

Nuclear Spins in a Solid Exceeding 10-Hour Coherence Times for Ultra-Long-Term Quantum Storage

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Long-term quantum storage for quantum communication applications will require optically accessible states with coherence times exceeding the desired storage time. Ground-state hyperfine levels in $\text{Eu}^{3+}:\text{Y}_2\text{SiO}_5$ have previously shown coherence times as long as 6 h at 1.4 K when operated at a zero-first-order-Zeeman (ZEFOZ) point. Here, we have characterized the limiting decoherence mechanisms acting in this regime, which shows that the coherence time is limited by the presence of spin impurities. This dephasing effect has been suppressed by cooling the sample to 125 mK, resulting in an extension in the coherence time to over 13 h. In the absence of these impurities, the dominant source of dephasing is the field inhomogeneity of the ZEFOZ point arising from the spin inhomogeneous broadening. We have shown that coherence times over 18 h can be achieved by operating using a spectrally narrow subensemble selected from within the inhomogeneous line. The observed coherence time is longer than that of any other system suitable for an optical quantum memory. Furthermore, at a much higher temperature of 6 K, we have realized a 6-h coherence time, which has been obtainable using single-stage cryocoolers. These results imply advanced engineering feasibility in designing and operating future quantum information-storage devices for a wide range of applications, including in satellites.

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I. INTRODUCTION

Rare-earth-ion- (REI) doped crystals have been identified as promising material platforms for optical quantum memories for light, due to the existence of stable solid-state optical centers possessing long coherence times for both optical and hyperfine transitions. The ability to work with a high density of these centers while maintaining good

coherence properties provides the potential to develop quantum memories with simultaneously high data storage densities and long storage times [1–3].

Several different storage protocols have been exploited in these systems and remarkable achievements in memory performance have been reported in recent years [3], including an efficiency of 76.3% [4], a storage mode number of 1650 [5,6], and a storage time of 20 ms [7]. Consequently, there is significant interest in using REI quantum memories for quantum communication, either as storage devices in quantum repeater systems or as a transportable quantum memory [8].

For a transportable quantum memory, quantum information is distributed by physically transporting the memory-material system rather than the light. Storage times over 1 h are likely to be required for practical communication applications. The observation of 6-h coherence times on a hyperfine transition of $\text{Eu}^{3+}:\text{Y}_2\text{SiO}_5$ has demonstrated the

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potential for developing a system suitable for transporting quantum information [9] and recent work has shown coherent optical storage for 1 h using the spin-wave atomic frequency comb in the same material [10]. In this system, the hyperfine decoherence of the Eu^{3+} is dominated by interactions with fluctuating magnetic fields of the host Y^{3+} spin bath. The observed long coherence time has been achieved by operating at a ZEFOZ transition occurring at a field of around 1.3 T and applying a dynamic decoupling (DD) sequence to decouple Eu^{3+} from the host spins. There are two mechanisms contributing to extending the hyperfine coherence time for a ZEFOZ transition. First, a zero gradient in the Zeeman splitting means a significant decrease in the sensitivity of the transition to magnetic field fluctuations. Second, the externally applied magnetic field induces a large magnetic moment on Eu^{3+} , resulting in the so-called “frozen-core effect” [11] by detuning the transition frequencies of nearby host ions from the bulk host ions.

While the coherence time obtained in $\text{Eu}^{3+}:\text{Y}_2\text{SiO}_5$ with ZEFOZ and DD methods is an improvement over the zero-field value by 7 orders of magnitude, it is not yet clear whether 6 h represents a limit for this system, nor how this value is affected by experimental factors such as temperature or crystal concentration. Here, we have addressed this subject by investigating the coherence time of the same ground-state hyperfine transition studied in previous works near the ZEFOZ point and over a range of temperatures. This study has been conducted on two samples with concentrations of 100 ppm and 1000 ppm (parts per million).

II. ZEFOZ FIELD ALIGNMENT

The system investigated here consists of the hyperfine transitions of the electronic ground state (7F_0) of $^{151}\text{Eu}^{3+}$ ions, located in site 1 of the two magnetically nonequivalent sites of C_1 symmetry in the Y_2SiO_5 crystal [12]. We have focused on the $|-3/2\rangle \leftrightarrow | +3/2\rangle$ ZEFOZ transition occurring at a field of around 1.3 T, the same ZEFOZ transition as in the previous work [9]. This transition is referred to as the “A” transition and the ZEFOZ field as $\mathbf{B}_{\text{cp}}$ in this paper, as illustrated in Fig. 1(a).

The studies have been performed mainly on a 100-ppm $\text{Eu}^{3+}:\text{Y}_2\text{SiO}_5$ crystal, while some measurements have also been conducted on a 1000-ppm sample to characterize the impact of the doping concentration. Both crystals are from Scientific Materials, with the same dimensions of $5.0 \times 5.5 \times 3.0 \text{ mm}^3$ ($D_1 \times D_2 \times b$). During all measurements presented in this work, the sample has been kept in a dilution refrigerator that incorporates a superconducting magnet, with a base temperature of approximately 80 mK, monitored using a sensor located at the mixing-chamber flange. The hyperfine transition has been excited by an eight-turn copper coil with a diameter of 12 mm,

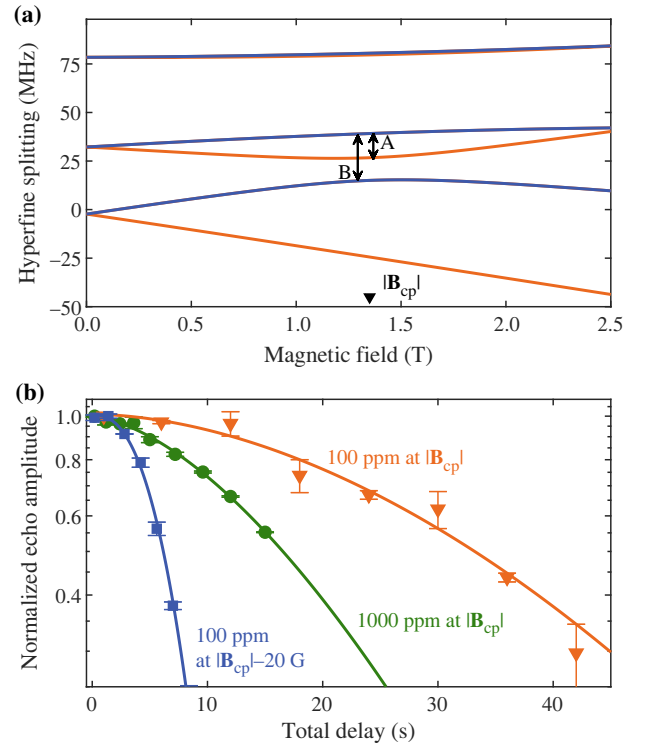


FIG. 1. The hyperfine energy levels and the two-pulse spin-echo (TSE) coherence-time results of the ZEFOZ transition. (a) The calculated energy levels of the hyperfine ground 7F_0 state of $\text{Eu}^{3+}:\text{Y}_2\text{SiO}_5$ with a magnetic field increasing from zero along the direction of $\mathbf{B}_{\text{cp}}$, using the spin Hamiltonian in Ref. [13]. The investigated ZEFOZ transition is $|-3/2\rangle \leftrightarrow | +3/2\rangle$ (the “A” transition), which occurs at $|\mathbf{B}_{\text{cp}}| = 1.3480 \text{ T}$. The transition $| +1/2\rangle \leftrightarrow | +3/2\rangle$ (B-transition) has also been involved in some measurements. (b) The two-pulse spin-echo decay with the measured echo amplitude plotted against the total delay, where the lines give the fit using the spectral-diffusion model $\text{Exp}[-(t/T_M)^2]$ [14]. The phase-memory time T_M is $44.0 \pm 1.0 \text{ s}$ and $7.2 \pm 0.2 \text{ s}$ in the 100-ppm sample measured at $\mathbf{B}_{\text{cp}}$ and $|\mathbf{B}_{\text{cp}}| = 20 \text{ G}$, respectively. T_M is $19.8 \pm 0.3 \text{ s}$ in the 1000-ppm sample at $\mathbf{B}_{\text{cp}}$. The error bars are from fitting uncertainty.

which was aligned perpendicular to the static magnetic field. The radio-frequency (rf) coil was driven by an arbitrary wave form generator, which was amplified using a 100-W amplifier. The spin echo has been optically measured using a Raman heterodyne method. The $^7F_0 \rightarrow ^5D_0$ optical transition has been driven by a Toptica laser tuned to 580.041 nm.

At the base temperature, the field magnitude and direction have been aligned to the target $\mathbf{B}_{\text{cp}}$ with a precision of 1 G by maximizing the two-pulse spin-echo (TSE) coherence time using machine-learning-assisted three-dimensional sampling. The value of $|\mathbf{B}_{\text{cp}}|$ has been found to be 1.3020 T, slightly higher than the 1.29 T reported in the previous work [9]. We attribute the field discrepancy to calibration uncertainty between the two magnet systems.

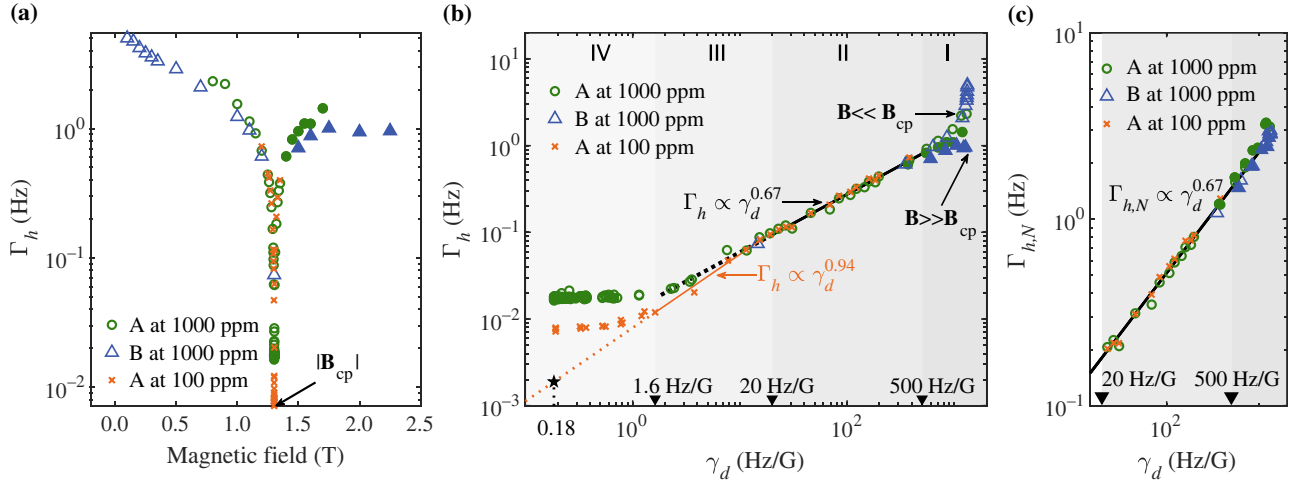


FIG. 2. The magnetic-field-dependent homogeneous line width near B_{cp} with power-law fitting. (a) The measured homogeneous line width Γ_h plotted against the applied magnetic field amplitude, which is detuned along the fixed direction of $\mathbf{B}_{cp}$. Similar to the “A” transition, the Γ_h value of the “B” transition approaches a minimum value because this transition is coincidentally close to another ZEFOZ point at B_{cp} . (b) The measured Γ_h plotted against the differential sensitivity γ_d , which has been calculated based on the applied field and the spin Hamiltonian in Ref. [13]. We identify four regimes (I–IV) showing qualitatively different decoherence behavior, as indicated by the background shading. Regime I contains magnetic fields far from a ZEFOZ point, $B \gg |\mathbf{B}_{cp}|$ (open circles and triangles) and $B \ll |\mathbf{B}_{cp}|$ (filled circles and triangles). Regime II contains intermediate fields where the field sensitivity of Eu^{3+} is reduced to be similar to that of the host Y^{3+} spins ($\gamma_Y = 209 \text{ Hz/G}$). In this regime, the Eu^{3+} is still strongly coupled to the host Y^{3+} . Regime III contains fields that are very close to $\mathbf{B}_{cp}$, with a γ_d value that is far smaller than that of the host Y^{3+} . In this regime, the Eu^{3+} is only weakly coupled to the host Y^{3+} and the coherence time is long enough for spin flips of the frozen-core Y^{3+} to affect the Eu^{3+} . Regime IV contains fields where the Γ_h values of both samples saturate. The black solid line is the fitting for power-law scaling of $\Gamma_h \propto \gamma_d^{0.67}$ using all the data in regime II ($20 \text{ Hz/G} < \gamma_d < 500 \text{ Hz/G}$), while the orange line is the fitting for the orange data points only in regime III ($1.6 \text{ Hz/G} < \gamma_d < 20 \text{ Hz/G}$). The dashed lines are the extension of the fitted lines into regions not included in the fit. The star on the orange dotted line labels the expected Γ_h at $\gamma_d = 0.18 \text{ Hz/G}$, which is the limit imposed by the magnetic field inhomogeneity of 1 G. (c) The normalized homogeneous line width $\Gamma_{h,N} = \Gamma_h \sqrt{\alpha_0 + \alpha_1 \gamma_d}$ of all the three data sets in regimes I and II, which can be described well with a single power-law fitting, plotted with a solid black line.

As shown in Fig. 1(b), the TSE decay can be described well using the spectral-diffusion model [14,15]:

$$M(t) = \text{Exp}[-(t/T_M)^\alpha], \quad (1)$$

where α is a stretching parameter. The fitting gives $\alpha = 2$ and phase-memory times T_M of $19.8 \pm 0.3 \text{ s}$ for the 1000-ppm sample and $44.0 \pm 1.0 \text{ s}$ for the 100-ppm sample at $\mathbf{B}_{cp}$. The latter is comparable to the 47-s coherence time reported in the previous work [9], obtained with a sample of the same doping concentration but at a temperature of 1.4 K.

III. MAGNETIC-FIELD-DEPENDENT DECOHERENCE

In order to explore more details of the decoherence mechanisms, TSE measurements have been conducted on both samples with the field amplitude detuned gradually along the fixed direction of $\mathbf{B}_{cp}$. During these measurements, the mixing chamber has been maintained at approximately 80 mK. With the goal of gaining insight into the transition-independent decoherence behavior, we have also performed coherence measurements in the 1000-ppm

sample under the same magnetic field conditions but corresponding to a different transition, which is the $|+1/2\rangle \leftrightarrow |+3/2\rangle$, labeled “B” in Fig. 1(a). As shown in Fig. 2(a), the measured homogeneous line width $\Gamma_h = 1/(\pi T_M)$ approaches a minimum value near $\mathbf{B}_{cp}$ for all three data sets. The Γ_h value of the “B” transition approaches a minimum value because this transition is also coincidentally close to another ZEFOZ point at $\mathbf{B}_{cp}$. For most discussions in this paper, we focus on the “A” transition, but data from the “B” transition, for which we happened to record more data points when well detuned from $\mathbf{B}_{cp}$, have been used when fitting the “frozen-core effect” model detailed later in this section.

For $\text{Eu}^{3+}:\text{Y}_2\text{SiO}_5$, the hyperfine decoherence of Eu^{3+} is mainly caused by bath magnetic field fluctuations $\vec{\zeta}_B(t)$ resulting from random flips and flip-flops of bath spins [9]. As a result, the transition frequency undergoes a stochastic process of $\zeta(t) = \vec{\gamma}_d \cdot \vec{\zeta}_B(t)$, where $\gamma_d = |\vec{\gamma}_d| = |\vec{\gamma}_e - \vec{\gamma}_g|$ is the differential sensitivity of the transition that can be determined from the spin Hamiltonian model and $\vec{\gamma}_e$ and $\vec{\gamma}_g$ are the sensitivity vectors of the hyperfine excited and ground states, respectively. Because the orientation of $\vec{\gamma}_d$ does not change in this field regime (although the

magnitude does), and under the assumption that the magnetic field fluctuation $\vec{\zeta}_B(t)$ is isotropic, there is a linear relation of $\zeta(t) \propto \gamma_d |\vec{\zeta}_B|$. We have plotted the measured homogenous line width Γ_h against the calculated γ_d in Fig. 2(b).

The relation between the homogenous line width Γ_h and the sensitivity γ_d depends on the nature and rate of the fluctuations of the spin bath. A common model, described by Klauder and Andersen [15], assumes that bath spins flip slowly (compared to the measurement time) between two states (the “sudden jump” model). Then, the homogeneous broadening is dependent on two parameters: δ , the half width at half maximum (HWHM) of the spectral broadening of the transition caused by the environmental noise spins, and the flipping rate R as $\Gamma_h = \sqrt{R\delta/(2\pi^2)}$ [14,15]. Provided that the range of magnetic fields considered around a central field ($\mathbf{B}_{cp}$ here) is small, the magnetic field fluctuation $\vec{\zeta}_B(t)$ can be assumed to maintain a constant distribution width, while δ is linearly dependent on the HWHM of $\zeta(t)$ and correspondingly linearly dependent on γ_d . Thus, the homogeneous broadening can be defined in terms of the sensitivity as

$$\Gamma_h \propto (R\gamma_d)^{1/2}. \quad (2)$$

Different models (discussed below) exist for different baths and measurement time scales, all giving a power-law scaling similar to Eq. (2) but with different exponents. Here, we use the experimentally derived power-law exponent to elucidate the nature of the fluctuations causing Eu^{3+} decoherence.

We identify four regimes in Fig. 2(b) showing quantitatively different decoherence behavior. Regime I ($\gamma_d > 500$ Hz/G) contains magnetic fields far from a ZEFOS point, $B \gg |\mathbf{B}_{cp}|$ (open circles and triangles) and $B \ll |\mathbf{B}_{cp}|$ (filled circles and triangles). In this regime, both the sensitivity and the line width are high, but the line width for $B \gg |\mathbf{B}_{cp}|$ is lower than for $B \ll |\mathbf{B}_{cp}|$, even with the same γ_d value. This can be attributed to the frozen-core effect [11]. A larger dipole moment, induced in Eu^{3+} ions by a higher magnetic field, expands the range of the frozen core and increases the detuning between host spins in the core, which in turn suppresses the effective fluctuation rate R of the bath noise and narrows Γ_h . In $\text{Eu}^{3+}:\text{Y}_2\text{SiO}_5$, the induced dipole moment is characterized by an average sensitivity $\gamma_A = |(\vec{\gamma}_e + \vec{\gamma}_g)/2|$. Previous research in REI systems gives the effective bath fluctuation rate $R \propto 1/(\alpha_0 + \alpha_1\gamma_A)$, where α_0 and α_1 are the fitting constants [16–19]. These constants can be extracted by fitting all data in regime I with the phenomenological function

$$\Gamma_h \propto \sqrt{\frac{1}{\alpha_0 + \alpha_1\gamma_A}} (\gamma_d)^\beta. \quad (3)$$

This fitting results in $\alpha_0 = 0.000 \pm 0.007$, $\alpha_1 = 0.023 \pm 0.001$, and a power-law exponent $\beta = 0.67 \pm 0.01$. When this fit is used to calculate a normalized homogeneous line width ($\Gamma_{h,N} = \Gamma_h \sqrt{\alpha_0 + \alpha_1\gamma_A}$) that removes this frozen-core effect, the $B \gg |\mathbf{B}_{cp}|$ and $B \ll |\mathbf{B}_{cp}|$ data now collapse onto a single line, as shown in Fig. 2(c).

In regime II, in which $20 \text{ Hz/G} < \gamma_d < 500 \text{ Hz/G}$ ($|\mathbf{B}_{cp}| \pm 0.1500 \text{ T}$), the Eu^{3+} is still strongly coupled to the host Y^{3+} and a power law of $\Gamma_h \propto \gamma_d^{0.67 \pm 0.01}$ has also been obtained with fitting of all three of the data sets. The power-law index $\beta = 0.67 \pm 0.01$ does not have the value of 0.5, given in Eq. (2), but it is consistent with the Gaussian spectral-diffusion model [14,15,20,21], as we briefly describe here (see Appendix C).

In this model, which assumes fluctuations that are Gaussian, stationary, and Markovian, the autocorrelation function of the fluctuation is

$$\langle \zeta(t)\zeta(t') \rangle = \sigma_\omega^2 e^{-|t-t'|/\tau_c}, \quad (4)$$

where $\tau_c = 1/R$ is the correlation time and σ_ω is the standard deviation of the broadened transition, which is linearly correlated with δ , the HWHM of the spectral broadening. Then, the echo-envelope evolution is given by [15,22]

$$M_G(t) = \text{Exp} \left\{ -\sigma_\omega^2 \tau_c^2 \left[\frac{t}{\tau_c} - 3 + 4e^{-t/(2\tau_c)} - e^{-t/\tau_c} \right] \right\}. \quad (5)$$

In the limit of $\sigma_\omega \tau_c \gg 1$ [15], for which the variation in the transition frequency is much larger than the fluctuation rate characterized by the correlation time τ_c , we recover the expected power law of

$$\Gamma_h \propto (\gamma_d)^{2/3} \quad (6)$$

seen in regimes I and II. This limit is termed the “strong-coupling” limit and it is reasonable to see such a “strong coupling” between Eu^{3+} and the host Y^{3+} in these regimes because the field sensitivity of Eu^{3+} is still larger than or comparable to that of the host Y^{3+} spins ($\gamma_Y = 209 \text{ Hz/G}$).

In contrast, regime III ($1.6 \text{ Hz/G} < \gamma_d < 20 \text{ Hz/G}$) is defined by a very different power-law dependency of $\Gamma_h \propto \gamma_d^{0.94 \pm 0.04}$, for the 100-ppm data (the 1000-ppm sample does not display the same rate, due to an earlier onset of the saturation behavior described below for regime IV). The higher power indicates an accelerated narrowing of Γ_h as the sensitivity γ_d is reduced, which is referred to as a “narrowing effect.”

Under the Gaussian assumption, Eq. (5) does lead to a narrowing effect in the limit of $\sigma_\omega \tau_c \ll 1$, namely, when the central system experiences weak but rapid field fluctuations. The power law expected from the Gaussian model in this limit is $\Gamma_h \propto \sigma_\omega^2 \propto \gamma_d^2$ (see Appendix C), with a

TABLE I. A summary of the value of the index β , which describes the power-law scaling of the measured homogeneous line width Γ_h , against the field sensitivity γ_d with $\Gamma_h \propto (\gamma_d)^\beta$.

Experiments	Diffusion model	
	Strong-coupling $\sigma_\omega \tau_c \gg 1$, regimes I and II	Weak-coupling $\sigma_\omega \tau_c \ll 1$, regime III
Gaussian	0.67 (✓)	2 (×)
Lorentzian	0.5 (×)	1 (✓)

much larger power-law index than the value obtained in our experiment.

The other popular model to describe spectrum diffusion is the Lorentzian model [14,15], for which the transition-line broadening is assumed to be Lorentzian (see Appendix C). In this model, the echo-envelope evolution is written as

$$M_L(t) = \text{Exp} \left\{ -\sigma_\omega \tau_c \left[\frac{t}{\tau_c} - 2 \ln(2 - e^{-t/(2\tau_c)}) \right] \right\}, \quad (7)$$

leading to a power law of $\Gamma_h \propto \gamma_d^1$ in the limit of $\sigma_\omega \tau_c \ll 1$, in good agreement with the measured data of the 100-ppm sample in regime III. In the limit $\sigma_\omega \tau_c \gg 1$, this model does not match the data, with a power law of $\Gamma_h \propto \gamma_d^{0.5}$.

The above discussion on the power-law dependency of $\Gamma_h \propto \gamma_d^\beta$ in regimes I–III is summarized in Table I. The spectral-diffusion process in regimes I and II can be described under the Gaussian strong-coupling model, corresponding to a power-law index of $\beta = 0.67$. When the magnetic field is tuned close to $\mathbf{B}_{cp}$, corresponding to regime III, it satisfies a Lorentzian weak-coupling model with $\beta = 1$.

The change of the coupling between the Eu^{3+} and the host Y^{3+} from strong-coupling to weak-coupling mode is in line with expectations. It occurs when the ZEFOZ field alignment is improved and γ_d is reduced; thus $\sigma_\omega = \gamma_d \delta B_Y \ll R$, namely, $\sigma_\omega \tau_c \ll 1$, where δB_Y is the field distribution caused by the host Y^{3+} and R is the effective flipping rate of these spins, including contributions from both the bulk and the frozen-core Y^{3+} . From the calculation detailed in Appendix D, the field distribution δB_Y is 1 μT for the strong-coupling regime and the flipping rate R is 0.2 Hz.

The transition from Gaussian to Lorentzian broadening can also be explained in terms of the underlying physics: it arises from the frozen-core effect. When the magnetic field is far from $\mathbf{B}_{cp}$, the transition is sensitive to the fluctuating magnetic fields, resulting in a short coherence time. There are hardly any spin flips of the Y^{3+} in the frozen core within the coherence-time period of the Eu^{3+} spins. Consequently, the main decoherence source is the small fluctuations caused by the rapid spin flips of Y^{3+} in the bulk, seen by the Eu^{3+} as a Gaussian-shaped homogeneous

broadening. As the field is aligned close to $\mathbf{B}_{cp}$, lowering the overall field sensitivity, these small fluctuations no longer strongly affect the Eu^{3+} . As a result, the coherence time becomes long enough for frozen-core Y^{3+} spin flips to affect the Eu^{3+} . This produces strong but slow magnetic field fluctuations on the Eu^{3+} ions, which gradually act as the dominating decoherence source, and make the transition broadening more Lorentzian.

In the final regime of the data, regime IV ($\gamma_d < 1.6 \text{ Hz/G}$), Γ_h of both samples saturates. There is an inhomogeneity of 1 G in the applied magnetic field, which was expected to accordingly limit the field alignment at a γ_d value of about 0.18 Hz/G. However, the saturation of both samples occurs much earlier, at $\gamma_d \approx 1.6 \text{ Hz/G}$ for the 100-ppm sample and $\gamma_d \approx 5 \text{ Hz/G}$ for the 1000-ppm sample (for which the saturation effect masks the narrowing effect in regime III). As the 1000-ppm sample saturates at a higher value and earlier than the 100-ppm sample, it can be attributed to the distribution of ZEFOZ fields among Eu^{3+} subgroups throughout the crystal, due to the concentration-dependent hyperfine inhomogeneous broadening. The measured hyperfine inhomogeneous broadening Γ_{inh} (HWHM) of the 1000-ppm sample is 18.0 kHz, much larger than the 6.0 kHz of the 100-ppm one, which implies a broader distribution of spin Hamiltonian parameters in the 1000-ppm sample and, accordingly, a larger distribution of ZEFOZ points. The shift in the Hamiltonian parameters, and therefore the ZEFOZ field associated with this line width, can be estimated assuming that it arises entirely from the quadrupole term in the spin Hamiltonian [13]. This estimation gives a ZEFOZ field range of 10 G for the 1000-ppm sample and 3 G for the 100-ppm one, respectively. This broader ZEFOZ field inhomogeneity for the higher-concentration sample implies that the ensemble coherence would decay faster and thus Γ_h would saturate at a higher value. Saturation will occur when the distribution of γ_d is larger than the mean γ_d value, which occurs earlier (at higher γ_d) for the 1000-ppm sample than for the 100-ppm one.

IV. TEMPERATURE-DEPENDENT DECOHERENCE

The TSE experiments above have been conducted at low enough temperatures that temperature did not limit the coherence decay [23]. In order to investigate how the decoherence process is affected by the temperature, we have measured the hyperfine homogeneous broadening in the 100-ppm and 1000-ppm samples over a temperature range from 80 mK to 17 K. Decoherence measurements have been conducted, respectively, using both TSE and DD sequences (the DD measurements are further detailed in Sec. V), with the results summarized in Fig. 3. We have also measured the lifetime and present $\Gamma_{T_1} = 1/(2\pi T_1)$ in the figure as a reference for the lower limit on Γ_h [24].

In the high-temperature regime, the spin-phonon interaction is strong and the coherence time is limited by the lifetime of the spin state. The lifetime-limited Γ_h is reached at 12 K here, while it has been observed at 19 K in a previous study performed at a magnetic field of 60 G [23]. We have reached the lifetime limit earlier because the ZEFOZ field suppresses other decoherence sources, leading to the lifetime dominating at a lower temperature. Indeed, an even lower lifetime-limit temperature of about 7 K has been reached when DD sequences were used to further suppress decoherence.

Between 2 K and their lifetime-limit temperatures, all the measured Γ_h are independent of temperature. This behavior is expected because all the transitions of the Eu^{3+} and Y^{3+} environment spins involved are less than 50 MHz (approximately 2.4 mK), much lower than the accessible phonon energy at the given temperature. However, at temperatures lower than 2 K, we have observed a decrease

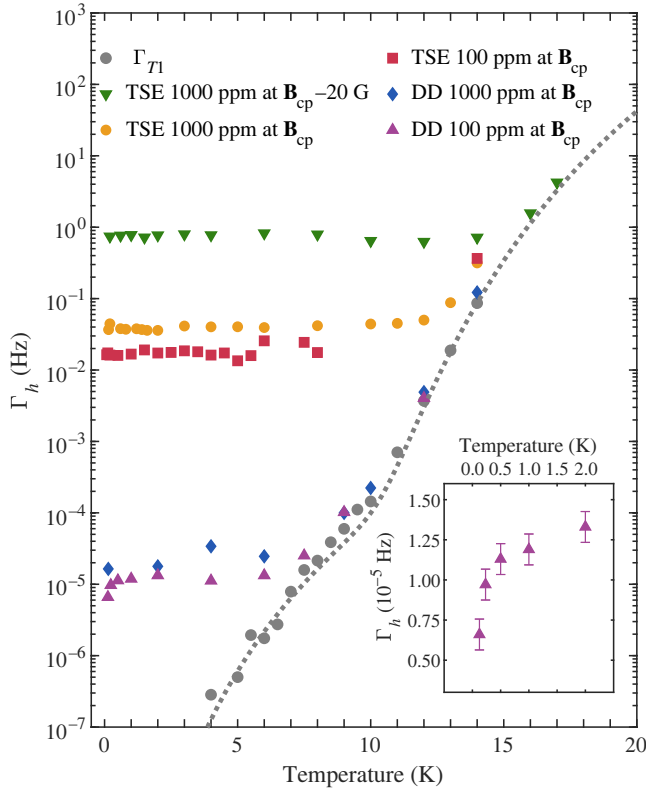


FIG. 3. The temperature-dependent homogeneous line width from the measured TSE coherence time T_M and the DD coherence time T_2 in the 100-ppm and 1000-ppm samples under different magnetic field conditions as labeled. The gray curve is a plot of the lifetime-limited $\Gamma_{T1} = 1/(2\pi T_1)$, derived from a fit to the measured T_1 using the model $T_1 = [A/(e^{\delta E/(kT)}) - BT^7]^{-1}$, where δE is the ${}^7F_0 \rightarrow {}^7F_1$ energy difference, A is the coefficient of the Orbach term and B is the coefficient of the Raman term [24]. The inset shows an enlargement of the temperature-dependent DD Γ_h in the ultralow-temperature regime for the 100-ppm sample, with error bars from fitting uncertainty.

in the DD Γ_h (increased T_2) in the 100-ppm sample with decreasing temperature. We attribute this to unidentified impurities with nonzero magnetic dipole moment. Such impurities lead to temperature-dependent spectral diffusion of the form $\Gamma_h \propto \text{Sech}^2[(\mu_{\text{eff}} B)/(2k_B T)]$ [17] and a fitting of Γ_h versus the temperature gives an effective magnetic dipole moment around $1.0 \mu_B$. A possible impurity candidate is Cr^{3+} ions, which have been identified using electron-paramagnetic-resonance (EPR) spectroscopy in Y_2SiO_5 crystals from the same manufacture at 100-ppm levels [25]. The dephasing contribution from these impurities must be faster than 30 Hz; otherwise, it would have been suppressed by the applied DD pulses ($\tau_c = 35$ ms).

In the 1000-ppm sample, such temperature dependence was not seen, as the line width saturated at a much higher temperature. Similar to the saturation behavior in the magnetic-field-dependent measurement, this can also be attributed to its broader inhomogeneous line width Γ_{inh} and, correspondingly, the broader inhomogeneity in the ZEFOZ field. Thus, the ensemble coherence decays rapidly before a temperature-dependent coherence-time extension can be observed. The same effect explains why the measured Γ_h of the 1000-ppm sample is larger than that of the 100-ppm one under all the magnetic field and temperature conditions.

V. LONG COHERENCE TIME

In order to further extend the TSE coherence time beyond the 44 s seen in Fig. 1(b), a series of DD sequences have been applied to the 100-ppm sample [26]. At the ZEFOZ point, the longest coherence times achieved have been 13.1 ± 1.1 h, 10.9 ± 0.9 h, and 12.9 ± 2.4 h, respectively, by using Carr-Purcell (CP) [27], Carr-Purcell-Meiboom-Gill (CPMG) [28], and KDD_x [29] sequences [Fig. 4(a)]. The temperature for these measurements was initially set at the base temperature of about 80 mK, but the equilibrium temperature increased to 125 mK with the heating effect from these DD pulses (Fig. 5). The variation in these DD T_2 values is probably due to magnetic field alignment uncertainty, magnetic field drift, and pulse errors in the DD pulses over the very long measurement time, rather than any differing effectiveness of the pulse sequences themselves. While these ultralong times do require millikelvin temperatures, a 6-h coherence time, matching the previous record, is available at temperatures up to 6 K [Fig. 4(b)]. The performance drops after this point, but even at 7.5 K, the coherence time exceeds 3 h.

Ideally, for the best decoupling effectiveness, we expect the DD pulses to have the same action on every individual ion in the ensemble. This would require the bandwidth of an applied pulse to be much larger than the transition inhomogeneous line width of the ensemble. However, due to the limited cooling capacity of the dilution refrigerator and imperfect impedance matching of the rf antenna, the

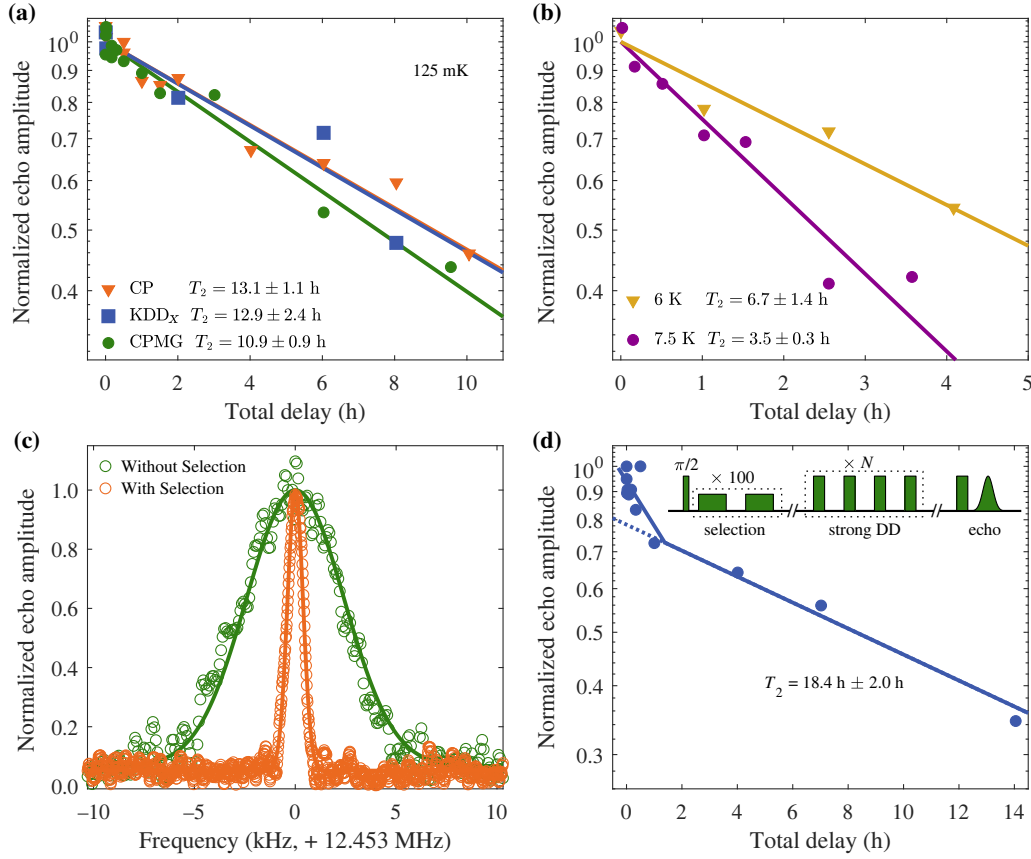


FIG. 4. The measured DD coherence times. (a) The measured echo decay of the 100-ppm sample at $\mathbf{B}_{cp}$ for the Carr-Purcell (CP), Carr-Purcell-Meiboom-Gill (CPMG), and KDD_x sequences. The temperature for these measurements was initially set at about 80 mK but the equilibrium temperature increased to 125 mK with the heating effect from these DD pulses (Fig. 5). (b) The measured echo decay of the 100-ppm sample at $\mathbf{B}_{cp}$ for the KDD_x sequence, with the temperature set at 6 K and 7.5 K. (c) A fast-Fourier-transformed (FFT) echo signal resulting from the KDD_x sequences and the corresponding Gaussian fitting. The orange and green curves correspond, respectively, to the echo signal with and without the preselection procedure at a delay of about 10 s. (d) The echo decay of the CPMG-selected KDD_x sequence inset with the applied rf pulses. There is an initial fast decay and the echo size reduced to about 75% with a time constant T_2 of 5.4 ± 2.6 h. There was then a long-term decay with a T_2 of 18.4 ± 2.0 h.

DD results have been obtained with a moderate rf Rabi frequency of about 3 kHz and a pulse separation τ_c of 35 ms in order to avoid heating up the system. Thus, this Rabi frequency is about half the measured line width of the hyperfine transition. In this situation, each subgroup in the ensemble has responded differently to the DD pulses and this leads to errors in the rephasing process, correspondingly undermining the DD effectiveness. To tackle this issue, we have first performed a frequency-selection process by applying a series of CPMG pulses for 10 s with a relatively low Rabi frequency of 0.6 kHz, with the pulse sequence as shown in Fig. 4(d). In Fig. 4(c), we show how the echo signal is narrowed through the selection process. As a result of these low-Rabi-frequency π pulses, the off-resonant subgroup ions have not been rephased and no contribution to the echo signal has been expected. After this process, DD has been applied, with pulses of higher Rabi frequency. The coherence measurement has

been conducted out to a delay of 14 h using the KDD_x sequence [Fig. 4(d)]. The measured echo amplitude has an initial fast decay of its size to approximately 75% for the first 1 h, with a coherence time of 5.4 ± 2.6 h. Then, there is a long-term decay with a coherence time of 18.4 ± 2.0 h. We attribute the initial fast decay to the nonideal selection.

During the DD measurement, we have tested the heating effect by applying either higher rf power or more frequent pulses in both samples and observed an obvious decline in the coherence time. This heating effect has been so substantial that the coherence times have decreased to less than half the maximum value when we have doubled the rf power or halved the pulse separation. While there is an increase in the equilibrium temperature of the mixing chamber associated with the rf, as shown in Fig. 5, this equilibrium temperature rise is small, nowhere near the 7 K needed to significantly affect the coherence time (from

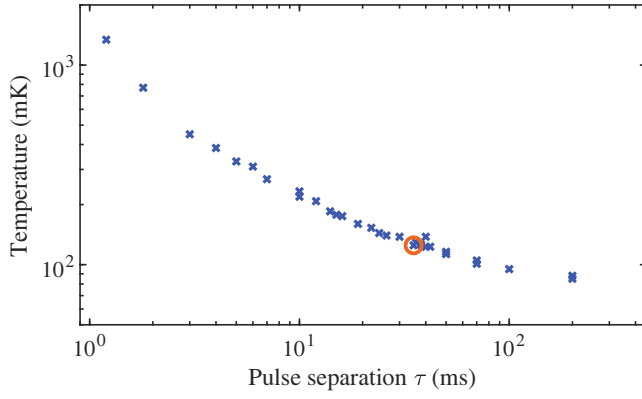


FIG. 5. The equilibrium temperature with an applied DD-pulse separation. The red circle labels the pulse separation applied in the coherence measurements for the present work.

Fig. 3). Instead, we attribute the decrease in the coherence time to a transient heating effect.

VI. DISCUSSION AND CONCLUSIONS

In this paper, we have studied the decoherence processes acting on a hyperfine transition in two $\text{Eu}^{3+}:\text{Y}_2\text{SiO}_5$ crystals for a magnetic field tuned around a ZEFOZ field. We have analyzed the Y^{3+} bath noise spectrum by investigating the scaling-law dependency of the Eu^{3+} homogeneous line width versus the transition sensitivity. The spectrum-diffusion process exhibits a Gaussian broadening profile when the magnetic field is far from $\mathbf{B}_{\text{cp}}$ but it converts to Lorentzian when the magnetic field is aligned approaching $\mathbf{B}_{\text{cp}}$. While in most cases, one needs to consider different ion species or crystals to analyze different diffusion models, here we demonstrate this unique possibility of being able to tune the coupling over a large range continuously, without changing the correlation time of the bath.

We have measured the ZEFOZ field coherence time for temperatures varying from 80 mK to 17 K and discussed the processes that limited the coherence times seen and what prospects exist for removing these limits. For the 100-ppm sample, we have observed hyperfine dephasing due to impurity spins for a temperature range from 2 K to 7 K. The EPR experiments could positively identify these impurities, informing efforts to remove them during the crystal-growth process. In the absence of these impurities, the dominant source of dephasing is the field inhomogeneity of the ZEFOZ point arising from the spin inhomogeneous broadening, which is 6 kHz for the 100-ppm sample and 18 kHz for the 1000-ppm sample. We have observed this limit here for both crystals below 1 K.

While the hyperfine inhomogeneous line width is linearly dependent on the dopant concentration, recent work has shown that other sources dominate the broadening at 100 ppm [30], so reducing the concentration further offers limited benefit (an approximately 30% reduction in line

width at 0 ppm). An alternative approach for ameliorating the effect of this broadening on the line width is to operate using a spectrally narrow subensemble selected from within the inhomogeneous line using hole-burning techniques, rather than the entire inhomogeneous line. Beyond reducing the ZEFOZ inhomogeneity, such a selection procedure has demonstrated additional benefits by improving the DD effectiveness. If a selection scheme works well to remove the distribution in the ZEFOZ significantly such that the magnet inhomogeneity of 1 G becomes the dominant limit of the field alignment, then a TSE coherence time of about 248 s is expected, which is more than 5 times the present result [Fig. 2(b)]. Based on a simple assumption that the DD technique can extend the TSE coherence time as it does in the present work, a DD coherence time of 73 h is expected. However, the selection technique or the lowering of the doping concentration will decrease the optical depth of the memory and thus the storage efficiency [24,31,32]. A low spectral density can be effectively compensated for by placing the system in an optical-impedance-matched cavity [33], although this does substantially increase the engineering complexity.

The preservation of quantum coherence over a period of 13 h allows the possibility of realizing entanglement distribution over a global scale by physically transporting the memory crystals. This is sufficient to allow a quantum memory to travel for several hours by road or air without significant decay of the quantum information if the considerable engineering challenges involved have been properly dealt with. It also opens up the possibility of sending the memory crystals to a place with different gravity, such as a spacecraft, for the testing of fundamental physics [34]. Moreover, the achieved long coherence time also allows the possibility of realizing very-long-baseline interferometry in the optical regime [8]. Further, we have demonstrated a coherence time of over 6 h at a temperature of 6 K, obtainable in single-stage cryocoolers, which are much more compact and transportable than research dilution or pumped liquid-helium cryostats, and which are, thus, more practical for a wider range of applications, including in satellites.

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APPENDIX A: MAGNETIC FIELD ALIGNMENT

In order to align the magnetic field to $\mathbf{B}_{\text{cp}}$, we have first measured the hyperfine splittings using a Raman heterodyne method, for magnetic fields in a three-dimensional array, and fitted the relative angle between the laboratory coordinate frame and the crystal coordinate frame using the spin Hamiltonian parameters [13]. We have then measured the TSE phase-memory time T_M for fields uniformly distributed around the calculated critical field point according to the results of the coarse alignment. We have used this uniform sampling data to train a machine-learning model that is deployed to optimize the coherence time [35]. Using a “query by committee” strategy, the active-learning method adopted needs much less time than other algorithms, such as the gradient-descent algorithm. Finally, we have carried out another uniform sampling about the field identified in the machine-learning step with the finest available magnetic field resolution (1 G). With these steps, we have achieved the optimized $T_M = 19.8 \pm 0.3$ s for the 1000-ppm sample and $T_M = 44.0 \pm 1.0$ s for the 100-ppm sample.

APPENDIX B: SENSITIVITY CALCULATION

The differential sensitivity

$$\gamma_d = |\vec{\gamma}_d| = |\vec{\gamma}_e - \vec{\gamma}_g| \quad (\text{B1})$$

and the average sensitivity

$$\gamma_A = |\vec{\gamma}_A| = |(\vec{\gamma}_e + \vec{\gamma}_g)/2| \quad (\text{B2})$$

of transitions “A” and “B” have been calculated using spin Hamiltonian parameters [9,13], where

$$\vec{\gamma}_a = \sum_{i=x,y,z} \frac{\partial E_a(\vec{\mathbf{B}})}{\partial B_i} \hat{e}_i \quad (\text{B3})$$

is the sensitivity vector of state a and $E_a(\vec{\mathbf{B}})$ is the energy of state a . The calculated results of transitions “A” ($| - 3/2 \rangle \leftrightarrow | + 3/2 \rangle$) and “B” ($| + 1/2 \rangle \leftrightarrow | + 3/2 \rangle$) are shown in Fig. 6. The average sensitivities of the two transitions are almost the same, since they are caused by the same quadratic Zeeman tensor [13].

The measured ZEFOZ point occurs at a magnetic field of $B_{\text{cp,exp}} = 1.3020$ T, slightly lower than the theoretical value $B_{\text{cp,th}} = 1.3480$ T. We have assumed that there is a calibration difference between the current system and the one in which the spin Hamiltonian was derived, and we

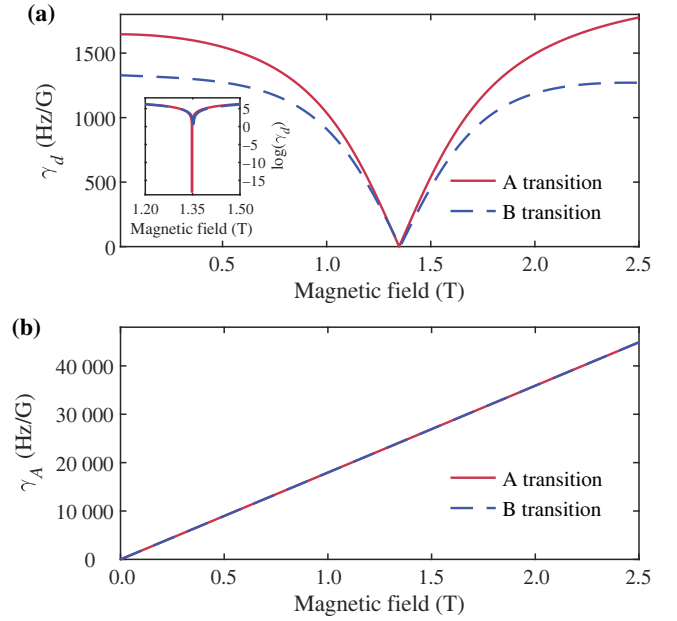


FIG. 6. The differential and average sensitivity of transitions “A” and “B” near the ZEFOZ point: (a) is differential sensitivity γ_d and (b) the average sensitivity γ_A of transitions “A” and “B,” respectively.

have taken the ratio $\kappa = B_{\text{cp,exp}}/B_{\text{cp,th}}$ into consideration in the numerical calculations. For a field close to $B_{\text{cp,exp}}$, we have treated the γ_d magnetic field as $\sqrt{B_{\text{cp,exp}} + \delta_B}$ in order to avoid a zero γ_d when doing the numerical calculations, where $\delta_B = 1.0$ G is the field resolution.

The transition undergoes a stochastic process of

$$\begin{aligned} \zeta(t) &= \vec{\gamma}_d \cdot \vec{\zeta}_B(t) = \sum_{i=x,y,z} \gamma_{d,i} \times |\vec{\zeta}_B|/\sqrt{3} \\ &\propto \gamma_d |\vec{\zeta}_B|, \end{aligned} \quad (\text{B4})$$

since $\vec{\gamma}_d$ of transitions “A” and “B,” as well as the anisotropy of $\vec{\zeta}_B(t)$, do not change direction for the field regime investigated in this work.

APPENDIX C: POWER-LAW SCALING

The decoherence of Eu^{3+} is dominated by the fluctuation of bath spins through magnetic dipole-dipole interactions. A quantitative understanding of the decoherence process requires details of the fluctuation of the Eu^{3+} transition frequency, which can be characterized with a broadened line shape $P(\omega)$, the probability density when the resonant frequency is ω , and with the diffusion kernel $P(\omega, t; \omega')$, the probability that the transition frequency is initially ω' and is ω at time t . There are two widely used models in solid-state systems [14,15,36], the Gaussian and Lorentzian spectral-diffusion models, with different assumptions on the line shape of the fluctuation-broadened

transition. In the Gaussian spectral-diffusion model, the fluctuation is assumed to be Gaussian, stationary, and Markovian. The line shape is given as

$$P_G(\omega) = \frac{1}{\sqrt{2\pi\sigma_\omega^2}} \text{Exp} \left[-\frac{(\omega - \omega_0)^2}{2\sigma_\omega^2} \right] \quad (\text{C1})$$

and the diffusion kernel is

$$P_G(\omega, t; \omega') = \frac{1}{\sigma_\omega \sqrt{2\pi(1 - e^{-Rt})}} \times \text{Exp} \left[-\frac{(\omega - \omega_0)^2}{2\sigma_\omega^2(1 - e^{-2Rt})} \right], \quad (\text{C2})$$

where $\sigma_\omega \propto \delta \propto \gamma_d$ characterize the spectral broadening of the Eu^{3+} transition, δ is the HWHM of the broadened line width, ω_0 is the central frequency, $R = 1/\tau_c$ is the effective fluctuation rate of the environmental noise, and τ_c is the correlation time of the correlation function

$$\langle \zeta(t)\zeta(t') \rangle = \sigma_\omega^2 e^{-|t-t'|/\tau_c}. \quad (\text{C3})$$

The two-pulse spin-echo evolution of Gaussian diffusion is calculated as [14,15,20,22]

$$M_G(t) = \text{Exp} \left\{ -\sigma_\omega^2 \tau_c^2 \left[\frac{t}{\tau_c} - 3 + 4e^{-t/(2\tau_c)} - e^{-t/\tau_c} \right] \right\} \quad (\text{C4})$$

Then the coherence time T_2 can be calculated with $M_G(t = T_2) = e^{-1}$. Similar to Ref. [37], the homogeneous line width is

$$\Gamma_h = 1/\left(\pi \mathcal{G} \left(\frac{1}{\sigma_\omega \tau_c} \right) \tau_c \right) \propto \kappa_G(\sigma_\omega \tau_c), \quad (\text{C5})$$

where

$$g\left(\frac{t}{\tau_c}\right) = \sqrt{\frac{t}{\tau_c} - 3 + 4e^{-t/(2\tau_c)} - e^{-t/\tau_c}} \quad (\text{C6})$$

and the inverse function of g is defined as $\mathcal{G}[g(x)] = x$ and

$$\kappa_G(\sigma_\omega \tau_c) = 1/\left(\pi \mathcal{G} \left(\frac{1}{\sigma_\omega \tau_c} \right) \right). \quad (\text{C7})$$

We assume that the correlation time τ_c is a constant in the regime that is close to the ZEFOZ point.

If the fluctuation is not Gaussian but is instead Lorentzian, the line shape is written

$$P_L(\omega) = \frac{\sigma_\omega}{\pi} \frac{1}{\omega^2 + \sigma_\omega^2} \quad (\text{C8})$$

and the diffusion kernel can be written as

$$P_G(\omega, t; \omega') = \frac{\sigma_\omega(1 - e^{-Rt})}{\pi} \times \frac{1}{(\omega - \omega_0 e^{-Rt})^2 + \sigma_\omega^2(1 - e^{-Rt})^2}. \quad (\text{C9})$$

Then, the echo decay function is calculated as [15]

$$M_L(t) = \text{Exp} \left\{ -\sigma_\omega \tau_c \left[\frac{t}{\tau_c} - 2\ln(2 - e^{-t/(2\tau_c)}) \right] \right\}. \quad (\text{C10})$$

With a definition of

$$l\left(\frac{t}{\tau_c}\right) = \frac{t}{\tau_c} - 2\ln(2 - e^{-t/(2\tau_c)}), \quad (\text{C11})$$

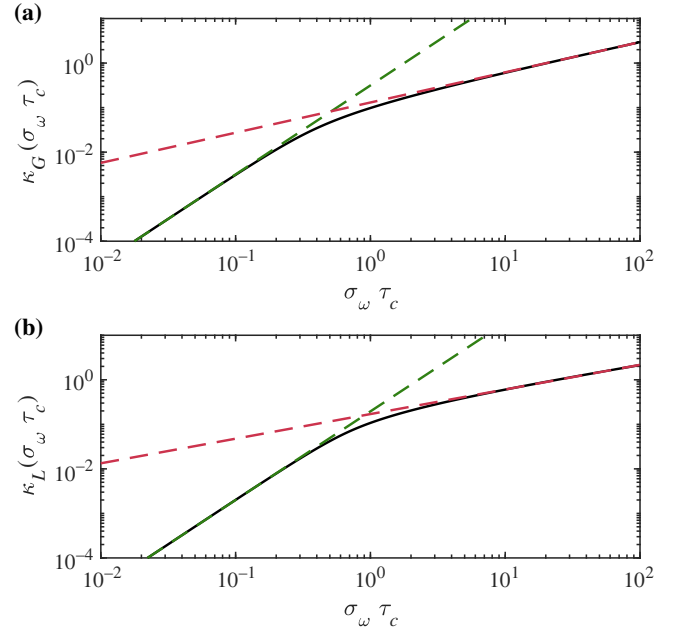


FIG. 7. Numerical calculations of the (a) Gaussian and (b) Lorentzian models, respectively: the numerically calculated inhomogeneous line width (with a numerical factor) (a) κ_G and (b) κ_L (black solid lines). There are two regimes with different power laws for both spectral-diffusion models. In the limit of large and small $\sigma_\omega \tau_c$, κ can be approximated by a power law $\kappa_{G,L} \propto (\sigma_\omega \tau_c)^\beta$, shown by red and green dashed lines, respectively. The fitted power indices are 0.67 and 2.00 for the Gaussian model and 0.50 and 1.00 for the Lorentzian one, in the large- and small- $\sigma_\omega \tau_c$ regimes, respectively.

the homogenous line width is

$$\Gamma_h = 1 / \left(\pi \mathcal{L} \left(\frac{1}{\sigma_\omega \tau_c} \right) \tau_c \right) \propto \kappa_L(\sigma_\omega \tau_c), \quad (\text{C12})$$

where $\mathcal{L}$ is the inverse function of l and

$$\kappa_L(\sigma_\omega \tau_c) = 1 / \left(\pi \mathcal{L} \left(\frac{1}{\sigma_\omega \tau_c} \right) \right). \quad (\text{C13})$$

The numerical-calculation results of $\kappa_{G,L}(\sigma_\omega \tau_c)$ are shown in Fig. 7. This figure shows that the homogenous line width obeys different power laws of $\Gamma_h \propto \gamma_d^\beta$ in the strong-coupling regime ($\sigma_\omega \tau_c \gg 1$) and in the weak-coupling regime ($\sigma_\omega \tau_c \ll 1$) for these two models. In Fig. 7, we have fitted the power-law exponent in the two limits, obtaining $\beta = 0.67$ and 2.00 for Gaussian spectral-diffusion model and 0.50 and 1.00 for Lorentzian spectral-diffusion model, in the strong-coupling and weak-coupling regime, respectively.

APPENDIX D: FIELD DISTRIBUTION AND FLIPPING RATE OF Y^{3+}

In Fig. 2(b), we show a conversion in the spectrum-diffusion process that agrees with the Gaussian strong-coupling ($\sigma_\omega \tau_c \gg 1$) model in regimes I and II and with the Lorentzian weak-coupling ($\sigma_\omega \tau_c \ll 1$) model in regime III. This conversion occurs at $\gamma_d = 20$ Hz/G and, as discussed in the main text, in the strong-coupling regime, the Eu^{3+}

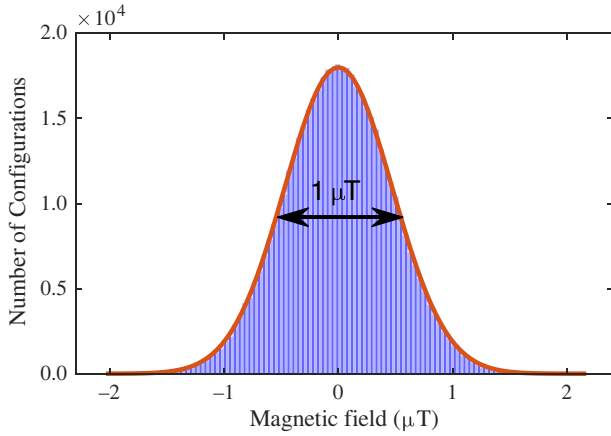


FIG. 8. The full width at half maximum (FWHM) line width of the field perturbation caused by the external bulk Y^{3+} . The calculation has been conducted using the Monte Carlo method, aiming to estimate the field perturbation from the external bulk Y^{3+} instead of the frozen-core Y^{3+} . The nearest 2000 Y^{3+} ions have been considered, but we have excluded the effect from the nearest 200 ions because they are included in the frozen core. This histogram of magnetic field values at the Eu site has been produced after 500 000 iterations of randomization of the Y^{3+} spin states.

hyperfine decoherence is mainly caused by the field fluctuations originating from the spin flips of the external bulk Y^{3+} instead of the frozen-core Y^{3+} . We can then calculate the effective spin flipping rate R of the host Y^{3+} , which is described as follows.

As shown in Fig. 8, the full width at half maximum (FWHM) line width of the field perturbation caused by the external bulk Y^{3+} has been calculated to be $\delta B_Y = 1 \mu\text{T}$ through a Monte Carlo method described in Ref. [38]. In this calculation, the nearest 200 Y^{3+} have been excluded because they were included in the frozen core based on the criterion that the Eu-Y magnetic dipole-dipole interaction strength is larger than that of Y-Y. The conversion of the coupling mode occurs at $\gamma_d = 20$ Hz/G and then the effective flipping rate has been estimated by making the equation $\sigma_\omega \tau_c = 1$, namely, $R = \gamma_d \delta B_Y = 0.2$ Hz, accordingly $\tau_c = 5$ s.

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