Ultra-narrow optical linewidths in rare-earth molecular crystals

Rare-earth ions (REIs) are promising solid-state systems for building light–matter interfaces at the quantum level. This relies on their potential to show narrow optical and spin homogeneous linewidths, or, equivalently, long-lived quantum states. This enables the use of REIs for photonic quantum technologies such as memories for light, optical–microwave transduction and computing. However, so far, few crystalline materials have shown an environment quiet enough to fully exploit REI properties. This hinders further progress, in particular towards REI-containing integrated nanophotonics devices. Molecular systems can provide such capability but generally lack spin states. If, however, molecular systems do have spin states, they show broad optical lines that severely limit optical-to-spin coherent interfacing. Here we report on europium molecular crystals that exhibit linewidths in the tens of kilohertz range, orders of magnitude narrower than those of other molecular systems. We harness this property to demonstrate efficient optical spin initialization, coherent storage of light using an atomic frequency comb, and optical control of ion–ion interactions towards implementation of quantum gates. These results illustrate the utility of rare-earth molecular crystals as a new platform for photonic quantum technologies that combines highly coherent emitters with the unmatched versatility in composition, structure and integration capability of molecular materials.

REI-doped materials are promising systems for optical quantum technologies. At cryogenic temperatures, REIs doped into high-quality bulk single crystals, such as Y2SiO5, show exceptionally narrow optical homogeneous linewidths, equivalent to long quantum coherence lifetimes, suitable for building quantum light–matter interfaces. Moreover, REIs can present optically addressable electron and/or nuclear spin degrees of freedom that can be leveraged to efficiently store and process quantum information. These unique properties in the solid state have been used to demonstrate quantum memories for light, light–matter teleportation, and frequency- and time-multiplexed storage. REI-doped crystals are also being actively investigated for optical to microwave conversion and quantum processing. Besides experiments on high-quality bulk single crystals, strong efforts have recently been launched towards combining REIs with nanophotonic structures. This has enabled single-REI detection and control, fast spontaneous-emission modulation, lifetime-limited single-photon emission and on-chip optical storage. Further developments of these exciting topics are, however, impeded by the difficulty to nanofabricate crystalline host materials that preserve REIs’ quantum properties for integration into high-quality nanophotonic devices. This difficulty is faced by many solid-state systems, although promising results have recently been reported (for example, with colour centres in diamond).

Molecular chemistry is very attractive in this respect because it offers unmatched flexibility in terms of material composition, fine structural tuning and integration into photonic structures, as demonstrated by numerous results obtained with single organic molecules embedded in crystalline host lattices. However, most organic molecules studied to date lack a spin degree of freedom. Recently, optically addressable molecular spins were demonstrated, although with limited optical coherence, hindering their use as coherent spin–photon interfaces. Narrow homogeneous linewidths are in effect key to high-fidelity optical spin control in quantum processors and high-efficiency light-to-spin transfer in long-storage-time quantum memories. Here we introduce REI molecular crystals containing trivalent europium that exhibit optical homogeneous linewidths between 5 and 30 kHz, 3 to 4 orders of magnitude narrower than those of any molecular system. This allows us to efficiently exploit the nuclear spin degree of freedom by demonstrating >95% spin initialization into a single level, and coherent optical storage using atomic frequency combs (AFCs). We also harness the narrow optical linewidth of Eu²⁺ to demonstrate ion–ion interactions, which are the basis for high-bandwidth two-qubit quantum gates.

The molecular crystal is composed of a mononuclear Eu²⁺ complex (Eu(BA),(pip)), in which BA and pip stand for benzoylacetonate.
transitions (see inset). All of the following results were obtained on the 7F0 → 5D0 transition metal ions, showing persistent optical properties, and no photodegradation was observed at low temperature or under high laser intensity (Extended Data Fig. 2).

Figure 1 shows the emission spectrum of the Eu3+ complex in a crystalline powder with grain sizes of at least 50 nm (see Supplementary Section 1). The Eu3+ complex showed high physical and chemical stability over time. Repeated cooling cycles had no noticeable effects on optical properties, and no photodegradation was observed at low temperature or under high laser intensity (Extended Data Fig. 1). Grey, carbon; white, hydrogen; pink, europium; red, oxygen.

In Extended Data Fig. 1, grey, carbon; white, hydrogen; pink, europium; red, oxygen. b, Eu3+ PL spectrum showing characteristic 7D0 → 7F(j = 0–4) transitions (see inset). All of the following results were obtained on the 7F0 → 5D0 transition metal ions, showing persistent optical properties, and no photodegradation was observed at low temperature or under high laser intensity (Extended Data Fig. 2).

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For clarity, it is referred to as the Eu3+ complex hereafter. The complex crystallized in the space group, belonging to the monoclinic crystal system, with a crystal lattice composed of anionic [Eu(BA)3]1 and cationic piperidin-1-ium units (Extended Data Fig. 1, see Supplementary Section 1 for more details). The all-optical coordination environment around the Eu3+ centre is best described as a biaugmented trigonal prism with the help of a continuous shape measure calculation. The point group symmetry around the Eu3+ centre is assigned to C2v, as inferred from the continuous shape measure calculation and luminescence spectrum depicted in Fig. 1b (see Supplementary Section 2). All experiments discussed in the following were performed on microcrystalline powders with grain sizes of at least 50 nm (see Supplementary Section 1). The Eu3+ complex showed high physical and chemical stability over time. Repeated cooling cycles had no noticeable effects on optical properties, and no photodegradation was observed at low temperature or under high laser intensity (Extended Data Fig. 1).

Figure 1b shows the emission spectrum of the Eu3+ complex in which the characteristic lines of trivalent europium are observed. We focused on the 7F0 → 5D0 transition at 580.3778 nm (vacuum) as a result of the stoichiometric composition of the molecular crystal and light scattering inside the powder. The 7F0 → 5D0 oscillator strength is estimated to be 6.7 × 10−17 cm (see Supplementary Section 4.2). Fluorescence decay experiments led to a lifetime value T1,exp of 540 μs for the 5D0 level (Fig. 1d). This sets a limit of only 1/2πT1,exp = 295 Hz on the optical homogeneous linewidth (Γh).

The photon echo (PE) technique, similar to Hahn’s spin echo experiment, enables accurate assessment of narrow homogeneous linewidths2,24. Owing to its high sensitivity, it can be applied to powders25, allowing us to measure the optical coherence lifetime, or quantum-state lifetime of the 5D0 → 7Fj transition in the Eu3+ complex (Fig. 2a). Γh = 1/πT1,exp was found to be equal to 30.2 ± 0.6 kHz, markedly lower than values in the tens of megahertz range reported for transition metal ions26, a previously reported Eu3+ complex and single molecules27, although in the last case, lifetime-limited linewidths have been observed. The Eu3+ linewidth in the complex is indeed comparable to those measured in 0.3% Eu3+:Y2O3 nanoparticles14 and even in some bulk stoichiometric crystals such as EuP5O14 (ref. 27), although still broader than the narrowest reported linewidths28. Supplementary Table 12 gathers data for a range of materials for a more comprehensive comparison. Line broadening was further explored using stimulated PEs, showing a increase in Γh of ≈105 kHz for 1 ms delay (Fig. 2b and Supplementary Section 4.2).

Insights into dephasing mechanisms occurring in the complex were also obtained from the evolution of Γh with temperature T, measured by two-pulse PE and spectral hole burning (SHB; Fig. 2c). Two regimes were identified: for temperatures below 3.5 K, dephasing is dominated by coupling to two-level systems (TLSs) and Γh increases linearly with T (Fig. 2c, inset)29; above 3.5 K, the exponential increase of Γh is attributed to quasi-localized low-frequency modes (LFMs)28. Data were modelled...
and 160 ground-state a series of diluted crystals. The non-exponential decays may indicate 2 = 10.5 ± 0.2 μs or exponential fit to data giving α using the expression Γ TLS = 15 ± 5 kHz K−1, as observed in some REI-doped crystals28, and an LFM transition energy Δ E LFM = 580 GHz (19 cm−1), in the range reported for single molecules29. The homogeneous linewidth extrapolated to 0 K (Γ0) is estimated at ± 5 ± 4 kHz. This remaining dephasing could be due to Eu3+–Eu3+ interactions22, as the crystal has a high Eu3+ concentration, C Eu = 9.6 × 1020 ions per cm3. To investigate this effect, an optically inactive REI, Y3+, was introduced into the crystal to reduce C Eu. Figure 2d shows two-pulse PE decays in a series of diluted crystals. The non-exponential decays may indicate distinct environments for the Eu3+ ions in these complexes. Increasing coherence lifetimes with decreasing C Eu are observed, especially for the long decay components (Fig. 2d, inset), confirming a contribution of Eu3+–Eu3+ interactions to dephasing. At the highest dilution investigated here (95%; that is, 5% Eu3+ content), we found a 1/e decay time 2 = 25 μs (that is, 2 = 12.7 kHz). Remarkably, in this sample, PEs could be detected even after evolution times of 300 μs, corresponding to a long decay component of 2 = 68 ± 4 μs (2 = 4.6 ± 0.2 kHz) (inset of Fig. 2d and Extended Data Fig. 3).

Narrow optical linewidths are key to efficiently address REI spins, which enabled detailed investigations of Eu3+ nuclear spin states in the complex. First, the nuclear spin structure was determined for the 5D0 excited state and 7F0 ground state by SHB in the stoichiometric complex, which is isotopically enriched in 153Eu3+. At zero magnetic field, it consists of three doubly degenerate levels split by quadrupolar interaction22, giving rise to a complex SHB spectrum after optical pumping. Thanks to the narrow optical linewidth, well-resolved spectral features could be recorded and analysed (Fig. 3a). This allowed us to assign ground- and excited-state splitting energies (Fig. 3b, left), and transition branching ratios between nuclear spin levels (Supplementary Fig. 13). The same study was performed on a sample with natural abundance, from which splitting energies and transition branching ratios were determined for the 153Eu3+ isotope (Supplementary Fig. 13). Notably, the obtained branching ratios show the existence of efficient quasi-localized LFMs (see text and Supplementary Section 4.3). Inset: zoom on lower temperature range. d. PE decays from Y3+-diluted complexes showing 2 increase with decreasing Eu3+ concentration. Red line: single (non-diluted crystals) and double (diluted crystals) exponential decay fits to data. Inset: 1/e echo amplitude decay time 2 (black dots) and long 2 component from double exponential fits (blue dots) as a function of Eu3+ concentration. Error bars in b, c and the inset of d represent one standard deviation of uncertainty.
spin levels and transfers population into \(|±1/2⟩_g\) (right panel of Fig. 3b and Supplementary Fig. 14). As displayed in Fig. 3d, nearly full transparency could be induced at \(v_1\) and \(v_2\), which translates to initialization of 95% of the spin population into a single level. This also proves that efficient spectral tailoring is possible in the Eu\(^{3+}\) complex, an essential feature for many quantum memory and processing protocols based on REIs\(^{5,12}\).

We next used the narrow optical linewidths of Eu\(^{3+}\) molecular crystals to demonstrate coherent optical storage and controlled ion–ion interactions. In the first case, we used the AFC protocol\(^{12}\) to store a light pulse in the molecular crystal. This protocol enables quantum memories based on an ensemble of ions with very low output noise and multiplexed storage, important assets for long-distance quantum communications. We first made use of the efficient optical pumping achieved in the Eu\(^{3+}\) complex to create an AFC spanning a 6 MHz range, with three 0.9-MHz-wide teeth separated by 1.75 MHz (Fig. 4a). A storage experiment was then performed with a 0.15-μs-long input pulse overlapping the AFC structure. As shown in Fig. 4b, the output pulse is observed at a delay \(t_s = 0.57 \mu s\) after the partially transmitted input pulse, in perfect agreement with the tooth spacing as \(1/t_s = 1.75 \text{ MHz}\) (ref. \(^{12}\)). By varying the tooth spacing and adapting the input pulse length to keep an identical spectral overlap with the comb, output pulses were observed up to about 1 μs storage time, clearly confirming the AFC process (Fig. 4b, inset). Storage efficiency, defined as the ratio between input and output pulse intensities, was 0.86% for a storage time \(t_s = 0.57 \mu s\), in agreement with theory (see Supplementary Section 6.1). This efficiency could be boosted up to 100% in an optical cavity\(^{31}\). This could be achieved by crystallizing the Eu\(^{3+}\) complex directly on a cavity mirror. Storage times up to several tens of microseconds could also be obtained by creating narrower teeth\(^{12}\) with a suitable laser, given the narrow homogeneous linewidths and limited spectral diffusion in the Eu\(^{3+}\) complex.

We finally investigated controlled interactions between Eu\(^{3+}\) ions. The scheme we used is based on the difference in permanent electric dipole moments between Eu\(^{3+}\) ground \((7F_0)\) and excited \((5D_0)\) states\(^{32}\). This difference occurs when Eu\(^{3+}\) ions sit in a low-symmetry site, which is the case in our molecular crystal (\(C_{3v}\) site symmetry). When a control ion is excited, the electric field it produces changes, causing a shift in transition frequency for a nearby target ion through the linear Stark effect. This mechanism is the basis for two-qubit gates and qubit read-out in some REI-based quantum computing proposals\(^{4}\). However, to be useful, ion–ion interactions must be significantly larger than the optical homogeneous linewidth, a condition well matched by our highly concentrated Eu\(^{3+}\) complex with 30 kHz linewidth.

Target and control ions were chosen at different frequencies within the absorption line (Fig. 1c) to allow for independent excitation and monitoring. As a result of the distribution of distances and orientations and therefore interaction strengths between Eu\(^{3+}\) ions, the excitation of control ions results in an additional line broadening \(Γ_c\) for target ions, as detailed in Supplementary Section 6.2 (ref. \(^{33}\)). \(Γ_c\) can be conveniently monitored by measuring the amplitude of a PE produced by target ions while an extra pulse excites the control ions (Supplementary Fig. 15b).
amplitude of target ions (at frequency $\nu_a$) as a function of evolution time $t_{\text{evol}}$ after excitation of control ions (at $\nu_c = \nu_a \pm 20$ MHz and $p = 0.16$; see below). Red line: exponential fit to data giving a broadening of the target ions’ linewidth by the control ions of $\Gamma_c = 14.5 \pm 1$ kHz (see text). d, PE amplitude of target ions as a function of control ions’ excitation probability $p$, normalized to the maximum value reached experimentally ($t_{\text{evol}} = 3.5$ μs). Red line: exponential fit. e, PE amplitude of target ions as a function of control pulse detuning ($p = 0.16$, $t_{\text{evol}} = 17$ μs). All experiments were performed at 1.45 K in the stoichiometric $^{151}$Eu$^{3+}$-enriched complex. Error bars represent one standard deviation of uncertainty.

The Eu$^{3+}$ complex investigated in this study is a very promising platform for optical quantum technologies as a robust system with narrow linewidth and long-lived optically addressable spins. Furthermore, a significant decrease in optical homogeneous linewidth could be obtained by lowering the temperature, tuning the REI concentration and optimizing synthesis to decrease residual defects or impurities. In such compounds, interactions with host nuclear spins such as $^{1}$H or $^{13}$C could become the dominant dephasing mechanisms, which can be reduced using specific isotopes with lower or zero magnetic moments. This could also be useful to obtain spin states with long coherence lifetimes, a property that remains to be measured in our samples. The huge possibility in molecular design also opens the way to accurate engineering of the ligand field acting on europium and more generally other REIs of interest for quantum technologies, such as Er$^{3+}$ or Yb$^{3+}$. This would allow optimization of transition strengths and frequencies for optimal coupling to light, tuning of electronic and spin level structures, and coupling to molecular vibrations for long coherence lifetimes. Complexes containing several REIs at close distance can also be synthesized, which would enable high-density optically addressable qubit architectures, as shown with REI magnetic molecules in the microwave range.

We first investigated the effect of changing the evolution time ($t_{\text{evol}}$) between the control pulse and the echo, as shown in Fig. 4c. In this case, the echo amplitude varies as $\exp(-\pi\Gamma_c t_{\text{evol}})$ (Ref. 35). A fit to the experimental data yields $\Gamma_c = 14.5 \pm 1$ kHz, in qualitative agreement with expected Eu$^{3+}$–Eu$^{3+}$ electric dipole interactions (see Supplementary Section 6.2) and previous experiments in non-molecular REI-doped crystals. It also provides an upper bound to the ion–ion interaction contribution in the measurements of $\Gamma_c$ by two-pulse PEs (see Supplementary Section 4.3). We further confirmed this analysis by varying the control pulse intensity $I_c$. In the weak excitation regime, $\Gamma_c$ is proportional to the fraction of excited control ions ($p$), with $p \approx I_c$. We indeed observed the predicted exponential decay of the echo amplitude with increasing $p$, as displayed in Fig. 4d. Finally, the control pulse frequency was varied over several tens of megahertz with fixed $t_{\text{evol}}$ and $p$. No significant change in echo amplitude was observed, ruling out a direct light-induced frequency shift of target ions, the so-called a.c. Stark shift (Fig. 4e). We therefore conclude that qubit gate and readout schemes based on electric dipole–dipole interactions could be implemented in the molecular crystal. As a result of the high Eu$^{3+}$ concentration and narrow linewidth, we estimate that one ion could control thousands of target ions, while the large inhomogeneous to homogeneous linewidth ratio ($2.2 \times 10^6$) enables individual addressing of a large number of single ions (see Supplementary Section 6.2). These are useful properties for scaling up REI-based quantum processors.
facilitated by the broad range of synthesis, functionalization and deposition methods that are available for molecular complexes6,10,34, as well as the scalable production of large amounts of identical molecules. REI molecular crystals could therefore emerge as a versatile platform for highly coherent light–matter–spin quantum interfaces for developing applications in quantum communications and processing and fundamental studies in quantum optics.

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Methods
Preparation of the microcrystalline powder form of the Eu⁺ complex
The europium complex discussed in this study was prepared in several forms including a stoichiometric natural abundance version, a series of yttrium-diluted versions and an isotopically (151Eu⁺) enriched version. This was achieved by using a slightly modified procedure to that reported by Melby et al.20. In a typical procedure, 1-phenylbutane-1,3-dione, hereafter referred to as ligand, (1.3 g, 8 mmol) was added to 20 ml of ethanol at 75 °C and dissolved by stirring. To the ligand solution, piperidine (0.8 ml, 8 mmol) was added, and the solution was stirred for 15 min. To the pale-yellow solution of the deprotonated ligand, EuCl₃·6H₂O (732 mg, 2 mmol) dissolved in 10 ml water was added. After the completion of the addition, heating was switched off, and the reaction mixture was stirred at room temperature for 24 h, yielding a half-white crystalline precipitate of the complex. The precipitate was filtered, washed with 50 ml ethanol several times, and dried under reduced pressure overnight. See Supplementary Section 1 for more details, including X-ray analyses and discussions. All optical experiments reported in the main text were performed on either the stoichiometric isotopically enriched complex or the Y⁺-diluted complexes.

Low-temperature optical spectroscopy setup
Optical absorption, photoluminescence (PL) decays, PE and SHB measurements were performed in a He bath cryostat (Janis SWT-200) under resonant excitation of the Dᵡ → Fᵩ transition of Eu⁺. The excitation source was a tunable continuous-wave dye laser (Sirah Matisse DS) with a 300 kHz linewidth. Temperature-dependent measurements were performed by tuning the He gas pressure (T < 4 K) and using a built-in resistance as a heater (T > 4 K). The temperature of the sample holder was monitored with a Si diode (Lakeshore DT-670). Pulse sequences were created using an acousto-optic modulator (AA Optoelectronic MT200-B100A0, 5-VIS, 200 MHz central frequency), in the double-pass configuration, driven by an arbitrary waveform generator (Agilent N8242A) with a 625 megasamples per second sampling rate.

The crystalline powders were placed in a home-built sample holder made of brass. This holder is composed of five individual sample containers that have front and rear optical access through glass windows (Supplementary Fig. 8). Each container was filled with 5 mg of powder, forming slabs of about 500 μm in thickness. The excitation beam was focused on the containers’ front openings by a 75-mm-focal-length lens placed in front of the cryostat window. The excitation beam and lens were vertically and/or horizontally translated to study one sample after another during measurements. Light scattered by the powders and transmitted through the back openings was collected by a lens sitting outside the cryostat as shown in Supplementary Fig. 9. Signals were detected with avalanche photodiodes (Hammamatsu C5460 with 10 MHz bandwidth for SHB, and Thorlabs 110 A/M with 50 MHz bandwidth for all other measurements).

PL measurements at 15 K were carried out for the isotopically purified ¹⁵¹Eu⁺ complex using a closed-cycle cold-finger cryostat. The Dₓ level was resonantly excited at 580.4 nm with a tunable optical parametric oscillator pumped by a Nd³⁺ YAG Q-switched laser (Ekspla NT342BSH, 6 ns pulse length and 10 Hz repetition rate). Spectra were recorded using an Acton SP2300 spectrometer equipped with an holographic grating with 1,200 grooves per mm and an ICCD camera (Princeton Instruments). PL decays were recorded at low temperature for the ¹⁵¹Eu⁺ complex (Fig. 1d) and Y⁺-diluted complexes after a single pulse (1 ms long), under resonant excitation of the Dₓ level, yielding comparable decay curves and population lifetimes (Tₜ₁/₂ = 540 μs) for all samples.

PE and SHB measurements
Echo amplitudes from two-pulse and three-pulse PE experiments were detected through the fast Fourier transform of the beating between the PE signal and a frequency-detuned laser pulse (heterodyne pulse, with detuning equal to 30 MHz). During measurements, the laser wavelength was scanned over 500 MHz in 1 s to prevent echo signal loss due to SHB. The length and intensity of the π/2 and π pulses were optimized to obtain maximum PE amplitudes, with typical pulse lengths between 1 and 2 μs, and a laser power before the cryostat of 300 mW. A short-pass filter was set in front of the avalanche photodiode detector to reject the strong PL emission in the Eu⁺ complexes. The echo amplitude obtained by fast Fourier transformation was averaged over 50 sequences to improve signal-to-noise ratio. For single exponential echo decays, the optical coherence lifetime (T₂) was directly derived as exp(−2τ/T₂). Diluted complexes presented non-exponential decays that were described by double exponential decay curves from which we derived short and long T₂ components.

SHB spectra from isotopically (¹⁵¹Eu⁺) enriched and natural abundance complexes, both stoichiometric, were obtained by applying a single pulse, referred to as the burn pulse, of 1 ms length and excitation power of 15 mW. After a waiting time of 10 ms, the spectrum was read out with a 2 ms-long scanning pulse with 5 mW power and 200 MHz scanning range around the burning frequency (Supplementary Fig. 11c). The time before readout was set to 10 ms (that is, ≈T₁/₂) to enable spontaneous relaxation from the optical excited state, followed by progressive trapping into non-pumped nuclear spin levels. The SHB spectra were corrected for the frequency-dependent response of the acousto-optic modulator by dividing by a readout trace obtained with the burn pulse turned off. A series of high-power pulses scanning over 200 MHz was applied at the end of the sequence to reset the ground-state population back to equilibrium. The population lifetime of the ground-state spin levels (T₁/₂) was determined by monitoring the hole depth as a function of waiting time between the burn and readout pulses. The hole decay curve presents two distinct decay rates, estimated by fitting a double exponential decay to the experimental data (Fig. 3c).

AFC storage
The AFC preparation in the ¹⁵¹Eu⁺ complex was carried out as follows: absorption was increased in a spectral region of about 9 MHz by burning a corresponding broad hole (spectral pit) about 30 MHz away (Extended Data Fig. 4a) as determined by the nuclear spin splittings. In a second step, population was burned away at regularly spaced frequencies in the high-absorption region yielding an AFC spanning a 6 MHz range with teeth widths of 0.9 MHz and 1.75 MHz separation (Extended Data Fig. 4b). The experiment was performed 4.3 GHz away from the transition peak at 580.370 nm (Supplementary Fig. 10) to reduce absorption and facilitate optical pumping. The input pulse intensity for the AFC efficiency estimation was measured through the spectral pit and corrected for remaining residual absorption (Extended Data Fig. 4a). The full AFC preparation and storage sequence is displayed in Supplementary Fig. 15a.

Data availability
Datasets generated and/or analysed during the current study are available in the Zenodo repository (https://doi.org/10.5281/zenodo.5652030). Source data are provided with this paper.
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Author contributions P.G., M.R. and D.H. conceived and supervised the project. D.S. and S.K.K. were involved in the conceptual development of the project. S.K.K. and M.R. were responsible for the synthesis and characterization of the isotopologue complexes. B.H. performed powder X-ray diffraction studies and indexed the patterns. O.F. solved the X-ray structure of the complex. D.S. and P.G. performed the optical experiments and analysed the results. D.S. and P.G. wrote the manuscript with input from all authors.

Competing interests The authors declare no competing interests.

Additional information

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Extended Data Fig. 1 | Eu³⁺ complex preparation and X-ray crystal structure. Upper: Schematic representation of the preparation of the europium complex discussed in this study. Lower: X-ray crystal structure of the complex. As shown, the piperidin-1-ium cation is involved in hydrogen bonding interactions with two oxygen atoms of benzoylacetonate ligands.
Extended Data Fig. 2 | Photostability of the Eu³⁺ complex. $^{7}D_{0}$ photoluminescence (PL) intensity under continuous wave excitation measured at 8 K for the $^{151}$Eu³⁺ isotopically enriched complex. The constant PL signal confirms absence of photobleaching. See SI section 3 for more details.
Extended Data Fig. 3 | 2-pulse photon echo decay from the 5% Eu³⁺-95% Y³⁺ diluted complex. The experimental decay (black circles) was fitted with a double exponential model (red curve) with $T_{2,\text{fast}} = 18 \mu s$ and $T_{2,\text{slow}} = 68 \mu s$ decay time constants.
Extended Data Fig. 4 | Spectral tailoring prior to AFC storage. a. Spectral pit of 9 MHz dug in the absorption profile at 30 MHz to create a high absorption region at 0 MHz. b. Atomic frequency comb (AFC) with teeth of 0.9 MHz separated by 1.75 MHz (finesse $F = 1.9$). The FFT of the storage pulse is presented over the AFC (blue line) showing good spectral overlap. The input pulse intensity was estimated by sending it through the spectral pit in a, taking advantage of the pit’s almost full transparency. A correction was made to account for residual absorption.