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 should have similar decoherence behaviors and stronger noises would cause faster decoherence. Here we show that in a quantum bath, the case can be the opposite. In particular, we predict that the multi-transition 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.

In modern quantum technologies, however, the relevant environments are of nanometer size \cite{6,17} and therefore their quantum nature becomes important. Quantum theories developed in recent years \cite{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 \cite{23}. The nuclear spin bath may also be exploited in quantum technologies such as information storage \cite{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 multi-transition and single-transitions of the spin-1 of a nitrogen-vacancy (NV) center in diamond \cite{Fig. 1(a)}, which are coupled to the same nuclear spin bath \cite{Fig. 1(b)}. Surprisingly, under the dynamical decoupling control \cite{25}, the multi-transition can have longer coherence time than the single-transitions, even though the former is subjected to 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.

In the semiclassical description, the effect on the center spin of the environment is a fluctuating local field $b(t)$, with a Hamiltonian $H = S_z b(t)$, where $S_z$ has eigenstates $|0\rangle$ 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 $|\Psi(0)\rangle = a_{-}|\rangle + a_{0}|0\rangle + a_{+}|+\rangle$ undergoes transitions due to manipulation of the bath evolution via flips of the center spin.

The spin decoherence of an NV center in high-purity (type IIa) diamond is mainly caused by hyperfine coupling to $^{13}$C nuclear spins \cite{13,15,17}. The NV center has a spin-1, with three eigen states $|0\rangle$ and $|\pm\rangle$ quantized along the NV (z) axis at zero field. The single-transitions $|0\rangle \leftrightarrow |\rangle$ and the multi-transition $|+\rangle \leftrightarrow |\rangle$ \cite{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.

![FIG. 1: (Color online) (a) The single-transition coherence $L_{0,\alpha}(t)$ and the multi-transition coherence $L_{\alpha,\gamma}(t)$ 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. The distance $\delta_{\alpha,\beta}$ (distinguishability) between the pathways determines the center spin coherence $L_{\alpha,\beta}(t)$. Under a flip of the center spin, the bath evolution directions are switched (from solid to dashed curves). (d) Free-induction decay of the center spin coherence $L_{0,\gamma}(t)$ (red dashed line) and $L_{\alpha,\gamma}(t)$ (blue solid line) under a magnetic field $B = 0.3$ T along the NV axis. The scaled single-transition coherence $L_{0,\alpha}(t)$ (black square symbols) is plotted for comparison.](image-url)
will evolve to $|\Psi(t)\rangle = a_-|\psi(0)\rangle + a_0|0\rangle + a_+e^{-i\phi(t)}|+\rangle$, with an accumulated random phase $\phi(t) = \int_0^t b_\alpha(\tau)d\tau$. The coherence of the single-transitions $|0\rangle \leftrightarrow |\pm\rangle$ is determined by the average of the random phase factor $L_{0,\pm} = \langle e^{i\phi(0)} \rangle$, while the multi-transition coherence $L_{\pm,\pm} = \langle e^{i\phi(t)} \rangle$. For Gaussian noises as commonly encountered $[18]$ $L_{0,\pm} = e^{-i\phi(0)}/2$ and $L_{\pm,\pm} = e^{-i\phi(t)}|0\rangle\rangle$, which satisfy a simple scaling relation

$$|L_{\pm,\pm}| = |L_{0,\pm}|^4.$$  

(1)

Decoherence of the multi-transition behaves essentially the same as that of the single transitions, but is faster since the multi-transition experiences as twice strong noises as the single-transitions.

In the quantum description, the random field $b_\alpha$ is a quantum operator of the bath. The bath itself has internal interaction $H_B$. The center spin decoherence is indeed caused by the entanglement with the bath during the quantum evolution $[24]$. From the initial state $|\Psi(0)\rangle = (a_-|0\rangle + a_0|0\rangle + a_+|+\rangle) \otimes |J\rangle$, the center spin and bath evolve as

$$|\Psi(t)\rangle = a_-|\psi(t)\rangle \otimes |J(t)\rangle + a_0|0\rangle \otimes |J(t)\rangle + a_+|+\rangle \otimes |J(t)\rangle,$$  

(2)

where $J_\alpha(t) = \exp\left(-iH_\alpha^0t\right)|J\rangle$ with $H_\alpha^0 = \alpha b_\alpha + H_B$ for $\alpha = 0$ or $\pm$. The bath evolves along protracted pathways in the Hilbert space conditioned on the center spin state $[Fig. 1(c)]$. The center spin loses its coherence as its which-way information is recorded in the bath. The coherence of the single- and multi-transitions are determined by the overlaps between the protracted bath states as $L_{0,\pm}(t) = \langle J_0(t)|J_\pm(t)\rangle$ and $L_{\pm,\pm}(t) = \langle J_\pm(t)|J_\pm(t)\rangle$, respectively. The bath evolution can be substantially different for different center spin states. Thus, the multi-transition may have different decoherence behavior from what the single transitions do, and in particular, the scaling relation in Eq. (1) does not hold in general.

A more striking difference between classical noises and quantum baths occurs when the center spin is under the dynamical decoupling control. In the case of classical noises, if the center spin is under stroboscopic flips between different states, the decoherence is controlled through modulation of the accumulated random phase as $\phi(t) = \int_0^tb_\alpha(\tau)d\tau$, where $F(\tau)$ jumps between $+1$ and $-1$ every time the center spin is flipped $[26]$. In the case of quantum baths, 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 $\tau$, the electron-nuclear spin system evolves as $a_0|\alpha\rangle \otimes e^{-iH_\alpha^0(\tau-\tau)}e^{-iH_\alpha^0\tau}|J\rangle + a_0|\beta\rangle \otimes e^{-iH_\alpha^0(\tau-\tau)}e^{-iH_\alpha^0\tau}|J\rangle$, 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.

In our specific system, the random field results from the hyperfine coupling to $^{13}$C spins, $b_\alpha = \sum_j A_j \cdot J_j$, where $A_j$ is the coupling coefficients for the $j$th nuclear spin $J_j$. The dipolar hyperfine interaction decays inverse cubically with distance and the center spin is effectively coupled to hundreds of nuclear spins located within a few nanometers (the bath) $[14] [27]$. The nuclear spins have dipolar interaction $H_B = \sum_{i<j}J_{ij}^2\cdot D_{ij}^2\cdot \mathbf{I}_i$ with the coupling tensor $D_{ij}$ of strength about 10 Hz for two nuclei at average distance, which is much weaker than the hyperfine coupling ($\approx$ 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 $[Fig. 1(b)]$. An example on the contrary is an NV center in a nitrogen-rich sample where nitrogen electron spins form the bath $[28] [30]$. In that case, the interaction between two bath spins at average distance is much stronger than that between the center and a bath spin, and therefore the coherence diffusion in the environment is faster than the decoherence of the center spin, which invalidates the definition of a closed quantum bath. Instead, the classical noise theory well describes the coupling to the nitrogen spin bath $[28] [30]$. There are also thermal noises resulting from random orientations of the nuclear spins at finite temperature $[31]$, which are of classical nature. As shown in $[Fig. 1(d)]$, the calculated free-induction decay of the single- and multi-transition coherence, which is mainly caused by the thermal fluctuations (also called inhomogeneous broadening) of the $^{13}$C nuclear spin bath, is well fitted with Gaussian decays and satisfies the scaling relation in Eq. (1). This reflects the classical nature of thermal fluctuations. Indeed, the thermal fluctuations are much stronger than the quantum fluctuations, but 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 that is 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 $\tau$, $3\tau$, $5\tau$ . . . ) $[17] [25] [30]$. The spin coherence is calculated with the cluster correlation expansion (CCE) method $[22]$. The center spin decoherence caused by a particular nuclear spin cluster $C$ is denoted as $F_{\alpha\beta}^{(C)}$. The irreducible correlation of the cluster is recursively defined as $F_{\alpha\beta}^{(C)} = F_{\alpha\beta}^{(C)} \prod_{\alpha\beta' \notin C} F_{\alpha\beta'}^{(C)}$, which excludes the irreducible correlations of the sub-clusters $C_\alpha$. Then the $M$th order CCE approximation (CCE-M) gives $L_{\alpha\beta} \approx \prod_{\alpha\beta \subset M} F_{\alpha\beta}^{(C)}$, with $|C|$ denoting the number of spins in the cluster. In this paper, inclusion of up to 5-spin clusters (CCE-5) is sufficient to produce converged results.

Figure 2(a) and (b) show the convergence of the CCE for the single- and multi-transition decoherence under a strong magnetic field and the 5-pulse PDD (PDD-5) control. The results show that under the strong magnetic field, the single-spin dynamics in the bath (CCE-1) is suppressed and causes 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. The dipolar interaction causes the transition $|\uparrow\downarrow\rangle \leftrightarrow |\downarrow\uparrow\rangle$ with a rate $X_{jk} \equiv \langle \downarrow \uparrow | [I_\sigma - I_\beta] \cdot \mathbf{e}_\sigma | \downarrow \uparrow \rangle$. The hyperfine interaction induces an energy cost of the flip-flop $Z_{jk}^{(a)} = \alpha (A_j - A_k) \cdot \mathbf{e}_\sigma$, for the electron spin state $|\alpha\rangle$. Thus the flip-flop is mapped to the precession of a pseudo-spin $\sigma_{jk}$ about a pseudo-field $h_{jk}^{(a)} = \{X_{jk}, 0, Z_{jk}^{(a)}\}$ [see Fig. 2(d)], which is conditioned on the electrons spin state $|\alpha\rangle$. The pronged bath evolution shown in Fig. 1(c), which causes the center spin decoherence, is reduced to pronged pseudo-spin precession. The center spin decoherence caused by pair flip-flops is factorized as

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

where $|\sigma_{jk}^{(\alpha/\beta)}(t)\rangle$ is the precession of the pseudo-spin about the pseudo-field $h_{jk}^{(\alpha/\beta)}$ for the center spin state $|\alpha/\beta\rangle$.

Figure 3(a) presents the main result of this paper. 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 multi-transition 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 multi-transition coherence even lasts longer than the single-transition coherence.

The anomalous decoherence effect, though counter-intuitive, can be understood using the pseudo-spin picture as illustrated in Fig. 3(b) and (c). The center spin decoherence is determined by the distance between the pseudo-spin pathways conditional on the electron spin states. In the Hahn echo, the decoherence due to the pair flip-flops is [21, 22]

$$L_{\alpha\beta}(t) \approx \prod_{jk} \left[ 1 - 2 \sin \left( h_{jk}^{(\alpha)} \tau / 2 \right) \times \sin \left( h_{jk}^{(\beta)} \tau / 2 \right) \right].$$

Actually in the short time limit $h_{jk}^{(\alpha)} \tau \ll 1$, the multi-transition coherence decays faster than 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 in the bath, the interaction between the two $^{13}$C spins is weaker than 100 Hz, while the hyperfine energy cost of the pairwise flip-flop is $> \text{kHz}$. Therefore in the case of multi-transition, the two pseudo-fields $h_{jk}^{(\alpha)}$ corresponding to the electron spin states $|\alpha\rangle$ are approximately anti-parallel. Thus the distance between the bifurcated pseudo-spin pathways and hence the induced decoherence are small. While in the single-transition case, the two pseudo-fields $h_{jk}^{(\alpha)}$ and $h_{jk}^{(\beta)}$ are in general not (anti-)parallel, and the bifurcated pseudo-spin pathways may deviate largely from each other, which induces strong decoherence. As the number of PDD pulses and hence the coherence time increase, such control effect on the
bath dynamics becomes significant, and therefore the multi-transition can have longer coherence time than the single-transitions.

The conditional flip-flops of the nuclear spin pairs also explain the observation that the oscillation features in the single-transition coherence are absent in the multi-transition coherence. A careful examination of decoherence caused by each individual pair reveals that the rapid and shallow modulations in the decoherence profile are induced by those pairs which have one $^{13}\text{C}$ located relatively close to the NV center. Such pairs have large hyperfine energy cost $Z_{ij}$ in the flip-flop. The large pseudo-fields cause rapid precession of the pseudo-spin when the center spin is in the states $|\pm\rangle$ but the pseudo-spin precession for the center spin state $|0\rangle$ is still slow. This causes rapid oscillation in the single-transition coherence. The slow and relatively deep modulations on the single-transition coherence are caused by flip-flops of pairs which have two $^{13}\text{C}$ spins close to each other. Neither the rapid nor slow oscillation features are visible in the multi-transition coherence. This is because of the fact that for the multi-transition, the two pseudo-fields $b_{ij}$ are nearly anti-parallel, and the decoherence induced by each pair is small.

In conclusion, we have discovered that the multi-transition 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 anomalous decoherence effect establishes the quantum nature and the controllability of an interacting nuclear spin ensemble in a solid-state system at room temperature.

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