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Feedback control of nuclear hyperfine fields in a double quantum dot

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Abstract – In a coupled double-quantum-dot system, we present a theory for the interplay between electron and nuclear spins when the two-electron singlet state is brought into resonance with one triplet state in moderate external magnetic field. We show that the quantum interference between first-order and second-order hyperfine processes can lead to a feedback mechanism for manipulating the nuclear hyperfine fields. In a uniform external field, positive- and negative-feedback controls can be realized for the gradient of the longitudinal hyperfine field as well as the average transverse hyperfine field in the double dot. The negative feedback which suppresses fluctuations in the longitudinal nuclear field gradient can enhance the decoherence time of a singlet-triplet qubit to microsecond regime. We discuss the possibility of enhancing the decoherence time of each individual spin in a cluster of dots using the negative-feedback control on the transverse nuclear field.

Introduction. – A single electron spin in a semiconductor quantum dot is an attractive candidate for a solid-state quantum bit [1]. Recent experiments have demonstrated the capability of reading, writing, and controlling of single spin in different III–V quantum dot systems, by optical means [2–7], or electric means [8–14]. An outstanding bottleneck towards spin-based quantum computation has been the fast dephasing of the electron spin by the nuclear spin environment in the III–V materials. Even at temperature as low as ∼100 mK, the thermal statistical fluctuation of the nuclear spin configurations still corresponds to a large inhomogeneous broadening in nuclear hyperfine field, which dephases the electron spin in a timescale of $T_2^* \sim 1$–10 ns [10,11,15–17]. The spin-echo approach can transiently remove inhomogeneous dephasing at certain spin-echo times for an ultrashort duration equal to T_2^* time [12,14], but this may be insufficient to allow the general quantum logic controls.

In principle, the nuclear field inhomogeneous broadening can be narrowed below its thermal value after proper procedures of nuclear state preparation [18–24], and the resultant enhancement on the T_2^* time can last for seconds or even longer as nuclear spin relaxation is extremely slow. This could be a solution for preparing a spin qubit with desirable coherence properties. For optically controllable electron spin in self-assembled dots, the enhancement of T_2^* up to microsecond timescales by nuclear state preparation has been achieved for a spin ensemble [25], and for a single spin [26]. For electrically controlled double quantum dots, an experimental paper has reported that a cyclic control of the two-electron state through the resonance between the singlet and one triplet state can enhance the inhomogeneous dephasing time of the two-spin states to microsecond timescale [27]. A phenomenological model was later proposed which shows that such control could result in negative feedback to suppress the fluctuation of longitudinal nuclear field gradient and as a result the two-spin dephasing time can be enhanced [28].

In this work, we present a theory for the interplay between electron and nuclear spins near the singlet-triplet resonances of a coupled double dot in moderate external magnetic field. The transition from the singlet to a triplet in resonance can either be realized through a first-order process by the electron-nuclear flip-flop term in the hyperfine coupling, or through a second-order hyperfine process mediated by another detuned triplet state. We show that the quantum interference between the first-order and the second-order hyperfine processes leads to

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a feedback mechanism for manipulating the nuclear field components. The theory provides a unified framework for realizing negative- and positive-feedback controls by bringing the system through the singlet-triplet resonances. A non-intuitive prediction is that negative and positive feedbacks can be realized not only for longitudinal nuclear field gradients, but also for the nuclear field components transverse to the external field while nuclear spins are dynamically polarized in the longitudinal direction. This has not been possible in other feedback mechanisms being explored [22,29–31]. We discuss the possibility of enhancing the dephasing time of each individual spin in a cluster of dots using this negative-feedback control on the transverse nuclear field.

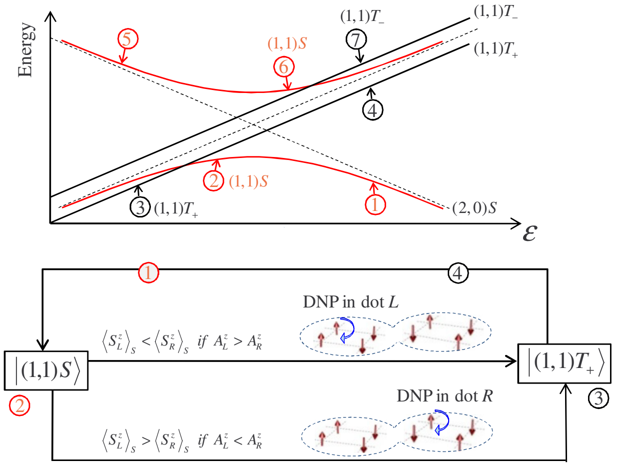

Fig. 1: (Colour on-line) Top: energy level scheme of the double-dot system in an external magnetic field in the z-direction. Bottom: feedback loop in the DNP cycle passing through the S–T_+ resonance. ε, the detuning between the $(2,0)$ and $(1,1)$ charge configurations, is gate controlled. The anti-crossing of the $(2,0)S$ and $(1,1)S$ is due to inter-dot tunneling which can be independently controlled. We use circled numbers to denote the two-electron spin states at different values of ε. ①→②→③→④→① form a DNP control loop utilizing the S–T_+ degeneracy. The loop consists of: the adiabatic evolution from $(2,0)S$ to $(1,1)S$ (①→②); the hyperfine-driven $S \to T_+$ transition which realizes the feedback on the nuclear field (②→③); the fast tuning of ε while the system remains in the $(1,1)T_+$ state (③→④); re-initialization to the $(2,0)S$ by relaxation (④→①). The fluctuation in the nuclear field gradient $A_L^z - A_R^z$ is suppressed (magnified) if the hyperfine constant $\mathcal{A}$ is positive (negative). (①→⑤→⑥→⑦→①) is a similar DNP control loop utilizing the S–T_- degeneracy. (①→⑤) is realized by fast tuning of ε with inter-dot tunneling switched off.

Electron-nuclear spin dynamics near the singlet-triplet resonances. – We first briefly explain the energy level scheme which is controlled by voltages applied to electrostatic gates in the double-dot system, as demonstrated in a number of recent experiments [12,27,32,33]. A schematic energy level diagram with (n,m) indicating the charge occupancies of the left and right dots is shown in fig. 1. A total of five two-electron states are involved: the singlet and triplet states of the $(1,1)$ charge configuration, and the singlet state of the $(2,0)$ charge configuration. All other states are well detuned and can be neglected. The detuning ε between the $(2,0)$ and $(1,1)$ charge configurations is gate controlled. The three triplet states (T_-, T_0, T_+) of the $(1,1)$ configuration are split in an external magnetic field along the z-direction. In the vicinity of the $(2,0)$–$(1,1)$ charge degeneracy, inter-dot tunneling causes the anti-crossing of the two singlet states $(2,0)S$ and $(1,1)S$. Away from the $(2,0)$–$(1,1)$ charge degeneracy point, the residual inter-dot tunneling results in an exchange splitting between $(1,1)S$ and $(1,1)T_0$, which allows the $(1,1)S$ state to be degenerate with $(1,1)T_+$ or $(1,1)T_-$ at negative or positive ε. Dynamical nuclear spin polarization (DNP) is highly efficient at the S–T_+ and S–T_- degeneracies since energy conservation can be directly satisfied [32]. Various aspects of the DNP processes in such a system in different parameter regimes have been studied in a recent theoretical work [34]. Here we focus on the feedback effects upon the crossing of singlet-triplet resonances.

In the double-dot system, each electron is coupled to the nuclear spins in the same dot by the contact hyperfine interaction $\hat{H}_{e\text{-}n} = \sum_{i=L,R}[\hat{S}_i^z \hat{A}_i^z + \frac{1}{2}(\hat{S}_i^+ \hat{A}_i^- + \hat{S}_i^- \hat{A}_i^+)]$. Here L and R denote the left and the right dot, respectively. $\hat{A}_L^z \equiv \sum_{n\in L} a_n \hat{I}_n^z$ and $\hat{A}_L^\pm \equiv \sum_{n\in L} a_n \hat{I}_n^\pm$ are the longitudinal and transverse nuclear field operators in the left dot, and $\hat{A}_R^z$ and $\hat{A}_R^\pm$ are the corresponding ones in the right dot. When the detuning between $(1,1)S$ and $(1,1)T_+$ states are much smaller than the electron Zeeman energy $g\mu_B B$ in the external field, the off-resonance hyperfine coupling of $(1,1)S$ and $(1,1)T_+$ to the other two far-detuned states $(1,1)T_0$ and $(1,1)T_-$ can be eliminated by a standard canonical transformation

$$\hat{W} = \exp\left[\frac{\hat{A}_L^z - \hat{A}_R^z}{g\mu_B B}|T_0\rangle\langle S| + \frac{\hat{A}_L^+ - \hat{A}_R^+}{4\sqrt{2}g\mu_B B}|T_-\rangle\langle S| + \frac{\hat{A}_L^+ + \hat{A}_R^+}{2\sqrt{2}g\mu_B B}|T_0\rangle\langle T_+| - \text{H.c.}\right],$$

and the residual second-order terms in the transformed Hamiltonian $\hat{W}\hat{H}\hat{W}^{-1}$ become effective couplings within the $(1,1)S$–$(1,1)T_+$ subspace. For the above perturbation expansion to be valid, we require $g\mu_B B \gg A$ where $A \sim \sqrt{N}a$ is the characteristic magnitude of the nuclear hyperfine field. Here a is the typical coupling strength between the electron spin and one nuclear spin, and N is the number of nuclear spins in one dot. For gate-defined III-V quantum dots, $N \sim 10^7$ and $a \sim \frac{\mathcal{A}}{N} \sim 10^4$ s $^{-1}$, with $\mathcal{A} \sim 10^{11}$ s $^{-1}$ being the hyperfine constant of the material [12,32].

The transformed Hamiltonian in the $(1,1)S$–$(1,1)T_+$ subspace is $\hat{H}_{S-T_+} = E_{T_+}|T_+\rangle\langle T_+| + E_S|S\rangle\langle S| + |T_+\rangle\langle S|\hat{D}_{(S-T_+)} + |S\rangle\langle T_+|\hat{D}^\dagger_{(S-T_+)} + |T_+\rangle\langle T_+|\hat{F}_{T_+} + |S\rangle\langle S|\hat{F}_S$. E_{T_+} and E_S are the energies of the singlet and triplet states determined by the electrostatic gates and the magnetic field (see fig. 1). $\hat{D}_{(S-T_+)}$, $\hat{F}_{T_+}$ and $\hat{F}_S$ are operators that acts on the nuclear spin bath

$$\hat{D}_{(S-T_+)} \equiv -\frac{1}{2\sqrt{2}}\left[(\hat{A}_L^- - \hat{A}_R^-) + \frac{(\hat{A}_L^- + \hat{A}_R^-)(\hat{A}_L^z - \hat{A}_R^z)}{2g\mu_B B}\right], \tag{1}$$

$$\hat{F}_{T_+} \equiv -\frac{(\hat{A}_L^- + \hat{A}_R^-)(\hat{A}_L^+ + \hat{A}_R^+)}{8g\mu_B B}, \tag{2}$$

$$\hat{F}_S \equiv -\frac{(\hat{A}_L^z - \hat{A}_R^z)^2}{4g\mu_B B} - \frac{(\hat{A}_L^- - \hat{A}_R^-)(\hat{A}_L^+ - \hat{A}_R^+)}{8g\mu_B B}. \tag{3}$$

Consider the electron spin initially on $|(1,1)S\rangle$ and the nuclear spin bath on an arbitrary state $|J\rangle$. Near the $(1,1)S$–$(1,1)T_+$ resonance, $\hat{H}_{S-T_+}$ causes transitions from the initial state $|(1,1)S\rangle \otimes |J\rangle$ to the final states $|(1,1)T_+\rangle \otimes \hat{D}_{(S-T_+)}|J\rangle$ and $|(1,1)S\rangle \otimes \hat{F}_S|J\rangle$. The former corresponds to the hyperfine-driven $(1,1)S \rightarrow (1,1)T_+$ transition accompanied by the simultaneous flip of a nuclear spin, and the latter corresponds to the hyperfine mediated inter-dot and intra-dot nuclear spin pair-flip while the electron spin state remains unchanged [21,35]. For typical nuclear state $|J\rangle$, the magnitude of the transition matrix elements are: $\sqrt{\langle J|\hat{D}^\dagger_{(S-T_+)}\hat{D}_{(S-T_+)}|J\rangle} \sim \sqrt{N}a$, and $\sqrt{\langle J|\hat{F}_S^\dagger \hat{F}_S|J\rangle} \sim \frac{Na^2}{g\mu_B B}$. For $g\mu_B B \gg \sqrt{N}a$, we have $\sqrt{\langle J|\hat{D}^\dagger_{(S-T_+)}\hat{D}_{(S-T_+)}|J\rangle} \gg \sqrt{\langle J|\hat{F}_S^\dagger \hat{F}_S|J\rangle}$, so the dominant process is the hyperfine-driven $(1,1)S \rightarrow (1,1)T_+$ transition.

One can similarly derive the effective Hamiltonian in the $(1,1)S$–$(1,1)T_-$ subspace when these two states are near resonance: $\hat{H}_{S-T_-} = E_{T_-}|T_-\rangle\langle T_-| + E_S|S\rangle\langle S| + |T_-\rangle\langle S|\,\hat{D}_{(S-T_-)} + |S\rangle\langle T_-|\,\hat{D}^\dagger_{(S-T_-)} + |T_-\rangle\langle T_-|\hat{F}_{T_-} + |S\rangle\langle S|\hat{F}_S$, where $\hat{D}_{(S-T_-)} \equiv \frac{1}{2\sqrt{2}}[(\hat{A}_L^+ - \hat{A}_R^+) + \frac{(\hat{A}_L^+ + \hat{A}_R^+)(\hat{A}_L^z - \hat{A}_R^z)}{2g\mu_B B}]$determines the back-action on the nuclear spin bath when the hyperfine-driven $(1,1)S \rightarrow (1,1)T_-$ transition occurs at the $(1,1)S$–$(1,1)T_-$ degeneracy.

Feedback controls of nuclear field fluctuations. – Upon the hyperfine-driven $(1,1)S \rightarrow (1,1)T_+$ transition, the effect of $\hat{D}_{(S-T_+)}$ is the flip down of a nuclear spin in the left or right dot, which polarizes the nuclear spin bath in the direction opposite to the external field. The interesting phenomenon comes from the interference of the leading-order term with the second-order term in $\hat{D}_{(S-T_+)}$. Consider an initial nuclear state $|A_L^z, A_R^z\rangle$ (an eigenstate of $\hat{A}_L^z$ and $\hat{A}_R^z$ with eigenvalues A_L^z and A_R^z, respectively), $\hat{D}_{(S-T_+)}$ brings it to the final state

$$\left[\left(1 + \frac{A_L^z - A_R^z}{2g\mu_B B}\right)\hat{A}_L^- - \left(1 - \frac{A_L^z - A_R^z}{2g\mu_B B}\right)\hat{A}_R^-\right]|A_L^z, A_R^z\rangle. \tag{4}$$

Namely, there is a larger probability for the flip-down of a nuclear spin to occur in left (right) dot if $A_L^z - A_R^z$ in the initial state is positive (negative). In either case, the magnitude of the nuclear field gradient, is reduced. This is a negative feedback which will reduce the magnitude of $A_L^z - A_R^z$.

For an ensemble of identical systems or the time ensemble averaged dynamics of a single system [12], we shall consider the ensemble average over evolutions initiated on various possible nuclear states. $\langle(\hat{A}_L^z - \hat{A}_R^z)^2\rangle$ gives the fluctuation of the nuclear field gradient in such ensemble dynamics[1], where $\langle\cdots\rangle$ stands for the quantum mechanical expectation value averaged over an ensemble of nuclear wavefunctions. We use $\delta\langle(\hat{A}_L^z - \hat{A}_R^z)^2\rangle$ to denote the change of this fluctuation when a hyperfine-driven $S \rightarrow T_+$ transition has occurred. Equation (4) then leads to

$$\delta\langle(\hat{A}_L^z - \hat{A}_R^z)^2\rangle = \frac{-2a}{|g\mu_B B|}\langle(\hat{A}_L^z - \hat{A}_R^z)^2\rangle + a^2. \tag{5}$$

It is clear that the fluctuation $\langle(\hat{A}_L^z - \hat{A}_R^z)^2\rangle$ gets suppressed in the DNP cycle if $g\mu_B B < \frac{2}{a}\langle(\hat{A}_L^z - \hat{A}_R^z)^2\rangle$. For a thermalized nuclear spin bath, we note that $\frac{1}{a}\langle(\hat{A}_L^z - \hat{A}_R^z)^2\rangle \sim \mathcal{A}$, corresponding to a magnetic field of $\sim 5\,\mathrm{T}$ [32]. The feedback effect is inversely proportional to the magnetic field. In the meantime, the perturbation treatment requires the magnetic field to be much larger than the nuclear field gradient which is of the typical value $\sim 2\,\mathrm{mT}$ for a thermalized nuclear spin bath. Thus, we expect the feedback effects to be pronounced in a moderate magnetic field of 10–100 mT.

A semiclassical picture helps to understand the underlying physics behind this feedback. The nuclear field gradient perturbs the electron spin eigenstates in the uniform external field, which results in an electron spin polarization gradient in the perturbed $(1,1)S$ state. The electron spin polarization gradient in turn determines the back-action to the nuclear spin bath upon the electron-nuclear flip-flop, which completes the feedback loop for manipulating the nuclear field gradient. One can further anticipate a similar feedback effect on the transverse nuclear field as well in the DNP process. In the presence of a transverse nuclear field, the total effective magnetic field for the electron is tilted from the z-direction, so the electron spin polarization in the perturbed $(1,1)T_+$ state has a transverse component. Upon the hyperfine-driven $S \rightarrow T_+$ transition, nuclear spins are then polarized along the axis of the total magnetic field, which changes the transverse nuclear field value.

To derive the effect on the transverse nuclear field, *e.g.* along the x-direction, it is convenient to use the x basis

[1]Note that $\langle\hat{A}_L^z - \hat{A}_R^z\rangle$ and $\langle\hat{\boldsymbol{A}}_L^\perp + \hat{\boldsymbol{A}}_R^\perp\rangle$ are always zero under the controls being discussed.

for the nuclear state and we rewrite eq. (1),

$$\hat{D}_{(S-T_+)} = \frac{1}{2\sqrt{2}}\Bigg[\hat{A}_L^x - \hat{A}_R^x - \frac{\left(\hat{A}_L^{x+} + \hat{A}_L^{x-} + \hat{A}_R^{x+} + \hat{A}_R^{x-}\right)\left(\hat{A}_L^{x+} - \hat{A}_L^{x-} - \hat{A}_R^{x+} + \hat{A}_R^{x-}\right)}{8g\mu_B B} + \left(1 + \frac{\hat{A}_L^x + \hat{A}_R^x}{2g\mu_B B}\right)\frac{\hat{A}_L^{x+} - \hat{A}_R^{x+}}{2\mathrm{i}} + \left(1 - \frac{\hat{A}_L^x + \hat{A}_R^x}{2g\mu_B B}\right)\frac{\hat{A}_L^{x-} - \hat{A}_R^{x-}}{2\mathrm{i}}\Bigg]. \tag{6}$$

Consider an initial nuclear state $|A_L^x, A_R^x\rangle$, an eigenstate of $\hat{A}_L^x$ and $\hat{A}_R^x$ with eigenvalues A_L^x and A_R^x, respectively, $\hat{D}_{(S-T_+)}$ brings it to the final state

$$\hat{D}_{(S-T_+)}|A_L^x, A_R^x\rangle = \frac{1}{2\sqrt{2}}\Bigg[(A_L^x - A_R^x)\,|A_L^x, A_R^x\rangle + \left(1 + \frac{A_L^x + A_R^x + a}{2g\mu_B B}\right)\frac{\lambda|A_L^x + a, A_R^x\rangle - \lambda|A_L^x, A_R^x + a\rangle}{2\mathrm{i}} + \left(1 - \frac{A_L^x + A_R^x - a}{2g\mu_B B}\right)\frac{\lambda|A_L^x - a, A_R^x\rangle - \lambda|A_L^x, A_R^x - a\rangle}{2\mathrm{i}}\Bigg], \tag{7}$$

where $\lambda \equiv \sqrt{\langle A_L^x, A_R^x|\hat{A}_L^{x-}\hat{A}_L^{x+}|A_L^x, A_R^x\rangle} \sim \sqrt{N}a$ and $|A_L^x - A_R^x| \sim \sqrt{N}a$. Here we have neglected terms such as $|A_L^x \pm a, A_R^x \pm a\rangle$ whose probability in the order of $(\frac{\sqrt{N}a}{g\mu_B B})^2$.

A measure of $\hat{A}_L^x + \hat{A}_R^x$ in the final state has three possible values: $A_L^x + A_R^x$, $A_L^x + A_R^x + a$ and $A_L^x + A_R^x - a$, with the probabilities of

$$\frac{(A_L^x - A_R^x)^2}{(A_L^x - A_R^x)^2 + \lambda^2}, \quad \frac{\lambda^2/2}{(A_L^x - A_R^x)^2 + \lambda^2}\left(1 + \frac{A_L^x + A_R^x}{g\mu_B B}\right), \quad \frac{\lambda^2/2}{(A_L^x - A_R^x)^2 + \lambda^2}\left(1 - \frac{A_L^x + A_R^x}{g\mu_B B}\right),$$

respectively. If $A_L^x + A_R^x$ is initially positive, the probability for this transverse nuclear field to increase is larger than the probability for it to decrease. Namely, there is a positive feedback to increase the magnitude of the total transverse field. For the y component of the nuclear field, we have the same result. Upon a hyperfine-driven $S \to T_+$ transition, the change in the fluctuation of transverse nuclear field $\hat{\boldsymbol{A}}^\perp \equiv \hat{A}^x\boldsymbol{i} + \hat{A}^y\boldsymbol{j}$ for a nuclear spin ensemble is given by (see footnote [1])

$$\delta\langle(\hat{\boldsymbol{A}}_L^\perp + \hat{\boldsymbol{A}}_R^\perp)^2\rangle = \alpha\frac{a}{|g\mu_B B|}\langle(\hat{\boldsymbol{A}}_L^\perp + \hat{\boldsymbol{A}}_R^\perp)^2\rangle + \beta a^2, \tag{8}$$

where α and β are both positive factors of the magnitude $\sim O(1)$. Here we note that the Larmor precession of nuclear spins in a moderate magnetic field of 10 mT is in the order of ~ 0.1 MHz, which is much smaller as compared to the magnitude of the transition matrix element for the electron-nuclear flip-flop $\sim \sqrt{N}a \sim 10$ MHz. Thus, the effect of nuclear spin Larmor precession on the hyperfine-driven $S \to T_+$ transition can be well neglected. Uniform precession of nuclear spins during other slower parts of the control cycle does not change the value of $\langle(\hat{\boldsymbol{A}}_L^\perp + \hat{\boldsymbol{A}}_R^\perp)^2\rangle$.

From eqs. (8) and (5), it is obvious that the sign of the feedback depends on the sign of the hyperfine constant a. In III–V materials, as the hyperfine constants for the isotopes of Ga, In, Al and As are all positive [36], the hyperfine-driven $S \to T_+$ transition results in a negative feedback to suppress fluctuation in the gradient of longitudinal nuclear field and a positive feedback to increase the fluctuation of the transverse nuclear field.

By similar analysis, we find that the $S \to T_-$ transition has the opposite effects, *i.e.* a positive feedback to increase fluctuation in the gradient of longitudinal nuclear field and a negative feedback to suppress the fluctuation of the transverse nuclear field.

For Ge/Si double quantum dot [37,38], since ^{29}Si and ^{73}Ge both have negative hyperfine constant [39], the same control shall result in feedbacks opposite to those in the III–V materials.

Below, we take parameters from realistic experimental systems and evaluate the efficiency of this feedback control. In the experiment by Reilly *et al.* [27], the double dot is initialized on the $(2,0)S$ state when ε is large and positive (see fig. 1). By tuning ε to negative value via gate control, the two-electron wave function can be adiabatically evolved to the $(1,1)S$ state. As the evolution is slowly passed through the S–T_+ degeneracy, the hyperfine-driven $S \to T_+$ transition is expected to occur with a simultaneous flip-down of one nuclear spin. The gate control can then be rapidly brought back to the large and positive ε, waiting for the double dot to be initialized again to the $(2,0)S$ state. By repeating the above cycle at a rate of 4 MHz in an external field $B_0 = 10$ mT, nuclear spins are expected to be dynamically polarized in the direction longitudinal to the external field. The experiment is performed at a temperature ($\sim$100 mK) many orders larger than the nuclear Zeeman energy ($\sim$0.01 mK), so the initial nuclear spin bath under thermal equilibrium has equal probability on every possible spin configuration. Before the DNP pump is applied, the observed ensemble dephasing time $T_2^* \sim 15$ ns between the spin states $|\uparrow\rangle_L|\downarrow\rangle_R$ and $|\downarrow\rangle_L|\uparrow\rangle_R$ is in agreement with the calculated fluctuation $\langle(\hat{A}_L^z - \hat{A}_R^z)^2\rangle$ for this thermal nuclear distribution [27]. T_2^* is found to be enhanced by the cyclic DNP pumping. After a pump time $\sim$1 s, T_2^* saturates at $\sim$1 μs, which indicates that $\sqrt{\langle(\hat{A}_L^z - \hat{A}_R^z)^2\rangle}$ has been suppressed by a factor of $\sim$70 [27]. The measurement also shows that this cyclic pumping establishes a steady-state nuclear spin polarization of $\sim$1%.

We use Γ to denote the DNP rate, *i.e.* the number of hyperfine-driven $S \to T_+$ transition per unit time. Equation (5) leads to the equation of motion

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle(\hat{A}_L^z - \hat{A}_R^z)^2\rangle = \frac{-2a\Gamma}{|g\mu_B B|}\langle(\hat{A}_L^z - \hat{A}_R^z)^2\rangle + a^2\Gamma, \tag{9}$$

which states that $\sqrt{\langle(\hat{A}_L^z - \hat{A}_R^z)^2\rangle}$ decreases in an exponential form: $\exp(-\frac{a}{g\mu_B B}\Gamma t)$. According to this exponential behavior, with a DNP rate of $\Gamma \sim 4\,\mathrm{MHz}$, suppression of the fluctuation by a factor of 100 can be achieved in a timescale of $\sim 0.1\,\mathrm{s}$. To achieve this degree of suppression, the total number of nuclear spins flipped down by the DNP cycles shall be $N\frac{g\mu_B B}{A}\ln(100) \sim 0.01\,N$, corresponding to a nuclear polarization of $\sim 1\%$. The steady-state value of the fluctuation is reached when the two terms on the RHS of eq. (9) cancel. So this residual fluctuation can be calculated $\sqrt{\langle(\hat{A}_L^z - \hat{A}_R^z)^2\rangle} = \sqrt{ag\mu_B B} \approx 10^6\,\mathrm{s}^{-1}$, corresponding to a dephasing time $T_2^* \sim \mu\mathrm{s}$.

By preparing the nuclear spin environment with the above cyclic negative-feedback control, the longitudinal nuclear fields in the two dots are set to the same value regardless of the initial state. The coherence of the physical qubit represented by the two-spin states is therefore protected from the statistical fluctuation of the nuclear field in each individual dot. Since nuclear spin relaxation is extremely slow, the resultant enhancement on the T_2^* time can last for seconds or even longer. It will be desirable to generalize this preparation scheme for single spin in individual dots.

Here we argue that the negative feedback on the average transverse nuclear field in the hyperfine-driven $S \to T_-$ transition may potentially be utilized to enhance the dephasing time of each single spin in a cluster of dots. As an example, we consider a three-phase control cycle in a three-dot system. In phase 1 of the cycle, dot 1 and dot 2 are coupled and both are decoupled from dot 3. The two-electron state in dot 1 and 2 is evolved in the sequence (① → ⑤ → ⑥ → ⑦ → ①) (see fig. 1) to realize the hyperfine-driven $S \to T_-$ transition. This evolution can be realized by an experimental control similar to the one used in ref. [27]. The loop consists of four parts: i) tuning ε with inter-dot tunneling switched off (① → ⑤); ii) the adiabatic evolution from $(2,0)S$ to $(1,1)S$ (⑤ → ⑥); iii) the hyperfine-driven $S \to T_-$ transition (⑥ → ⑦); iv) re-initialization to the $(2,0)S$ by relaxation (⑦ → ①). In phase 2 (phase 3) of the cycle, we repeat the same control for dot 2 and 3 (dot 3 and 1). The fluctuations in $\hat{\boldsymbol{A}}_1^\perp + \hat{\boldsymbol{A}}_2^\perp$, $\hat{\boldsymbol{A}}_2^\perp + \hat{\boldsymbol{A}}_3^\perp$ and $\hat{\boldsymbol{A}}_1^\perp + \hat{\boldsymbol{A}}_3^\perp$ are all reduced in this three-phase control cycle. Since these three linearly independent vectors remain uncorrelated throughout the control, we have

$$\langle(\hat{\boldsymbol{A}}_1^\perp)^2\rangle = \frac{1}{4}\langle(\hat{\boldsymbol{A}}_1^\perp + \hat{\boldsymbol{A}}_2^\perp)^2\rangle + \frac{1}{4}\langle(\hat{\boldsymbol{A}}_2^\perp + \hat{\boldsymbol{A}}_3^\perp)^2\rangle + \frac{1}{4}\langle(\hat{\boldsymbol{A}}_1^\perp + \hat{\boldsymbol{A}}_3^\perp)^2\rangle.$$

Therefore, the fluctuation of the transverse nuclear field in each individual dots gets reduced in this three-phase control cycle. After the preparation of nuclear spin bath with the above cycles, if the external magnetic field is rotated to one of the transverse direction, an enhanced single-spin dephasing time T_2^* in the new external field is expected.

For the qubit represented by the two-spin states $|\uparrow\rangle_L|\downarrow\rangle_R$ and $|\downarrow\rangle_L|\uparrow\rangle_R$, the gradient in the longitudinal nuclear field can realize a rotation about the z-axis. Positive feedback which increases the magnitude of the nuclear field gradient is also of interest since a larger gradient means a faster gate operation. A recent experiment has reported universal quantum control of the two-spin qubit in a coupled double dot where the nuclear field gradient is magnified by nuclear state preparation [40]. The experiment uses two types of controls which realize the $S \to T_+$ transition and the $T_+ \to S$ transition, respectively, in an external field of $\sim 1\,\mathrm{T}$. Such a large external field will deactivate the negative feedback in the $S \to T_+$ transition. For the $T_+ \to S$ transition, the back-action on the nuclear state is described by the operator $\hat{D}^\dagger_{(S-T_+)}$ which does result in a positive feedback on the longitudinal nuclear field gradient. However, the large external field will make it difficult to initiate the magnification by the positive feedback. The validity of our theory is likely to be restricted in moderate external field for both negative and positive feedbacks, and the mechanism for the magnification of the nuclear field gradient in this experiment is still unclear.

Summary. – For a coupled double-dot system in moderate external magnetic field, we have shown that the interference of first-order and second-order hyperfine processes results in a feedback mechanism for manipulating the nuclear hyperfine fields when the two-electron singlet state is brought into resonance with one triplet state. In principle, the feedback controls here do not need explicit measurement steps. Re-initialization of the control loop is simply realized by the relaxation to the ground state $(2,0)S$ at large positive ε, which can be ensured after sufficiently long waiting time. The non-determinacy lies in the electron-nuclear flip-flop at the singlet-triplet resonance, but whether it has occurred or not, the double dot will return to the initial state $(2,0)S$ at the end of a control cycle. The probability for the electron-nuclear flip-flop in a single cycle corresponds to the ratio between the actual DNP rate Γ and the repetition rate of the control cycles.

For negative-feedback controls aimed at suppressing the nuclear field fluctuations, the spectral diffusion of nuclear hyperfine fields caused by various processes [35,41–45] could be a competing mechanism in determining the residual fluctuations in the steady state. This element has not been included in the present discussion. Further studies are needed to investigate the quantitative effects of spectral diffusions.

* * *

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