Anomalous Decoherence Effect in a Quantum Bath

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Decoherence of quantum objects in noisy environments is important in quantum sciences and technologies. It is generally believed that different processes coupled to the same noise source have similar decoherence behaviors and stronger noises cause faster decoherence. Here we show that in a quantum bath, the case can be the opposite. We predict that the multitransition of a nitrogen-vacancy center spin-1 in diamond can have longer coherence time than the single transitions, even though the former suffers twice stronger noises from the nuclear spin bath than the latter. This anomalous decoherence effect is due to manipulation of the bath evolution via flips of the center spin.

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Decoherence of quantum objects in noisy environments is of paramount importance in quantum sciences and technologies [1–5]. In particular, decoherence of electron spins coupled to nuclear spin baths in quantum dots [6–12] or solid-state impurity centers [13–16] is a key issue in spin-based quantum information processing [2], magnetic resonance spectroscopy [13,16,17], and magnetometry [3–5]. The seminal spectral-diffusion theories [18,19] treat couplings to the environments as classical noises. It is generally believed that different processes coupled to the same noise source have similar decoherence behaviors and stronger noises cause faster decoherence [18,19].

In modern quantum technologies, however, the relevant environments are of nanometer size [6–17] and therefore their quantum nature becomes important. Quantum theories developed in recent years [20–22] suggest that a quantum nuclear spin bath, in contrast to classical noises, possesses a great extent of controllability and has surprising coherence recovery effects on an electron spin embedded in it [23]. The nuclear spin bath may also be exploited in quantum technologies such as information storage [24].

In this Letter, we report an anomalous decoherence effect of a spin higher than 1/2 in a nuclear spin bath. We consider the multitransition and single transitions of the spin-1 of a nitrogen-vacancy (NV) center in diamond [Fig. 1(a)], which are coupled to the same nuclear spin bath [Fig. 1(b)]. Surprisingly, under the dynamical decoupling control [25], the multitransition can have longer coherence time than the single transitions, even though the former is subjected to stronger noises. This anomalous effect is due to manipulation of the bath evolution via the center spin flips.

The spin decoherence of an NV center in high-purity diamond is mainly caused by hyperfine coupling to ^{13}C nuclear spins [14,15,17]. The NV center has a spin-1, with three eigenstates $|0\rangle$ and $|\pm\rangle$ quantized along the NV (z) axis at zero field. The single transitions $|0\rangle \leftrightarrow |\pm\rangle$ and the multitransition $|+\rangle \leftrightarrow |-\rangle$ [Fig. 1(a)] are subjected to noises from the same nuclear spin bath, with the noise amplitude for the latter being twice that for the former.

In the semiclassical description, the effect on the center spin of the environment is a fluctuating local field $\mathbf{b}(t)$, with a Hamiltonian $H = S_z b_z(t)$, where S_z has eigenstates

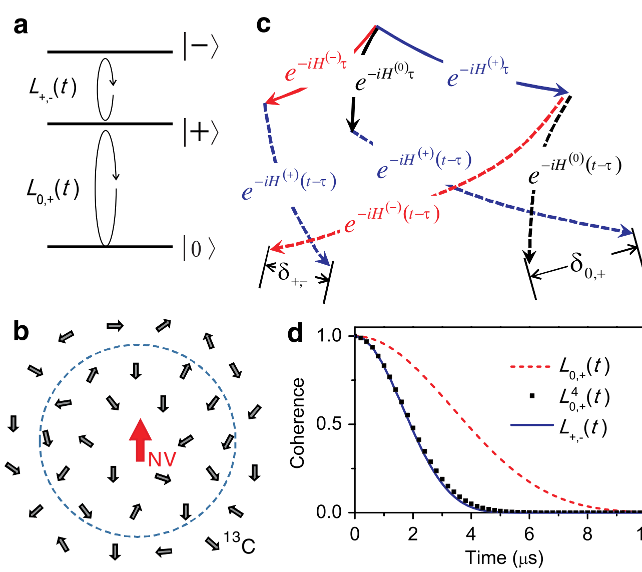

FIG. 1 (color online). (a) The single-transition coherence $L_{0,+}(t)$ and the multitransition coherence $L_{+,-}(t)$ of an NV center spin. (b) Schematic of an NV center spin coupled to a ^{13}C nuclear spin bath (enclosed by the circle). (c) Pronged quantum evolution pathways of the nuclear spin bath conditioned on the center spin states. Under a flip of the center spin, the bath evolution directions are switched (from solid to dashed curves). The distance $\delta_{\alpha,\beta}$ (distinguishability) between the pathways determines the center spin coherence $L_{\alpha,\beta}(t)$. (d) Free-induction decay of the center spin coherence $L_{0,+}(t)$ (red dashed line) and $L_{+,-}(t)$ (blue solid line) under a magnetic field $B = 0.3$ T along the NV axis. The scaled single-transition coherence $L^4_{0,+}(t)$ (black square symbols) is plotted for comparison.

$|0\rangle$ and $|\pm\rangle$ with eigenvalues 0 and ± 1, respectively. Here we consider only the fluctuations along the NV axis, since the perpendicular components are too weak to cause spin-flip relaxation. An initial state of the center spin $|\Psi(0)\rangle = a_-|-\rangle + a_0|0\rangle + a_+|+\rangle$ will evolve to $|\Psi(t)\rangle = a_- e^{i\varphi(t)}|-\rangle + a_0|0\rangle + a_+ e^{-i\varphi(t)}|+\rangle$, with an accumulated random phase $\varphi(t) = \int_0^t b_z(\tau)d\tau$. The coherence of the single transitions $|0\rangle \leftrightarrow |\pm\rangle$ is determined by the average of the random phase factor $L_{0,\pm} = \langle e^{\pm i\varphi(t)}\rangle$, while the multitransition coherence $L_{+,-} = \langle e^{2i\varphi(t)}\rangle$. For Gaussian noises as commonly encountered [18,19], $L_{0,\pm} = e^{-\langle\varphi(t)\varphi(t)\rangle/2}$ and $L_{+,-} = e^{-2\langle\varphi(t)\varphi(t)\rangle}$, which satisfy a simple scaling relation

$$|L_{+,-}| = |L_{0,\pm}|^4. \tag{1}$$

Decoherence of the multitransition behaves essentially the same as that of the single transitions, but is faster.

In the quantum description, the random field $\mathbf{b}$ is a quantum operator of the bath. The system Hamiltonian is

$$H = S_z b_z + H_B \equiv \sum_{\alpha=\pm,0} |\alpha\rangle\langle\alpha| \otimes H^{(\alpha)}, \tag{2}$$

where $b_z = \sum_j \mathbf{A}_j \cdot \mathbf{I}_j$ with $\mathbf{A}_j$ denoting the hyperfine coupling to the jth nuclear spin $\mathbf{I}_j$, and $H_B = \sum_{j<k} \mathbf{I}_j \cdot \mathbb{D}_{jk} \cdot \mathbf{I}_k + \omega_N \sum_j I_{j,z}$ contains dipolar coupling ($\mathbb{D}_{jk}$) between nuclear spins and the nuclear Zeeman splitting (ω_N) under the external field (along the z axis), and $H^{(\alpha)} \equiv \alpha b_z + H_B$. The electron spin splitting is dropped by working in the interaction picture. From an initial state $(a_-|-\rangle + a_0|0\rangle + a_+|+\rangle) \otimes |J\rangle$, the center spin and bath evolve as

$$\begin{aligned}|\Psi(t)\rangle = {} & a_-|-\rangle \otimes |J_-(t)\rangle + a_0|0\rangle \otimes |J_0(t)\rangle \\ & + a_+|+\rangle \otimes |J_+(t)\rangle,\end{aligned} \tag{3}$$

where $J_\alpha(t) \equiv \exp(-iH^{(\alpha)}t)|J\rangle$. The bath evolves along pronged pathways in the Hilbert space conditioned on the center spin state [Fig. 1(c)]. The center spin loses coherence as its which-way information is recorded in the bath [21]. The coherence of the transition $|\alpha\rangle \leftrightarrow |\beta\rangle$ is

$$\langle\Psi(t)|\alpha\rangle\langle\beta|\Psi(t)\rangle = a_\alpha^* a_\beta \langle J_\alpha(t)|J_\beta(t)\rangle \equiv a_\alpha^* a_\beta L_{\alpha,\beta}(t), \tag{4}$$

determined by the overlaps between the pronged bath states. Explicitly, the single-transition coherence $L_{0,\pm}(t) = \langle J_0(t)|J_\pm(t)\rangle$ and the multitransition coherence $L_{+,-}(t) = \langle J_+(t)|J_-(t)\rangle$. The bath evolutions for different center spin states can be substantially different. Thus, the single- and multitransitions may have different decoherence behaviors, and the scaling relation in Eq. (1) does not hold in general.

Even more interesting is the dynamical decoupling control of the center spin [26]. In the classical picture, if a transition of the center spin is flipped, the decoherence is controlled through modulation of the random phase as $\varphi(t) = \int_0^t b_z(\tau)F(\tau)d\tau$, where $F(\tau)$ jumps between $+1$ and -1 at every flip [27]. In the quantum picture, the bath evolution along different pathways is manipulated when the center spin is flipped between different states. For example, after a flip operation $|\alpha\rangle \leftrightarrow |\beta\rangle$ at time τ, the system evolves as $a_\beta|\alpha\rangle \otimes e^{-iH^{(\alpha)}(t-\tau)}e^{-iH^{(\beta)}\tau}|J\rangle + a_\alpha|\beta\rangle \otimes e^{-iH^{(\beta)}(t-\tau)}e^{-iH^{(\alpha)}\tau}|J\rangle$; i.e., the bath evolutions conditioned on the center spin state exchange their directions in the Hilbert space [Fig. 1(c)]. This results in decoherence control dramatically different from the case of classical noises.

The quantum description requires the definition of a relatively closed quantum system including the center spin and bath. In diamond, the dipolar hyperfine interaction decays inverse cubically with distance and the NV center spin is effectively coupled to hundreds of nuclear spins located within a few nanometers (the bath) [14,28]. The dipolar interaction between nuclear spins has strength about 10 Hz for two nuclei at average distance, which is much weaker than the hyperfine coupling ($\gtrsim$ kHz for nuclei within 4 nm). During the decoherence process, which occurs within milliseconds, negligible is the diffusion of quantum coherence from the bath to outside. Thus, the center spin and bath evolve as a relatively closed quantum system [Fig. 1(b)]. A counterexample is NV centers in nitrogen-rich samples where nitrogen electron spins form the baths [29–31]. In that case, the interaction between two bath spins at average distance is much stronger than the coupling between the center and a bath spin, and therefore the coherence diffusion in the environment is faster than the center spin decoherence, which invalidates the definition of a closed quantum bath. Instead, the classical noise theory well describes the nitrogen spin baths [29–31].

There are also thermal noises resulting from random orientations of the nuclear spins at finite temperature [32], which are of classical nature. Indeed, the thermal noises (also called inhomogeneous broadening) are much stronger than the quantum fluctuations. As shown in Fig. 1(d), the calculated free-induction decay of the single- and multitransition coherence, which is mainly caused by the inhomogeneous broadening, fits very well the scaling relation in Eq. (1). The inhomogeneous broadening effect can be totally removed by spin echo. Such coexistence of classical and quantum fluctuations, and their different effects in spin echo, can be used for *in situ* test of the semiclassical and quantum theories.

We calculate the coherence of an NV center electron spin coupled to a nuclear spin bath generated by randomly placing ^{13}C atoms on the diamond lattice with natural abundance 1.1%. Inclusion of about 500 ^{13}C nuclear spins within 4 nm from the NV center is sufficient for a converged result. For the decoherence control, we adopt the periodic dynamical decoupling (PDD) control by an equally spaced sequence (applied at $\tau, 3\tau, 5\tau \ldots$) [17,25,31].

The spin coherence is calculated with the cluster correlation expansion (CCE) [22]. The center spin decoherence caused by a particular nuclear spin cluster C is denoted as $L_{\alpha,\beta}^{(C)}$. The irreducible correlation of the cluster is recursively defined as $\tilde{L}_{\alpha,\beta}^{(C)} \equiv L_{\alpha,\beta}^{(C)}/\prod_{C'\subset C}\tilde{L}_{\alpha,\beta}^{(C')}$ which excludes the irreducible correlations of the subclusters C'. Then the Mth order CCE approximation (CCE-M) gives $L_{\alpha,\beta} \approx \prod_{|C|\leq M}\tilde{L}_{\alpha,\beta}^{(C)}$ with $|C|$ denoting the number of spins in the cluster. In this Letter, inclusion of up to 5-spin clusters (CCE-5) is sufficient to produce converged results.

Figures 2(a) and 2(b) show the convergence of the CCE for the single- and multitransition decoherence under a strong magnetic field and the 5-pulse PDD (PDD-5) control. Under the strong magnetic field ($\gg$ 100 Gauss), the nuclear Zeeman splitting is too large for the hyperfine interaction ($\omega_N \gg A_j$) to cause single nuclear spin flips, so the single-spin dynamics in the bath (CCE-1) contributes negligible decoherence. Actually, CCE-2 gives almost converged results. This means that the main mechanism of the decoherence is the nuclear spin pair correlations.

The nuclear spin pair dynamics is essentially the flip-flop between the two states $|\uparrow\downarrow\rangle$ and $|\downarrow\uparrow\rangle$ [see Fig. 2(c)]. The polarized states $|\uparrow\uparrow\rangle$ and $|\downarrow\downarrow\rangle$ are stationary, since the nuclear spin Zeeman energy is much greater than the dipolar interaction strength ($\omega_N \ggg |\mathbb{D}_{jk}|$). The dipolar interaction causes the transition $|\uparrow\downarrow\rangle \leftrightarrow |\downarrow\uparrow\rangle$ with a rate $X_{jk} \equiv \langle\downarrow\uparrow|\mathbf{I}_j\cdot\mathbb{D}_{jk}\cdot\mathbf{I}_k|\uparrow\downarrow\rangle$. The hyperfine interaction induces an energy cost of the flip-flop $Z_{jk}^{(\alpha)} = \alpha(\mathbf{A}_j - \mathbf{A}_k)\cdot\mathbf{e}_z$, for the electron spin state $|\alpha\rangle$. Thus the flip-flop is mapped [21] to the precession of a pseudospin $\boldsymbol{\sigma}_{jk}$ about a pseudofield $\mathbf{h}_{jk}^{(\alpha)} = (X_{jk}, 0, Z_{jk}^{(\alpha)})$ [see Fig. 2(d)], which is conditioned on the electron spin state $|\alpha\rangle$. The pronged bath evolution shown in Fig. 1(c), which causes the center spin decoherence, is reduced to pronged pseudospin precession. The center spin decoherence caused by pair flip-flops is factorized as

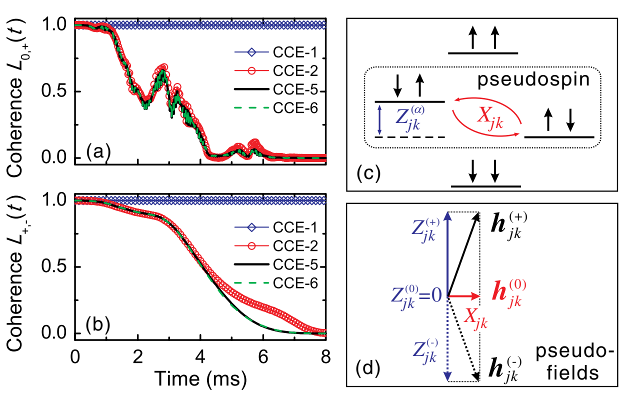

FIG. 2 (color online). (a) The NV center spin coherence $L_{0,+}(t)$ under PDD-5 control and magnetic field $B = 0.3$ T along the NV axis, calculated with different orders of CCE. (b) The same as (a), but for the multitransition coherence $L_{+,-}(t)$. (c) The pseudospin picture for the nuclear spin pair dynamics. (d) Schematic of the pseudofields $\mathbf{h}_{jk}^{(\alpha)}$ for different NV center spin states $|\alpha\rangle$.

$$L_{\alpha,\beta}(t) \approx \prod_{jk}|\langle\sigma_{jk}^{(\alpha)}(t)|\sigma_{jk}^{(\beta)}(t)\rangle|, \tag{5}$$

where $|\sigma_{jk}^{(\alpha/\beta)}(t)\rangle$ is the precession of the pseudospin about the pseudofield $\mathbf{h}_{jk}^{(\alpha/\beta)}$ for the center spin state $|\alpha/\beta\rangle$.

Figure 3(a) presents the main result of this Letter. Under the Hahn-echo (PDD-1) control, the inhomogeneous broadening effect is eliminated and the decoherence is determined by the quantum fluctuations resulting from the many-body interaction in the bath. The multitransition coherence decays faster than the single-transition coherence, but the simple scaling relation in Eq. (1) is violated. More surprisingly, when the number of control pulses is increased (from two- to five-pulse PDD control), the multitransition coherence even lasts longer than the single-transition coherence.

The anomalous decoherence effect, though counterintuitive, can be understood using the pseudospin picture as illustrated in Figs. 3(b) and 3(c). The decoherence is determined by the distance between the bifurcated pseudospin pathways. In the Hahn echo (PDD-1), the decoherence due to the pair flip-flops is [21,28]

$$L_{\alpha,\beta}(2\tau) = \prod_{jk}[1 - 2|\sin(\mathbf{h}_{jk}^{(\alpha)}\tau/2)\times\sin(\mathbf{h}_{jk}^{(\beta)}\tau/2)|^2]. \tag{6}$$

In the short time limit $h_{jk}^{(\alpha)}\tau \ll 1$, the multitransition coherence decays faster than the single-transition coherence

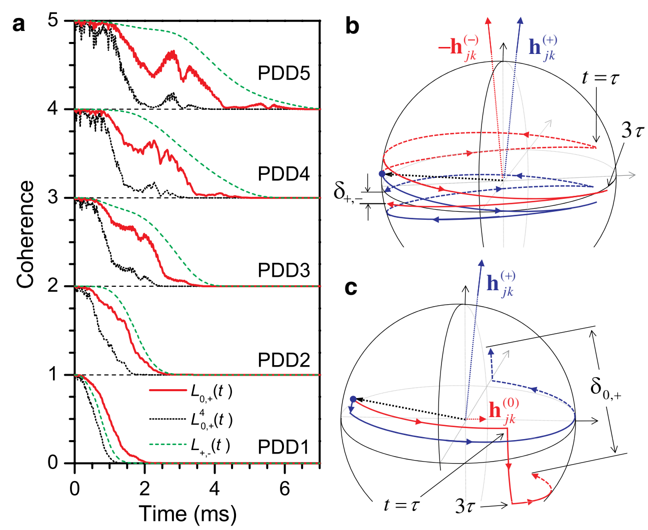

FIG. 3 (color online). (a) The single-transition coherence $L_{0,+}(t)$ (red solid lines), the multitransition coherence $L_{+,-}(t)$ (green dashed lines), and the scaled single-transition coherence $L_{0,+}^4(t)$ (black dotted lines), under a magnetic field $B = 0.3$ T along the NV axis and one- to five-pulse PDD control (PDD-1 to PDD-5, from bottom to top, vertically shifted for the sake of clarity). (b) The bifurcated pseudospin precession about the pseudofields $\mathbf{h}_{jk}^{(\pm)}$ for the multitransition coherence under the PDD-2 control, with the initial state indicated by a solid circle at the end of a dotted arrow. Upon the center spin flip (at $t = \tau$ or 3τ, the pseudospin alternates its pseudofield. (c) The same as (b), but for the single-transition coherence.

and the scaling relation in Eq. (1) is satisfied. As the time increases, however, the scaling relation is violated. For most nuclear spin pairs, the interaction is <100 Hz, while the hyperfine energy cost is $>$kHz. Therefore in the multitransition case, the two pseudofields $\mathbf{h}_{jk}^{(\pm)}$ corresponding to the electron spin sates $|\pm\rangle$ are nearly antiparallel. Thus the distance between the bifurcated pseudospin pathways [Fig. 3(b)] and hence the induced decoherence are small. While in the single-transition case, the two pseudofields $\mathbf{h}_{jk}^{(0)}$ and $\mathbf{h}_{jk}^{(+)}$ are in general not (anti-)parallel, and the bifurcated pseudospin pathways may deviate largely from each other [Fig. 3(c)], which induces strong decoherence. Such control effect on the bath dynamics becomes more significant when the coherence time is prolonged by dynamical decoupling.

The pseudospin picture also explains the oscillation features in the single-transition coherence and the smooth decoherence of the multitransition. A careful examination reveals that the rapid and shallow oscillations are induced by those pairs which have one ^{13}C located relatively close to the NV center. Such pairs have large hyperfine energy cost $Z_{jk}^{(\pm)}$ in the flip-flop. The large pseudofields cause rapid precession of the pseudospin when the center spin is in the states $|\pm\rangle$, but the pseudospin precession for the center spin state $|0\rangle$ is still slow. This induces rapid oscillations in the single-transition coherence. The slow and deep oscillations are caused by pairs which have two ^{13}C spins close to each other [5]. For the multitransition, however, the two pseudofields $\mathbf{h}_{jk}^{(\pm)}$ are nearly antiparallel, and the decoherence contributed by each individual pair is small, so the decoherence is smooth.

The higher-order cluster correlations will not affect the anomalous decoherence effect. In general, the effect should exist if the center-bath coupling is nonuniform and much stronger than the intrabath interaction. The center-bath coupling provides not only noises to the center spin but also back action to the bath [the bath Hamiltonian depends on the center spin state, as shown in Eq. (2)]. The elementary excitations in the bath will be suppressed by large energy cost due to the strong, nonuniform center-bath interaction, unless the center spin is in the unpolarized state $|0\rangle$. Thus, the effective dynamical fluctuations and hence the decoherence are relatively weak for the transitions not involving $|0\rangle$.

In conclusion, we have discovered that the multitransition and single-transitions of an NV center spin in diamond, though coupled to the same nuclear spin bath, have different decoherence features, and more strikingly, the former can have longer coherence time though it suffers stronger noises. This discovery establishes the controllability of quantum baths and paves the way for exploiting nuclear spin ensembles in solids for quantum information processing [1,2,24] and magnetometry [3–5].

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