Sensing Individual Nuclear Spins with a Single Rare-Earth Electron Spin

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Rare-earth related electron spins in crystalline hosts are unique material systems, as they can potentially provide a direct interface between telecom band photons and long-lived spin quantum bits. Specifically, their optically accessible electron spins in solids interacting with nuclear spins in their environment are valuable quantum memory resources. Detection of nearby individual nuclear spins, so far exclusively shown for few dilute nuclear spin bath host systems such as the nitrogen-vacancy center in diamond or the silicon vacancy in silicon carbide, remained an open challenge for rare earths in their host materials, which typically exhibit dense nuclear spin baths. Here, we present the electron spin spectroscopy of single Ce$^{3+}$ ions in a yttrium orthosilicate host, featuring a coherence time of $T_2 = 124\ \mu$s. This coherent interaction time is sufficiently long to isolate proximal $^{89}$Y nuclear spins from the nuclear spin bath of $^{89}$Y. Furthermore, it allows for the detection of a single nearby $^{29}$Si nuclear spin, native to the host material with $\sim5\%$ abundance. This study opens the door to quantum memory applications in rare-earth ion related systems based on coupled environmental nuclear spins, potentially useful for quantum error correction schemes.

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Hybrid quantum systems, consisting of a read-out electron spin and long-lived nuclear spins, have demonstrated remarkable properties for quantum memory applications [1–3]. At the same time individually controllable, interacting spins enable active quantum processing used in, e.g., quantum error correction [4]. Implementing them in scalable quantum networks based on single rare-earth ions (REI) doped in solids potentially combines long distance entanglement distribution via single telecom band photons [5] with error corrected long-lived quantum memories.

Based on the efficient isolation of REI’s $4f$ electrons, their narrow and stable optical and spin levels have been used for demonstration of storage and retrieval of single photons [6] and exceptional coherence times [7], rendering them particularly suitable for quantum repeater protocols. With additional control over single rare-earth ions, however, these capabilities can be extended to high-fidelity spin readout and generation of entanglement of rare-earth electrons and nuclei in a scalable fashion [3]. Consequently, an increasing number of REIs are isolated as single emitters [5,8–11]. Based on ancillary electron spins of these single emitters, sensing of nuclear spins [12–16] is an important next step for REI-based quantum network applications. So far, only dilute nuclear spin bath host materials such as diamond [17], silicon carbide [18], and silicon [19] were successfully used for detection of individual nuclear spins.

In this study, we use the yttrium orthosilicate ($\text{Y}_2\text{SiO}_5$, YSO) crystal to investigate the nuclear environment of individual Ce$^{3+}$ electron spins, which simultaneously act as a proxy for other REI species, owing to their interchangeable doping into yttrium containing solids. We demonstrate spin initialization and coherent manipulation of Ce$^{3+}$ electron spins in a YSO host crystal. Surprisingly, we were able to sense individual dipolar coupled $^{29}\text{Si}$ nuclear spins despite the strong yttrium spin bath. $^{29}\text{Si}$ signal is distilled furthermore with basic decoupling sequences. Signatures of yttrium nuclear spins also reveal dipolar coupling with the nearby Ce$^{3+}$ superimposed on the yttrium spin bath. It is worth mentioning that the hyperfine structure of single REIs was already probed, and it was shown that the nuclear spin of REIs can be potentially used as a quantum memory [20–22]. Our findings, however, reveal another potentially useful memory resource, namely, nuclear spins of the surrounding host atoms similar to nitrogen-vacancy (N-V) center in diamond [23].

Trivalent cerium substitutes $\text{Y}^{3+}$ in 95% of the cases at the seven-oxygen-coordinated site of the YSO crystal with a $C_1$ symmetry [24]. The remaining 5% of Ce$^{3+}$ ions substituting $\text{Y}^{3+}$ in the six-oxygen-coordinated site of the crystal can be neglected from further considerations due to different optical (red shifted) and magnetic (different $g$ tensor) properties [24,25]. Individual Ce$^{3+}$ ions were identified in an ultrapure YSO crystal using laser scanning confocal microscopy. The experimental setup of the microscope is described in detail in the Supplemental Material [26]. To improve the collection efficiency and spatial
FIG. 1. (a) SEM image of SILs on a YSO crystal surface. The MW copper wire position is indicated by an orange stripe. (b) Electronic level structure of Ce$^{3+}$ ion in YSO crystal. Purple arrows indicate laser excitation and blue arrows fluorescence. Under circularly polarized excitation light, directed along the same axis as the applied external magnetic field (along the b axis of the crystal), different strengths of optical transitions between the lowest $4f$ level and the lowest $5d$ level are indicated by different widths of arrows. (c) Confocal scan of cerium centers under pulsed laser excitation, taken at the focus of an SIL. (d) Low temperature, high-resolution spectrum of 0.01% Ce$^{3+}$:YSO crystal in the vicinity of the zero-phonon line at 371 nm. Inset: typical spectrum of a 0.01% Ce$^{3+}$:YSO crystal at 8 K. The gap in the spectrum around 532 nm is an artifact of the used notch filter.

resolution of the microscope, solid immersion lenses (SILs) were fabricated on the surface of the sample by focused ion beam milling. A scanning electron microscopy (SEM) image of the sample with milled SILs and an indicated wire used for microwave (MW) spin manipulation is shown in Fig. 1(a). A picosecond pulsed laser at 355 nm wavelength was used to off-resonantly excite Ce$^{3+}$ ions from the $4f$ ground state into the excited $5d$ band based on their phonon-assisted absorption band [25]. The corresponding energy level structure of Ce$^{3+}$ in YSO is shown in Fig. 1(b). For the purpose of acquiring a fluorescence spectrum, $5d \rightarrow 4f$ emission was collected from a 0.01% doped Ce$^{3+}$:YSO crystal at $T \approx 8$ K [inset Fig. 1(d)]. A high-resolution spectrum of Ce$^{3+}$:YSO, shown in Fig. 1(d), reveals the zero-phonon line at 371 nm, characteristic for Ce$^{3+}$ ion fluorescence at cryogenic temperatures [31]. Performing confocal microscopy with the pulsed 355 nm laser on the SILs, however, allows for resolving individual Ce$^{3+}$ ions. A typical laser scanning fluorescence image of optically resolved Ce$^{3+}$ ions located in the focus of an SIL is shown in Fig. 1(c).

Even in ultrapure crystals (from Scientific Materials), cerium is an unavoidable native impurity for yttrium-based hosts and the estimated residual density of 0.3 ppb in our crystal is the main contribution to background signal and can result in more than one Ce$^{3+}$ ion to be probed within the focal volume. In principle, stimulated-emission-depletion-based superresolution microscopy is available for Ce$^{3+}$:YSO (see the Supplemental Material [26]) [32]; however, spin initialization and readout were found to be prevented by the high power depletion laser used in the experiment.

All spin manipulation experiments were conducted at cryogenic temperatures. In a magnetic field parallel to the optical excitation beam, optical transitions between $4f^1$ and $5d^1$ spin doublets show different relative strengths under $\sigma^-$ circularly polarized excitation [10]. While the $|4f^1 \downarrow \rangle \rightarrow [5d^1 \uparrow \rangle$ spin-flip transition exhibits the strongest optical dipole moment out of the four possible transitions under $\sigma^+$ circularly polarized excitation [indicated in Fig. 1(b)], the radiative decay originating from the excited $5d^1$ level ends up in both ground state spin levels with equal probability. A repeated excitation of the selectively driven spin-flip transition eventually results in a polarized ground state spin level. When applying the optical polarization procedure, the spin is pumped into the optically “dark” state [33], which results in a reduced fluorescence signal. MW radiation resonant with the ground $4f^1$ state spin transition can then be used to flip the optically polarized spin. This results in an increased fluorescence signal, which allows for optically detected magnetic resonance (ODMR) measurements on individual cerium ions.

Initialization measurements, shown in Fig. 2(a), capture the dynamic evolution of the fluorescence signal as a function of the laser pulse number. Starting with a thermal polarization, the fluorescence signal drops with increasing number of laser pulses because of optical pumping of the spin. Initialization signal contrast ranges between 5% and 15%, depending on the individual Ce$^{3+}$ ion under investigation. The relatively high background of densely packed Ce$^{3+}$ ions in the sample can contribute to measuring a decreased signal contrast.

In our experiments, a magnetic field with a strength of 970 G was applied parallel to the optical beam and the b axis of the YSO crystal, for which Ce$^{3+}$ has a $g$ factor of $g_{\text{Ce}} \approx 1.4$ and a magnetic resonance frequency of 1930.5 MHz [24]. MW structures created in proximity of SILs were then used to sweep the MW radiation frequency in order to observe ODMR spectra of individual Ce$^{3+}$ ions. A typical spectrum is shown in Fig. 2(b) and exhibits a linewidth of 2–3 MHz, slightly different for different cerium ions.

Coherent Ce$^{3+}$ spin manipulation is demonstrated in a measurement of spin Rabi oscillations under strong MW driving, shown in Fig. 2(c). A typical sequence for such a Rabi measurement contains spin initialization, control and readout, as shown in the inset of Fig. 2(c). An exponentially decaying sine function was fitted to the Rabi measurement.
signal, revealing a Rabi frequency of 5.6 MHz and a decay time of 2 μs.

A free induction decay (FID) experiment is capable to reveal the electron spin coherence in the thermal noise of the characteristic nuclear spin bath of YSO, featuring $^{89}$Y and $^{29}$Si nuclear spins. Figure 2(d) presents a typical FID of Ce$^{3+}$ and quantifies the inhomogeneous broadening of Ce$^{3+}$ spin transition to $T_2 = 310$ ns, by fitting with a Gaussian. These dephasing times are in good agreement with ODMR linewidth measurements. The main contribution to inhomogeneous broadening may emerge from the nuclear spin bath and can also come from short-pulsed optical excitation of Ce$^{3+}$, causing ionization of electron traps nearby the electron spin, which induces charge fluctuations capable of Stark shifting the resonance.

We measured the lifetime of the spin state $T_1$ after optical initialization by reading out the spin state after a variable waiting time $\tau$. The spin relaxation measurements are shown in Fig. 2(e) and reveal $T_1 = 610$ μs at a temperature of $T = 3.8$ K, measured at the heat exchanger of our cold-finger cryostat. At an indicated temperature of $T = 4.5$ K, the spin lattice relaxation measurement yields $T_1 = 280$ μs. By comparing our measured $T_1$ values with electron paramagnetic resonance-based values [34], we can identify the actual sample temperatures to be approximately 4 K higher than measured on the heat exchanger, because of insufficient cooling power.

The Hahn spin echo sequence decouples the spin from slow (compared to the waiting time $\tau$) changes in the environment and allows for more detailed spin spectroscopy of the nuclear spin environment of Ce$^{3+}$ electron spins. A representative Hahn-echo measurement on Ce$^{3+}$ is shown in Fig. 3(a). We observe periodic revivals related to yttrium ions in the crystal, which have 100% abundance of nuclear spin $I_Y = 1/2$ with magnetic moment $\mu_Y = -0.137\mu_N$, with $\mu_N$ as the nuclear magneton. The overall decaying signal corresponds to a decoherence time of $T_2 = 124$ μs and was fitted to exp$[-(2\tau/T_2)]$ [17]. The coherence of the electron spin is simulated by considering dipole-dipole interactions in a noninteracting nuclear spin bath and plotted as blue line in Fig. 3(a). For the Ce$^{3+}$ electron spin located in a complex nuclear environment such as the YSO matrix, the simulated behavior of its coherence under spin echo control agrees strikingly well with our experimental results. Details of the simulations are described in the Supplemental Material [26]. The fast Fourier transform (FFT) of the Hahn-echo signal [see Fig. 3(b)] reveals two particular facts. First, yttrium bath related signatures, found at the Larmor precession frequency $\omega_Y Larmor \approx 200$ kHz, similarly known from single N-V centers in diamond [17]. Additionally, we find frequency components related to weakly coupled yttrium nuclear spins based on magnetic dipole interaction with the electron spin [35]. According to our simulations, up to eight neighboring yttriums contribute to this class of dipolar coupled nuclear spins with coupling strengths of a few tens of kHz. Their expected coupling is listed in the form of the hyperfine tensor in the Supplemental Material [26].

Using confocal microscopy, we can study spatially resolved Ce$^{3+}$ ions and their unique environment. In the following, we show coupling between an individual Ce$^{3+}$ electron spin and one $^{29}$Si nuclear spin. For each silicon ion in the crystal lattice, there is a 4.7% natural abundance of $^{29}$Si isotope with a nuclear spin $I_{Si} = 1/2$ and
depends on the number of pulses and scales approximately with \( f/(N/2) \) at frequency \( f = 1/2\tau \) [36]. With an increasing number \( N \) of \( \pi \) pulses, the noise filter function causes the \(^{29}\text{Si} \) signature to be split in \( N \) coherence dips. This can be seen in the insets of Figs. 4(b)–4(d), where the \(^{29}\text{Si} \) signal is isolated by plotting the difference between signals from the two Ce\(^{3+} \) ions under investigation. Solid lines in Fig. 4 are simulated noise spectra of Ce\(^{3+} \) ions located in a specific nuclear spin environment. In order to account for differences in the depth of coherence dips between experiment and simulation, we phenomenologically add a relaxation and dephasing mechanism to the dynamics of \(^{29}\text{Si} \) (see [26]). One possible reason for a reduced depth of the coherence dip in the experiment is external noise for proximal \(^{29}\text{Si} \). Broadening mechanisms can be related to magnetic field fluctuations introduced by MW manipulation of Ce\(^{3+} \), the optical initialization as described for the N-V center system [37] or residual RE Kramers ion impurities in the crystal (such as Er\(^{3+} \) or Yb\(^{3+} \), acting as a noise source on a short timescale.

Based on our simulations, we can localize a coupled \(^{29}\text{Si} \) nuclear spin within the nearest neighbor position in the lattice (ion \( B \), magenta). Furthermore, the comparison spectrum without \(^{29}\text{Si} \) nuclear spin signatures (ion \( A \), blue) reveals information about the absence of \(^{29}\text{Si} \) within 6 Å distance from the Ce\(^{3+} \) ion. The probability to find a Ce\(^{3+} \) ion with the same nuclear spin environment as ion \( A \) in YSO is \( \sim 70\% \). In conclusion, we show coherent control of an individual Ce\(^{3+} \) electron spin in a YSO matrix. Using spin decoupling techniques, our spin spectroscopy reveals the single REI electron spin to be dipolar coupled to nearby nuclear spins. Based on high density of yttrium in the host crystal, the future challenge is to distinguish spectrally between individual coupled nuclear spins. Carefully designed DD sequences can improve detection of nuclear spin signals, which tend to be hidden by the noisy spin bath [38]. \(^{29}\text{Si} \) nuclear spin sensing was demonstrated, for \(^{29}\text{Si} \) being located within the nearest neighbor shell.

Nuclear spins (either \(^{29}\text{Si} \) nuclear spin or yttrium nuclear spin) in proximity to the Ce\(^{3+} \) electron spin are quantum resources for quantum memory protocols. By establishing polarization transfer techniques, for example, based on Hartmann-Hahn double resonance techniques [39], single \(^{29}\text{Si} \) nuclear spin could be initialized and potentially used as memory. A key concern is the coherence time of nuclear spins in this context. Decoherence caused by the nuclear spin bath will set the ultimate limitation for the coherence time. Because of weak dipolar interaction between nuclear spins in YSO compared to the Zeeman energy at applied magnetic fields, only the pure-dephasing interaction can have significant effect on the decoherence [40]. The characteristic decoherence timescale can be estimated by the nuclear spin dipolar interaction and gives \( T_2 \sim 10 \text{ ms} \) for \(^{29}\text{Si} \) nuclear spin and \( T_2 \sim 50 \text{ ms} \) for yttrium nuclear spin.

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**FIG. 3.** (a) Hahn-echo measurement shows collapse and revivals. The decay is fitted to \( \exp\left(-\left(2\tau/T_2\right)^3\right) \) (gray dashed line), with \( T_2 \approx 124 \mu s \), plotted offset on the intensity axis to envelope the signal. Simulated coherence is plotted as blue continuous line. (b) FFT of the Hahn-echo signal (orange) and FFT of the simulated Hahn-echo signal (blue). Highlighted in orange are the contributions originating from dipolar coupled yttrium nuclear spins in close proximity to Ce\(^{3+} \) [35].

\[ \mu_Y = -0.555\mu_N. \] For a Ce\(^{3+} \) ion under investigation, this leads to a chance of approximately 20% to have a \(^{29}\text{Si} \) as nearest neighbor. At the nearest neighbor location, the close distance (≤6 Å) between electron spin and \(^{29}\text{Si} \) nuclear spin leads to a detectable hyperfine coupling based on magnetic dipole interaction (tens to a few hundreds of kilohertz, see [26]), superimposed on the hyperfine interaction with close by yttrium spins and the yttrium bath [schematically shown in Fig. 4(a)].

Dynamic decoupling (DD) of the Ce\(^{3+} \) spin from the nuclear bath allows to extract \(^{29}\text{Si} \) related signatures. Carr-Purcell-Meiboom-Gill (CPMG) control sequences were used to acquire noise spectra for two different Ce\(^{3+} \) ions, as shown in Figs. 4(b)–4(d). The center of the coherence dip [in Fig. 4(b) and inset] at \( \tau_{\text{ddp}} \approx 600 \) ns corresponds to a revival time of \( \tau_r = 2\tau_{\text{ddp}}. \) 1/\( \tau_r \) matches well with the gyromagnetic ratio \( g_{\text{SI}} = -8.465 \text{ MHz/T} \), thus confirming the nearby \(^{29}\text{Si} \) nuclear spin. Specifically, CPMG-\( N \) decoupling sequences were used, with \( N = 1, 2, 5 \) denoting the number of \( \pi \) pulses used in the sequences. The bandwidth

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**FIG. 4.** (a) Hahn-echo measurement shows collapse and revivals. The decay is fitted to \( \exp\left(-\left(2\tau/T_2\right)^3\right) \) (gray dashed line), with \( T_2 \approx 124 \pm 5 \mu s \), plotted offset on the intensity axis to envelope the signal. Simulated coherence is plotted as blue continuous line. (b) FFT of the Hahn-echo signal (orange) and FFT of the simulated Hahn-echo signal (blue). Highlighted in orange are the contributions originating from dipolar coupled yttrium nuclear spins in close proximity to Ce\(^{3+} \) [35].
Finally, this work motivates the realization of controllable multispin quantum registers based on single REIs embedded in the YSO matrix. Access to local nodes based on environmental spins, as demonstrated in [4], provides functionality of quantum memories, such as error correction. Furthermore, presented findings are applicable to other Kramers ions doped into YSO, such as erbium, for which coherent spin control and readout were demonstrated recently [41].

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[21] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.124.170402 about (1) laser scanning confocal microscope setup, (2) super-resolution microscopy experiments, (3) Hahn-echo measurement sequence, (4) performed simulations, and (5) coupling between cerium electron spin and proximal nuclear spins, which includes Refs. [27–30].

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