{\mathrm{d}t} = \alpha \overline{S}_{y,0} \overline{m}_z = -\frac{\alpha^3 \overline{S}_{y,0} C_x m_\perp^2}{2\omega \Omega},\tag{7}$$

where the averaged m_y is equals to zero: $\bar{m}_y = 0$. Generally, the right-hand side of Eq. (7) depends on m_x via $S_{y,0}$ and C_x dependence on Ω_{eff} . One can neglect this dependence if Ω_{eff} is not very close to the PSC. In this case, we obtain for $m_x(t)$:

$$\frac{m_x(t)}{m_\perp(0)} \approx \frac{t}{\tau_{\rm nf}}, \quad \frac{1}{\tau_{\rm nf}} = -\frac{\alpha^3 m_\perp(0)}{2\omega\Omega} \overline{S}_{y,0} C_x, \tag{8}$$

where $m_{\perp}(0)$ is the initial value of the perpendicular component of the NSP. One can see from Eq. (8) that $m_x(t)$ and, consequently, Ω_{eff} grow linearly with time. The analytical dependence $\Omega_{\text{eff}}(t)$ shown by the solid line in Fig. 1(c) is in good agreement with results of the numerical calculations.

In the case where Ω_{eff} is close to the phase synchronization condition, which is fulfilled if $m_x = m_x^{\text{PSC}}$, one can rewrite Eq. (7) using Eq. (3) as

$$\frac{\mathrm{d}m_x}{\mathrm{d}t} = \frac{\left(m_x - m_x^{\mathrm{PSC}}\right)^2}{m\tau_{\mathrm{nf}}^2},\tag{9}$$

where

$$\frac{1}{\tau_{\rm nf}'} = \frac{\alpha^5 m T_R}{16\omega\Omega^2} \frac{1+Q}{1-Q} \left[m^2 - \left(m_x^{\rm PSC} \right)^2 \right].$$
 (10)

The dynamics of $m_x(t)$ in this case is described by

$$m_x(t) = m_x^{\text{PSC}} - m \frac{\tau'_{\text{nf}}}{t - t_0},$$
 (11)

where t_0 is an arbitrary constant, chosen to merge the time dependencies given by Eqs. (8) and (11) at $t \sim \tau_{nf}$. The corresponding long-time asymptote of Ω_{eff} is plotted in Fig. 1(c) by a dashed line.

Although $dm_x/dt = 0$ at $m_x = m_x^{\text{PSC}}$, these m_x 's are only the saddle points in the m_x time dependence. For positive τ'_{nf} , the points are stable if $m_x < m_x^{\text{PSC}}$ and unstable otherwise. This means that m_x returns to m_x^{PSC} only if its fluctuation $\delta = m_x - m_x^{\text{PSC}} < 0$. If $\delta > 0$, the fluctuation causes the deterministic growth of m_x until the next PSC with larger m_x is met. Including a weak nuclear spin relaxation in Eq. (1b) gives two $m_x = m_x^{\text{PSC}} \pm \sqrt{m_x^{\text{PSC}}m\tau_{nf'}/T_1}$, at which $dm_x/dt = 0$, where $T_1 \gg \tau_{nf'}$ is the nuclear spin relaxation time. One of these solutions, $m_x = m_x^{\text{PSC}} - \sqrt{m_x^{\text{PSC}}m\tau_{nf'}/T_1}$, is a stable point of $m_x(t)$. A switch of the light polarization from σ^+ to σ^- does not change the direction of the m_x growth, as can be seen from Eqs. (8) and (11).

Figure 3 shows the dependence of the NIFF time, τ_{nf} , defined Eq. (8) on the pump pulse area, Θ . One can see that τ_{nf} becomes extremely long for $\Theta \ll 1$ because electron spin orientation is inefficient and the averaged electron spin S_0 is very small under these conditions. Growth of Θ increases S_0 and consequently the Knight field, αS_0 , which in turn shortens τ_{nf} . Further increase of τ_{nf} with Θ seen in Fig. 3



FIG. 3. (Color online) (a) Dependence of the NIFF time, τ_{nf} , on the pulse area, Θ . The three curves were calculated at the magnetic fields that give the following electron spin precession frequencies: $\Omega T_R/(2\pi) = 50.5$ (black), 100.5 (red), and 150.5 (blue). The parameters used, $\alpha = 5 \times 10^6 \text{ s}^{-1}$, $T_R = 13 \text{ ns}$, $\omega/\Omega = 10^{-3}$, and m = 126, which could be created by 6×10^4 nuclei with spin 1/2, were selected to keep the calculations relevant to Refs. 1,18. (b) and (c) Time dependence of the average z component of the electron spin $\overline{S}_z(t)$ during 3997 $T_R < t < 3999 T_R$ time interval. The averaging was conducted over 25 initial directions of NSP *m*. Panel (b) is calculated for the frozen nuclear fluctuation [$\alpha = \omega = 0$ in Eq. (1b)], and panel (c) is calculated taking the nuclear spin precession into account ($\alpha =$ 0.4, $\omega = \Omega/500$). Other parameters are the same as in Fig. 1.

is connected with the periodic dependence of S_0 on Θ . The frequency focusing time $\tau_{nf} \propto \Omega^2 \omega$ increases significantly with a magnetic field. This explains the rapid increase of τ_{nf} with Ω seen in Fig. 3.

We have considered electron-nuclear spin dynamics in a single QD with a certain initial orientation of NSP. To describe a QD ensemble, we average over different initial orientations of the NSP. The time dependence of the average *z* component of the electron spin $\overline{S}_z(t)$ is shown in Figs. 3(b) and 3(c). The initial NSP orientations m(0) were chosen to be isotropically distributed, with the magnitude m(0) = m = 23.5 used to describe the spin dynamics of a single QD in Fig. 1. Figure 3(b)

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shows the electron spin dynamics in the absence of nuclear spin dynamics, which is simulated using $\alpha = 0$ and $\omega = 0$ in Eq. (1b). One can see that $S_z(t)$ partially decays between the pump pulses and the phase of spin beats jumps at each repetition period. Comparison with Fig. 1(a) shows that the amplitude of $\overline{S}_{7}(t)$ after 4000 repetition periods is the same as at the initial precession stage. The nuclear spin precession in the external magnetic field and in the Knight field tunes up the electron spin precession frequency and leads to a very pronounced mode-locking of electron spin coherence seen in Fig. 3(c). One can see a clear rise of $\overline{S}_{\tau}(t)$ before pulse arrival. The NIFF effect also significantly increases the $\overline{S}_{z}(t)$ amplitude relative to those shown in Fig. 3(b). For the parameters used in this calculation, $\alpha m T_R/(2\pi) \approx 1.5$, the main contribution to $\overline{S}_{z}(t)$ comes from the two Ω_{eff} 's satisfying the PSC $\Omega_{\rm eff}T_R/2\pi = 8$ and 9.

We note that if the NIFF effect is a consequence of random fluctuations of the NSP as suggested in Refs. 1,13, the rate of this process can be estimated as $\gamma_n \sim (1 - Q^2)\alpha^2/(\Omega^2 T_R)$. The current classical model leads to a much faster NIFF in the QD ensemble studied in Ref. 1 because $1/(\gamma_n \tau_{nf}) \sim \alpha m/\omega \sim 10$ in these experiments. Quantum-mechanical extension of the suggested model was carried out by M. Yu. Petrov.²⁵

In summary, we have suggested a new physical mechanism of the NIFF effect for the electron spin precession.¹ This mechanism leads to a monotonic shift of the electron spin precession frequency with time and allows this frequency to reach phase synchronization condition with the train repetition period much faster than in the case when the NIFF is a consequence of random fluctuations of the nuclear spins as was suggested earlier. Further experimental studies of the NIFF time dependence on a magnetic field and a pulse area should provide evidence for the suggested mechanism.

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