Probing the collective dynamics of nuclear spin bath in a rare-earth ion doped crystal

Zong-Quan Zhou,1,2 Yu Ma,1,2 Tao Tu*,1,2 Pei-Yun Li,1,2 Zong-Feng Li,1,2 Chao Liu,1,2 Peng-Jun Liang,1,2 Xiao Liu,1,2 Yi-Lin Hua,1,2 Tian-Shu Yang,1,2 Jun Hu,1,2 Xue Li,1,2 Yi-Xin Xiao,1,2 Yong-Jian Han,1,2 Chuan-Feng Li1,1,2 and Guang-Can Guo1,2
1CAS Key Laboratory of Quantum Information, University of Science and Technology of China, CAS, Hefei, 230026, China
2Synergetic Innovation Center of Quantum Information and Quantum Physics, University of Science and Technology of China, Hefei, 230026, China
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Probing collective spin dynamics is a current challenge in the field of magnetic resonance spectroscopy and has important applications in material analysis and quantum information protocols. Recently, the rare-earth ion doped crystals are an attractive candidate for making long-lived quantum memory. Further enhancement of its performance would benefit from the direct knowledge on the dynamics of nuclear-spin bath in the crystal. Here we detect the collective dynamics of nuclear-spin bath located around the rare-earth ions in a crystal using dynamical decoupling spectroscopy method. From the measured spectrum, we analyze the configuration of the spin bath and characterize the flip-flop time between two correlated nuclear spins in a long time scale (∼1 s). Furthermore, we experimentally demonstrate that the rare-earth ions can serve as a magnetic quantum sensor for external magnetic field. These results suggest that the rare-earth ion is a useful probe for complex spin dynamics in solids and enable quantum sensing in the low-frequency regime, revealing promising possibilities for applications in diverse fields.

Introduction

Quantum memories are essential building blocks for a large-scale quantum network [1, 2] and can be constructed with various physical systems [3, 4]. There are important evaluation criteria with relevance for practical applications, and the key requirement hereby is the ability to store quantum states for coherence times that are longer than the direct transmission time of the channel [1]. Strategies such as environmental and materials engineering [3, 4], quantum error correction [7] and optimal control pulses [8] can improve the coherence time, but, in general, the efficiency of these methods is sensitive to the specific knowledge of the dynamics of environmental fluctuations.

Rare-earth (RE) ion doped crystals have been recognized as a promising candidate system for photonic quantum memory [9–14]. Storage of single photons with extended lifetime [10], wide bandwidth [11], large multimode capacity [12], with telecom interface capability [13] and in a nanoscale medium [14] have been demonstrated using this system. However, these remarkable results are achieved using different samples and various experimental configurations. Further combination of these techniques into a single system still remains a significant challenge. In the RE doped crystal, the coherence of the RE ions is affected by the magnetic fluctuations caused by surrounding nuclear spins. Characterization of the environmental spectrum of the nuclear spin bath will be particularly useful for identifying the optimal sample configuration and design of novel memory protocols.

Dynamical decoupling (DD) spectroscopy has been employed as the useful tool for detecting the spin dynamics in various systems [15–19]. Pioneering work has focused on sensing individual nuclear spins and nuclear-spin clusters in diamond [20–22], and resolving the bath of electronic spins on diamond surface [23–25]. Despite these remarkable progress, probing the spin bath dynamics in photonic quantum memories, especially in the low-frequency regime, is still unexplored. In this work, we experimentally detect the collective dynamics of the Yttrium (Y) nuclear spins in a Eu3+ ions doped Y2SiO5 crystal using DD spectroscopy method. To achieve the goal of understanding the bath dynamics, we solve two important problems: characterization of the spectrum of bath fluctuations (a double-Lorentzian spectrum), and determination of the correlation time of spin bath (∼1 s). Using the Eu3+ ions as the probe, we further present a proof-of-principle experiment on quantum sensing of an external magnetic field. These results promise new applications in a wide range of fields from detection and analysis of the complex many-body effects in solids to design of the advanced quantum information processing protocols.

Results

Probing the spectrum of spin bath dynamics

The physical platform is based on the hyperfine structure of the electronic ground state (7F0) for 151Eu3+
ions occupying site 1 Y positions in the Y$_2$SiO$_5$ crystal. As illustrated in Fig. 1a, the doubly degenerate hyperfine state $|m_I = \pm \frac{3}{2}\rangle$ of $^{151}$Eu$^{3+}$ ion is split by the external magnetic field $B_{dc}$. Since the dynamics of nearby Y nuclear spins cause a randomly fluctuating magnetic field $\delta B(t)$ at the $^{151}$Eu$^{3+}$ ion site, the perturbation contributing to detuning of the transition frequency (i.e., decoherence between the energy levels) is $\delta_B(t) = \omega_0 + S_1 \delta B(t) + S_2 \delta B(t)^2 + ...$, where $\omega_0$ is the transition frequency at the bias field $B_{dc}$, $S_1$ (or $S_2$) is the first (or second) order Zeeman coefficient (The complete characterization of the spin Hamiltonian for $^{151}$Eu$^{3+}$ is given in Supplementary Information). Since we are only interested in small perturbation field $\delta B(t)$ in this work, high orders Zeeman effect including $S_2$ can be ignored. Thus the measured coherence time $T_2$ of the $^{151}$Eu$^{3+}$ ions can be used as an indicator of the Y spin bath dynamics $\frac{1}{T_2} \approx S_1 \Delta B$, where $\Delta B$ is the variance of the magnetic field fluctuations. When $B_{dc}$ approaches a critical magnetic field where $S_1 \approx 0$, this is the so-called zero first order Zeeman (ZEOFZ) point where $T_2$ of the considered spin transition will be greatly extended.

![FIG. 1: Schematic of the RE doped crystal and experimental scheme.](Image)

We detect the spin bath dynamics by employing the spectroscopy method based on DD [15-18]. The experimental scheme are schematically shown in Fig. 1b. In the intuitive picture, the collective dynamics of nuclear spin bath consists of different kinds of components in the frequency domain. One component of the spin bath dynamics induces an a.c. magnetic field fluctuation with a characteristic frequency $\nu$. The application of multiple short $\pi$ pulses filters out the spin bath fluctuation $\delta B(t)$ at a narrow-band range of frequency $\nu$ [15-18].

$$U(t) = \int_0^\infty F(\nu)S(\nu)d\nu. \tag{1}$$

Here, $C = \exp(-U(t))$ corresponds to spin-echo signal amplitude of the $^{151}$Eu$^{3+}$ ions, $S(\nu)$ is the spectral density of the spin bath fluctuation and $F(\nu)$ is the filter function determined by the pulse sequences (see Methods). This allows one to extract a wealth of information.
about the environmental spectrum $S(\nu)$ from the measured spin-echo data and hence probe the particular dynamics of spin bath.

Details about the experimental procedures are presented in Methods and Supplementary Information. In short, optical pumping is firstly employed to polarize the $^{151}$Eu$^{3+}$ ions into the $|m_J = +3/2\rangle$ spin level. After radiofrequency (RF) multiple-pulse manipulation, the finally emitted spin-echo signal is probed optically. The measured spin-echo amplitude $C$ under the Carr-Purcell-Meiboom-Gill (CPMG) pulse sequence as a function of pulse spacing $\tau$ is displayed in Fig. 2a and 2b. Here, we set the working condition as $B_{dc}$ being 200 G greater than the ZEFOZ field (Fig. 2a) and near the ZEFOZ point (Fig. 2b), respectively. The ZEFOZ field is chosen as approximately $[0.685 \text{ T}, -0.812 \text{ T}, 0.714 \text{ T}]$ along the [D1, D2, b] crystal axis. Then, we fit the experimental data with an exponential form $\exp(-t/T_2)$, from which we can extract the coherence time $T_2$ of the RE ion for different pulse intervals $\tau$, as shown in Fig. 2c. Diagram Fig. 2c shows several interesting features of the coherence behaviors: First, the RE ion has much longer coherence times with more applied pulses, which clearly illustrates the efficiency of the DD pulse sequences for the present system. The coherence time of the RE ion is measured to be $T_2 = 73.8$ s for pulse interval $\tau = 12$ s, while it extends significantly to $T_2 = 259$ min (4.3 hours) for pulse interval $\tau = 0.05$ s. Second, the decoherence rate can be described by a scaling formula $1/T^2 \sim \tau^\beta$ with power $\beta = 1$ (the solid line in the Fig. 2c). A similar scaling has been observed in the nitrogen-vacancy (NV) centers in diamond system [27], while in our case, the scaling spans over a several-hour-long time scale. Moreover, the value of the power $\beta = 1$, obviously deviates from the number $\beta = 2$ in the NV center system [27], which means that there is a different mechanism at work. The robust scaling behaviors provide evidence that the decoherence of central RE ions originates from the intrinsic dynamics of the spin bath around the RE ions.

In Fig. 3a, we present the extracted spectrum of the spin bath dynamics with $B_{dc}$ of 200G greater than ZEFOZ field. The typical spectrum obtained is dominated by the contribution from the low-frequency spin bath dynamics. We fit the detected result as a double-Lorentzian spectrum $S(\nu) = \frac{1}{\pi} \frac{2b^2 \tau_c^2}{\nu^2 (\tau_c^2)^2 + 1} + \frac{1}{\pi} \frac{2b^2 \tau_f^2}{\nu^2 (\tau_f^2)^2 + 1}$, where $b = 2.46$ Hz is the average coupling strength of the bath to the central RE ions, and $\tau_c$ is the correlation time of the bath dynamics. In particular, the environmental spectrum is characterized by a “two-time-scale”: $\tau_c = 2.54$ s for the slow dynamics component (see the green region in Fig. 3a), and $\tau_f = 0.25$ s for the fast dynamics component (see the yellow region in Fig. 3a).

In Fig. 3b we show the probed environmental spectrum with $B_{dc}$ near the ZEFOZ field. The measured spectrum are in overall similar to that far away from the ZEFOZ case, confirming that the “two-time-scale” environmental dynamics are responsible and universal for the present system. The characterized “two-time-scale”
values are $\tau_c^s = 28.6$ s for the slow dynamics component and $\tau_c^f = 0.25$ s for the fast dynamics component. In addition, the spectrum have a much smaller amplitude of $b = 0.12$ Hz, one order of magnitude smaller than those far away from the ZEFOZ case. Comparing the data in Fig. 3a, we find that the dynamics strength of the bath spins is clearly suppressed (i.e., slower $\tau_c$ and smaller $b$) near the ZEFOZ point.

**Analysis of the spin bath dynamics**

Diagram Fig. 3 shows several remarkable features of the collective spin bath dynamics: First, we find that the typical environmental spectra in solids such as power law spectra $\frac{1}{v^\alpha}$ in a superconducting flux qubit [13, 28] and simple Lorentzian spectra $\frac{1}{1+R^2}$ in bulk diamond systems with various NV center densities and impurity concentrations [18], cannot describe our measured data (the fitting is rather poor, please see Fig. S2 in the Supplementary Information). This suggests different environmental dynamics in the present system compared to that in other solids. Second, the environment spectrum at different working points have a similar fast correlation time $\tau_c^f = 0.25$ s, while they have different slow correlation time $\tau_c^s$. Under the same multiple-pulse control, the RE ions can achieve different coherence time $T_2$ for different slow correlation time $\tau_c^s$; for comparison, the maximum $T_2 = 40$ s for $\tau_c^s = 2.54$ s (200G away from the ZEFOZ point), the maximum $T_2 = 259$ min for $\tau_c^s = 28.6$ s (near the ZEFOZ point). We also note that the observed characteristic time $\tau_c$ is orders of magnitude longer than that of other candidate system for quantum memories [27], which suggests that the probed spin bath dynamics are in the low-frequency regime.

We initially explain this double-Lorentzian spectra is attributed to the special dynamics of the spin bath. The dynamics of Y spin bath include single nuclear-spin precession and flip-flop processes of nuclear-spin pairs. Our experiments are performed in strong magnetic fields, where the single nuclear-spin precession is strongly suppressed. Thus, the dynamics of the spin bath are dominated by the interacting nuclear-spin pairs. When one bath spin $i$ flips, a corresponding bath spin $j$ flops in the opposite direction. A large number of flip-flop events show that the configuration of spin bath changes over time, which can be considered as the collective dynamics of spin bath with different kinds of flip-flop rates $R_{ij}$.

Moreover, the strong coupling between the central $^{151}$Eu$^{2+}$ ions and neighboring Y bath spins induces that Larmor frequencies of these bath spins are detuned from others and cannot readily exchange. As shown in Fig. 1b, this create a so-called “frozen core” region [9], where the flip-flop rate $R_{ij}$ of these bath spins is significantly suppressed. Therefore, the dynamics of the bath spins can be characterized by two distinct time scales: slow dynamics $\tau_c^s \sim 1/R^s$ (small flip-flop rate $R^s$) in the “frozen core” region and fast dynamics $\tau_c^f \sim 1/R^f$ (large rate $R^f$) outside the “frozen core” region, respectively.

The flip-flop events show that the configuration of the spin bath changes in time. As a result, the bath dynamics create a fluctuating magnetic field $\delta B(t) \sim \sum_i n_i(t)$ as a sum of a large number of independent configurations $\{...n_i(t)...n_j(t),...\}$, where $n_i(t) = 1$ if $i$-th bath spin at time $t$ is $|\uparrow\rangle$, and $n_i(t) = -1$ for $|\downarrow\rangle$. Thus the environmental dynamics can be modeled as a random process with Gaussian distribution according to the central limit theorem [27]. Due to lack of coherence between different configurations of spin bath, this dynamics also does not depend on history, i.e. it is Markovian. Summarizing all of the above, we can expect the environment dynamics in each region can be represented as a correlation function $\langle \delta B(t)\delta B(0) \rangle \sim \exp(-t/\tau_c)$ [27, 28]. Here $\tau_c = 1/R$ is the correlation time of this process. Since
there are two regions with different time scales or flip-flop rates $\tau^x = 1/R^x$, $\tau^f = 1/R^f$, we conclude that there is a double-Lorentzian spectrum consisting of two components $S(\nu) \sim \frac{1}{v^2(\tau^x)^2+1} + \frac{1}{v^2(\tau^x)^2+1}$. (More quantitative analysis are provided in the Supplementary Information).

Detection of an external magnetic field

The above results demonstrate that the RE ions can be used for detecting the spin-bath spectrum inside the crystal. Inspired by this experimental precursor, next we report that the RE ions can also be employed for detection of an external field. An example measurement for an external ac magnetic field $B_{ac}(t)$ is presented in Fig. 4. Details about the experimental procedures are presented in the Supplementary Information. Here, we set the working condition as $B_{dc}$ being 200 G greater than the ZEFOZ field. Simple two-pulse spin-echo sequence is employed for sensing an synchronized external field $B_{ac}$, the accumulated phase during the two-pulse sequence can be written as $\phi = \int_0^{\tau} S_1 B_{ac}(t) dt - \int_0^{2\tau} S_1 B_{ac}(t) dt = \pi \tau S_1 B_{ac}$, where $\tau$ is the total evolution time and $B_{ac}(t) = B_{ac} \sin(2\pi\nu t)$, with frequency $\nu = 1/(2\tau)$, is the external field to be measured. Thus, detecting the relative phase shift $\phi$ induced by the magnetic field enables precise determination of the applied ac magnetic field $B_{ac}(t)$. The real part (X) and the imaginary part (Y) of the spin-echo signal are measured simultaneously to minimize the affects from the fluctuations of the laser power and RF power (Detailed discussions are given in the Supplementary Information). As shown in Fig. 4a, when the amplitude of the ac magnetic field increases, due to the accumulation of the phase, the spin-echo signal varies periodically. A sensitivity of 10.8 nT/$\sqrt{\text{Hz}}$ is achieved at a detecting frequency of 0.75 Hz. Another example measurement performed with a working condition of $B_{dc}$ being 6 G greater than the ZEFOZ field is presented in Fig. 5 in the Supplementary Information. We achieved a sensitivity of 118 nT/$\sqrt{\text{Hz}}$ at a detecting frequency of 66 mHz. Therefore, the present RE ion magnetometer enables sensitive detection of external fields in the low-frequency regime near 0.1 Hz, which is not accessible by other quantum sensors with much shorter coherence times.[31]

**Discussion**

In conclusion, we have measured and characterized the collective dynamics of nuclear-spin bath in a RE ion doped crystal. From the coherence measurements of the central $^{151}$Eu$^{3+}$ ions under multiple-pulse control and various magnetic fields, we determine the spectrum of the Y bath dynamics and its correlation times. The RE ions doped in solids can also serve as a quantum sensor for external magnetic fields. With this observation, this proof-of-principle work could be the first step towards the detection of various environmental processes in other RE doped solids, especially in the Eu$^{3+}$: Y$_2$SiO$_5$ crystal, a candidate for a quantum memory working in the 1550 nm communication band.[32] Recent investigations have also facilitate the detection and manipulation of single RE ion in a nanocrystal with long coherence time.[33, 34] Hence, this work also opens up wide perspective for applications in the field of detecting the interesting

![FIG. 4: Demonstration of quantum sensing of an external ac magnetic field.](image-url)
dynamics of complex materials with high-sensitivity, in the low-frequency regime and with nano-scale resolution.

Methods

Experimental setup

The sample is a 0.01% doped Eu$^{3+}$:Y$_2$SO$_5$ crystal with a thickness of 1 mm along the crystal's b-axis. The sample is oriented within a superconducting magnet contained in a cryogen-free cryostat and its temperature is maintained at 1.80±0.01 K. The 580-nm laser beam from a frequency-doubled semiconductor laser is further modulated with an acoustic-optic modulator to generate desired preparation and probe light. The RF field, which is controlled by an arbitrary waveform generator and amplified by a 300-W amplifier, is employed to stimulate different RF pulse schemes.

The RF spin-echo signal is readout by optical Raman heterodyne detection [9]. Before the RF manipulation sequence, the pump laser prepares the large population difference in the considered spin transition to obtain a sequence, the pump laser prepares the large population heterodyne detection [9]. Before the RF manipulation sequence, the pump laser prepares the large population difference in the considered spin transition to obtain a large signal. After the RF manipulation, a synchronized laser pulse at $\omega_L$ ($\sim 517.471$ THz) converts the RF spin-echo into an optical pulse at $\omega_L + \omega_{RF}$ through a two-photon process and Raman scattering. The spin echo is observed by beating the generated light at $\omega_L + \omega_{RF}$ and the probe light at $\omega_L$. The phase accumulation of the spin-echo is observed by mixing the beat signal of the spin-echo with two separate RF local oscillator channels separated in phase by 90°. More details about the experimental setup can be found in the Supplementary Information.

Spectroscopy method based on dynamical decoupling control

We quantify the coherence of central RE ions by using the function $C = \exp[-U(t)]$, which is defined as the off-diagonal elements of the density matrix of the spin state of RE ions. Formally, the dynamics of the RE spin are analyzed using the evolution operator. On the one hand, the decoherence process from the environmental fluctuation $\delta B(t)$ is introduced by $\exp(-i\frac{\sigma_z}{2}\delta B(t)t)$. On the other hand, DD sequences can be considered as applying a certain number of $\exp(-i\frac{\delta B(t)}{2}\sigma_z) = -i\sigma_x$ at time $t_i$. Therefore, we can write the evolution operator by slicing the time interval into small pieces:

$$C(t) = \left\langle \exp(-i \int_0^t F(t') \delta B(t') dt') \right\rangle,$$

where $F(t)$ is the temporal filter function, which takes value $-1$ and $+1$ and changes the sign at each pulse time $t_i$. Then, using the Fourier transformation of the above equation, $U(t)$ can be calculated through the spectral density $S(v)$ of the environmental fluctuation as Eq. (1).

The filter function for the n-pulse CPMG control is calculated as $F(vt) = 8 \sin^2(vt) \sin^2(vt/4n)/\cos^2(vt/2n)$ [35]. Since we employ the pulse sequences with a large number $n$, the filter function $F(vt)$ can be effectively approximated as a $\delta$-function in the frequency domain. Flipping the central RE ions by a sequence of $\pi$ pulses will suppress the effect of the fluctuating magnetic field components at every other frequency while filtering the fluctuating magnetic filed component oscillating synchronously with the pulse interval $\tau = 1/2v$. Thus, repeating the experiment for different pulse spacings $\tau$, we can scan the entire spectrum $S(v)$ of the environment.

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Author Contributions Z.-Q. Z., T. T. and C.-F. L. designed experiment. Z.-Q. Z. carried out the experiment assisted by Y. M., P.-Y. L., Z.-F. L., C. L., P.-J. L., X. L., Y.-L. H., T.-S. Y., J. H., X. L., and Y.-X. X. Z.-Q. Z., T. T. and Y. M. analyzed the experimental results. T. T. analyzed the dynamical decoupling spectroscopy. Y. M. and Y.-J. H. did the calculation for the spin Hamiltonian. C.-F. L. and G.-C. G. advised on the project. Z.-Q. Z. and T. T. wrote the paper with input from other authors. All authors discussed the experimental procedures and results.

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