Purcell Enhancement of Erbium Ions in TiO$_2$ on Silicon Nanocavities

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ABSTRACT: Isolated solid-state atomic defects with telecom optical transitions are ideal quantum photon emitters and spin qubits for applications in long-distance quantum communication networks. Prototypical telecom defects, such as erbium, suffer from poor photon emission rates, requiring photonic enhancement using resonant optical cavities. Moreover, many of the traditional hosts for erbium ions are not amenable to direct incorporation with existing integrated photonics platforms, limiting scalable fabrication of qubit-based devices. Here, we present a scalable approach toward CMOS-compatible telecom qubits by using erbium-doped titanium dioxide thin films grown atop silicon-on-insulator substrates. From this heterostructure, we have fabricated one-dimensional photonic crystal cavities demonstrating quality factors in excess of $5 \times 10^4$ and corresponding Purcell-enhanced optical emission rates of the erbium ensembles in excess of 200. This easily fabricated materials platform represents an important step toward realizing telecom quantum memories in a scalable qubit architecture compatible with mature silicon technologies.

KEYWORDS: Purcell enhancement, rare earth ions, erbium, quantum optics

are earth ion defects in solid-state hosts are key candidate qubits for applications in quantum computing and communication owing to their inherent spin–photon interface and long coherence times. These properties have enabled critical demonstrations of optical quantum memory protocols based on light-matter entanglement and entanglement distribution. Quantum technologies geared toward distribution of quantum information over long distances require qubits that interface with telecom photons to avoid huge propagation losses over optical fiber networks. Because the trivalent ion of erbium (Er$^{3+}$) has a $^4I_{15/2} \rightarrow ^4I_{13/2}$ transition in the telecom C-band, there has been interest in using erbium ions as optically addressable quantum memories using persistent spectral hole burning techniques and spin-based quantum memories in quantum communication, including at the level of single ions. However, significant engineering steps are needed to enhance the low photon emission rates from individual rare earth ions, which tend to have long radiative lifetimes.

One direct way to reduce the radiative lifetime is to use an optical cavity to enhance the emission through the Purcell effect. The majority of Purcell enhancement studies on rare earth ions have involved devices derived from rare earth-doped bulk crystals, such as heterogeneous integration with deposited amorphous Si resonators or bonded Si photonics, focused ion beam milling of bulk crystals, or incorporating relatively small erbium-doped samples into tunable distributed Bragg reflector-based fiber cavities. These approaches are all appealing because they leverage the generally good performance of well-studied host materials for Er$^{3+}$ (Y$_2$SiO$_5$, YVO$_4$, CaWO$_4$). Furthermore, these prior approaches have led to important cutting-edge demonstrations toward single rare-earth ion quantum memories, including single-shot spin state readout. However, such approaches may not allow for the on-chip scalability needed for wide deployment of quantum memories in large-scale quantum networks. For instance, estimates for fault-tolerant quantum repeater protocols show that hundreds or thousands of communication qubits are necessary to perform the requisite (operational and loss) error correction for quantum communication over long distances ($\geq 1000$ km). In addition, the implementation of a fully CMOS-compatible process flow can enable easy integration with filters, phase shifters, and beamsplitters for on-chip photon routing. In the vein of technological scalability with Si, there has also been considerable interest in low concentration implantation of erbium ions into Si nanostructures. While this approach
might have the greatest technological upside for directly unifying Si photonics with erbium-based quantum memories, early work has shown that the size mismatch between Er\(^3+\) and the crystalline lattice leads to myriad substitutional and interstitial sites with closely spaced spectral emission,\(^{25,26}\) and further experiments are needed to resolve the best defects for various applications.

In addition to ease of Si photonic integration—assessed by the ability to grow on Si and perform straightforward fabrication—there are a variety of additional considerations for a candidate material to serve as a high-quality host for Er\(^3+\). These include a minimal or controlled level of background nuclear spins, a wide bandgap, the number of substitutional sites available, and site symmetries.\(^{27-29}\) To that end, previous measurements have suggested that rutile TiO\(_2\) would be a good host for erbium because of narrow optical and spin line widths in implanted bulk crystals owing to the nonpolar symmetry of the substitutional erbium site and the low natural abundance of nuclear spins, respectively.\(^{30}\) Furthermore, TiO\(_2\) thin films can be grown on Si using a variety of deposition techniques,\(^{31,32}\) are CMOS-compatible, and are amenable to standard fluorine- and chlorine-based dry etch chemistries for top-down device fabrication.\(^{33}\) Recent work has shown Er-doped TiO\(_2\) thin films grown on Si substrates via molecular beam deposition.\(^{32}\) Some salient findings from that recent work are that the TiO\(_2\) thin films were polycrystalline and the dominant TiO\(_2\) phase (rutile or anatase) could be tuned by varying the substrate temperature during growth. Furthermore, the inhomogeneous line widths for the best buffered devices were as low as \(5 \, \text{GHz}\), which was substantially narrower than some of the epitaxial thin film control samples grown on better lattice-matched substrates (SrTiO\(_3\) or sapphire). Molecular beam deposition is particularly appealing for these types of heterostructures because of the inherent control of the Er\(^3+\) doping profile and doping density, in order to spectrally resolve single ions. Moreover, unintentional Er\(^3+\) doping from the TiO\(_2\) itself is anticipated to be lower than for the more common yttrium-containing oxides because of the relatively high concentrations of trace lanthanides in yttrium precursors.

In this Letter, we show early progress on top-down nanofabrication of photonic crystal cavities comprising thin layers of Er-doped TiO\(_2\) grown directly atop Si films. The thin films explored are similar to those discussed previously,\(^{32}\) but consist of an Er-doped TiO\(_2\) heterostructure grown on commercial silicon-on-insulator (SOI) substrates typically used by the Si photonics community: Si device layer thickness of \(220 \, \text{nm}\) atop a \(2 \, \mu\text{m}\) buried oxide (see Supporting Information (SI)). Within each TiO\(_2\) heterostructure, there are three TiO\(_2\) layers of equal thickness: an erbium-doped layer sandwiched between undoped top and bottom buffer layers,
and each layer is \( \sim 7.5 \) nm, giving a total film thickness of \( \sim 22 \) nm (Figure 1a). The estimated Er\(^{3+}\) density within the doped layer is 35 ppm (see SI). The TiO\(_2\) layers for this sample are grown at a substrate temperature of 520 °C. As shown in Figure 1b, transmission electron microscopy (TEM) cross-sectional imaging reveals that the TiO\(_2\) layer is polycrystalline, and there is a thin oxide layer \( \sim 2 \) nm at the Si/TiO\(_2\) interface, as was seen previously for lower temperature growth. However, unlike the previous demonstration, the polycrystalline film has both large and small grains visible, and the grains nearest the Si interface are bigger (13(3) nm) than those near the top of the TiO\(_2\) layer (\( \sim 1 \) nm). The cause of this gradient in grain size and effect on eventual Er properties is currently still under investigation. The roughness of the top surface of the TiO\(_2\) is \( \sim 1 \) nm according to the cross-sectional TEM.

Using 3D finite-difference time-domain (FDTD) simulations, we have designed one-dimensional photonic crystal cavities for our Si/TiO\(_2\) heterostructure. The photonic crystal devices consist of a waveguide with identical, elliptically shaped holes and a parabolic reduction of the lattice constant to generate a defect in the photonic bandgap (see SI). Figure 1c shows the computed fundamental (dielectric) mode with a predominant TE-like polarization in the plane of the TiO\(_2\) film.

Our strategy relies on the evanescent field in the Si waveguide coupling to ions in the doped layer of the nearby TiO\(_2\) film. The electric field intensity for a simulated electric dipole oriented normal to the waveguide, parallel to the plane of the TiO\(_2\), and 10 nm above the Si/TiO\(_2\) interface is approximately 37% of the maximum intensity in the center of the Si waveguide (see SI). The photonic crystal cavities (Figure 1d, bottom) are patterned via conventional electron-beam lithography and dry etching through both the TiO\(_2\) and Si device layers (see SI). After etching and hardmask removal, the waveguide is cleaved along alignment marks under a microscope, beyond the inverse-taper (Figure 1d, top). We employ an end-fire measurement configuration, whereby a lensed optical fiber is used to couple light in/out of the device. As a demonstration of the inherent scalability of this scheme, we reliably fabricate hundreds of photonic crystal devices on each chip, a representative array of which is shown in Figure 1e (see SI).

A schematic of our experimental setup used to measure these devices is shown in Figure 2a. We perform our measurements in a closed-cycle cryostat with a base temperature of \( T = 3.1 \) K. The sample is fixed to the coldfinger and the lensed fiber is mounted on a 3-axis nanopositioner to enable addressing of different devices. Using a tunable telecom C-band laser, we primarily probe devices with pulse lengths ranging from 1 to 1000 \( \mu \)s and collection times after the pulse that are sufficient to enable decay of the emission (see SI). The pulses are routed to the sample via a fiber circulator and polarization controller to match the polarization of the cavity mode. We then measure the fluorescence from the sample, back through the lensed fiber and circulator, and directed to either a superconducting nanowire single photon detector (SNSPD), a spectrometer with InGaAs camera, or a photodiode. The one-way coupling efficiency (see SI) is fairly poor at approximately 15%, but it is sufficient to probe these particular devices because of the relatively large number of ions in each device. We also have a fiber wavelength division multiplexer (WDM) that can be added into the path of port 2 of the circulator for off-resonant photoluminescence (PL) measurements using a 1480 nm diode laser. Off-resonant PL measurements on a bare waveguide device (no cavity) reveal a variety of peaks from 1520 to 1560 nm, as shown in Figure 2b. The largest peak near 1520.5 nm is attributed to substitutional Er in the rutile phase of TiO\(_2\), whereas the other dominant

Figure 2. Optical characterization of Er\(^{3+}\): TiO\(_2\) on Si waveguides. (a) Schematic of the experimental configuration. An Er\(^{3+}\): TiO\(_2\) waveguide device is situated in a cryostat \( T = 3.1 \) K. A tunable laser in combination with three fiber-coupled acousto-optic modulators (AOMs) enable the production of short pulses of light that are directed to the sample through a lensed fiber (LF) mounted on a nanopositioner (NP). The return light can be routed to either a photodiode (PD), IR spectrometer (Spec), or superconducting nanowire single-photon detector (SNSPD). A fiber polarization controller (FPC) is used to rotate the polarization to match that of the cavity. We can insert optional components (dashed boxes), such as an electro-optic phase modulator (phEOM) to generate sidebands for transient spectral hole burning and a fiber wavelength division multiplexer (WDM) for off-resonant excitation. Additional details are given in the Supporting Information. (b) PL spectrum of a bare waveguide (no cavity) device pumped with 1480 nm laser light and detected via spectrometer. The tallest peak (highlighted in yellow) is centered at 1520.56 nm and originates from Er\(^{3+}\) in the rutile phase of polyphase TiO\(_2\). Inset: A resonant laser scan showing a similar inhomogeneous line width of 0.4 nm (52 GHz) for the rutile peak. (c) Measurement of the spectral diffusion line width for the rutile transition at 1520.56 nm. The inverted Lorentzian fit (dashed magenta line) yields a spectral diffusion line width of 267(17) MHz.
peak near 1533 nm is attributed to substitutional Er in anatase, as confirmed previously with electron diffraction measurements. This particular sample is polyphase, which has been seen previously for TiO$_2$ thin films grown at intermediate temperatures. The intensity of the substitutional rutile transition—relative to the substitutional anatase peak—is much stronger in this sample compared to previous thin film results when grown near 520 °C. The origin of this TiO$_2$ phase discrepancy is still under investigation. Similarly, the origin of the minor peaks could be due to a variety of factors, such as different localized phases within the TiO$_2$, Er$^{3+}$ residing in different substitutional/interstitial sites, or crystal field-split transitions, but additional control samples and experiments are needed.

Resonant photoluminescence excitation (PLE) measurements on the bare waveguide device near 1520.56 nm are performed using a narrowband, tunable CW laser and modulated into pulses with emission from the Er$^{3+}$ detected during a collection window following each pulse. The lifetime decay curve and associated total PLE intensity at that pump wavelength are integrated after numerous pulses (see SI). Resonant PLE extracts a line width of 0.4 nm (Figure 2b inset), which is similar to the result from off-resonant pumping. The inhomogeneous line width represents the distribution in Er$^{3+}$ transition energies along the entire device that is over 40 μm long, and this inhomogeneous line width is similar to that measured in predominately rutile thin film samples using a confocal microscope geometry with a spot size of approximately 1 μm$^2$. Given that the inhomogeneous line width of the Er$^{3+}$ in rutile transition is relatively broad, it is important to investigate if the homogeneous line width of the emitters is sufficiently narrow to enable cavity-based lifetime enhancement (i.e., the “bad cavity” limit). To that end, we have performed transient spectral hole burning measurements on a bare waveguide device to find an upper bound on the homogeneous line width and spectral diffusion for the rutile transition (Figure 2c). Using the pulsed resonant laser PLE scheme described above, we have employed an additional phase modulator (ϕEOM, Figure 2a) to generate two sidebands each with a frequency detuning, Δ, from the carrier frequency. We can then measure the relative PLE intensity change as Δ is swept but the laser carrier wavelength is maintained at the rutile transition, 1520.56 nm, and the total pump power is near saturation for the ensemble. If Δ is larger than the spectral diffusion line width of the erbium ions, the ions are no longer pumped to saturation and the total fluorescence increases. If we plot the normalized intensity as a function of laser sideband-carrier detuning (Figure 2c), we can fit a Lorentzian line shape with a half-width-half-maximum (HWHM) of 267(17) MHz, which sets the upper bound on the spectral diffusion-limited homogeneous line width. While this homogeneous line width is substantially narrower than the inhomogeneous line width (Figure 2b), it is still quite large compared to the transform-limited line width of Er$^{3+}$ in rutile TiO$_2$ (sub kHz). It is possible that further optimization of the top capping layer in these devices can reduce the spectral diffusion of the Er emitters.

We have the ability to tune the cavity resonance in situ using a combination of controlled gas adsorption and desorption techniques while near base temperature. We use N$_2$ condensation via a gas nozzle directed at the sample to deposit a thin layer of ice on the cavity, which increases the refractive index of the mode, thereby resulting in a redshift of its resonance. We can also deterministically desorb the ice through localized heating, whereby we use relatively strong CW laser excitation tuned directly to the cavity resonance to induce two-photon absorption and heating in the Si, but only at the cavity region, which leads to a blueshift of the cavity resonance (see SI). Furthermore, this heating is sufficiently localized that cavities in neighboring waveguides—separated by only 6 μm—can be tuned independently of one another, which can be useful for future two-cavity interference experiments. We can directly measure the spectral position and line width of the cavity resonance by detecting the wavelength-dependent reflection of the narrowband laser light using a photodiode. If we perform this measurement close to the rutile transition, we typically see cavity quality factors (Q) near 5 × 10$^5$. A prototypical cavity reflection scan at T = 3.1 K is shown in Figure 3a. Based on measurements of control devices with and without TiO$_2$ films, the quality factors in general are currently limited by scattering due to the roughness of the top surface of the TiO$_2$ rather than sidewall roughness of the waveguide or cavity holes from the dry etching.

Our resonant pulsed laser measurements reveal the optical lifetime of all ions that couple to the device. This includes ions well-coupled to the cavity, ions poorly coupled to the cavity—whether because of position or polarization—and ions that couple only to the bare waveguide. Therefore, we use a stretched exponential function to capture the variety of decay times, where the time constant (τ) in the fit represents the fastest time decay within the ensemble. Using this approach (see SI), the Er ions coupled to a bare waveguide device give a natural lifetime of τ = 4660(20) μs (Figure 3b), which is close to the time constant of 5.1 ms for Er implanted/annealed bulk TiO$_2$ measured at low temperature. In comparison, the ions which couple most strongly to the cavity—when it is tuned onto the rutile transition at 1520.56 nm—show a greatly decreased decay time of τ = 23(1) μs. This represents a 200-fold decay rate enhancement.

We next measure the emission rate enhancement as a function of the detuning between the cavity resonance and the Er$^{3+}$ rutile transition. To do so, we perform a systematic redshift of the cavity resonance via gas condensation and measure the optical lifetime with the resonant laser fixed at the rutile transition. Figure 3c shows the increase in decay rate relative to the natural decay rate (Γ$/\Gamma_0$) as the cavity resonance is tuned across the transition. The cavity-enhanced emission has a Lorentzian line width of 5.06(0.17) GHz, which is modestly broader than the cavity line width measured via reflection of 3.71(0.17) GHz. However, this can be explained by reduction in Q when the cavity is resonant with the Er$^{3+}$ transition versus the reflection scan, which is measured at 90 pm (11.7 GHz) to the blue of the transition (see SI). The general agreement of the decay rate enhancement line shape with that of the cavity suggests that the radiative speedup is due to the cavity rather than nonradiative recombination induced by device fabrication. In addition, the lack of a plateau in Figure 3c confirms that the system is in the bad cavity limit, that is, Γ$/\Gamma_0 < \kappa$, where Γ$_0$ is the homogeneous line width of the emitters and $\kappa$ is the cavity line width, in agreement with the transient spectral hole measurement in bare waveguide devices. On the basis of our simulated Purcell factor for the ideally placed TE-oriented electric dipole at the center of the cavity (see SI)—after adjusting for the experimental Q (40 000) of this device at T = 3.1 K at 1526.56 nm—we estimate that the maximum Purcell factor in the ensemble should be 326. The
discrepancy between this estimate of the maximum Purcell factor and our experimentally determined value is likely due to a combination of the dipole orientations in the polyphase TiO$_2$ film and the specific location of erbium ions in the rutile phase grains probed by this experiment.

In this Letter, we have shown optical addressing of Er$^{3+}$ ions in rutile phase TiO$_2$ grown on commercial SOI wafers. We have fabricated top-down 1D photonic crystal cavities from the Si/TiO$_2$ heterostructure with Purcell enhancements up to 200. Systematic detuning of the cavity resonance from the Er$^{3+}$ transition in rutile phase TiO$_2$ reveals a good match of the Purcell enhancement to the Lorentzian cavity line shape, thereby indicating the erbium emission rates are limited by the cavity quality factor, not by the emitters’ homogeneous line widths. The next goal for this platform is to isolate and address individual Er ions as single spin quantum memories by cooling to lower temperatures and applying magnetic fields to address various spin levels. To that end, it is important to push the platform forward in three key areas.

First, it is necessary to reduce the homogeneous and inhomogeneous broadening in this system. The current upper bound of the homogeneous line width and spectral diffusion is estimated to be in the range of a few hundred MHz and could be attributed to a variety of factors, including trapped charges in the interfacial SiO$_x$ fabrication-induced damage, and the presence of mixed phases of TiO$_2$. Further improvement of the thin film growth conditions to isolate a single TiO$_2$ phase and to reduce the interfacial oxide, along with optimization of the buffer layer thicknesses could enable large reductions of the homogeneous and inhomogeneous line widths, as was seen already in predominantly anatase thin films. Second, it is important to reduce the number of Er$^{3+}$ coupled to the cavity to spectroscopically isolate and address single ions. By further reducing the metallic Er source flux and restricting doping to a single delta-doped TiO$_2$ layer, intentional doping of these films to the single ion level is straightforward via molecular beam deposition. Additional improvements to the homogeneous line width may also be achieved naturally with the reduction in the Er-doping density that would be needed to address single ions. Importantly, telecom quantum technologies based on this platform will be enabled by improving the photonic device performance. Most notably, the current fiber-waveguide coupling efficiency will need to be increased through improved mode-matching via cladding layers or a full undercut of the Si inverse tapered waveguides. Finally, a reduction in the TiO$_2$ surface roughness during growth can lead to improvements in cavity $Q$ for higher photon rates.

This demonstration represents an exciting first step toward a scalable on-chip quantum light source and memory device platform grown on an SOI wafer. This telecom-ready device architecture can be integrated with standard Si photonics and MEMS foundry process lines, enabling integration with other needed photonics elements such as on-chip filters, phase shifters, and beamsplitters. Furthermore, the Si platform is amenable to combination with other important cryogenic quantum technologies, such as on-chip SNSPDs, superconducting qubits, semiconductor quantum dot qubits, and microwave-to-optical transducers to enable powerful quantum computational and communication nodes. With further improvements to the materials and device fabrication, the scalable nature of this Er-doped TiO$_2$-on-SOI platform can meet the future demands for large numbers of communication qubits and quantum light sources needed for fault-tolerant long-distance quantum networking.

- **ASSOCIATED CONTENT**
- **Supporting Information**

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.nanolett.2c01561.

The Supporting Information contains additional simulation, fabrication, and experimental details, including: parameters for 3D FDTD simulations, the expected Purcell factor for dipoles in the TiO$_2$ layer, a full device
fabrication process flow, the cryogenic experimental configuration, rough estimates of the number of ions that couple to the cavity, and characterization of the cavity Q during gas condensation tuning.

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**REFERENCES**

1. Lvovskiy, A. I.; Sanders, B. C.; Tittel, W. Optical quantum memory. Nat. Photonics 2009, 3, 706–714.
2. Clausen, C.; Usmani, I.; Bussiérès, F.; Sangouard, N.; Afzelius, M.; De Riedmatten, H.; Gisin, N. Quantum storage of photonic entanglement in a crystal. Nature 2011, 469, 508–512.
3. Lago-Rivera, D.; Grandi, S.; Rakonjac, J. V.; Seri, A.; De Riedmatten, H. Telecom-heralded entanglement between multimode solid-state quantum memories. Natur 2021, 594, 37–40.
4. Saglamyurek, E.; Jin, J.; Verma, V. B.; Shaw, M. D.; Marsili, F.; Nam, S. W.; Oblak, D.; Tittel, W. Quantum storage of entangled telecom-wavelength photons in an erbium-doped optical fibre. Nat. Photonics 2015, 9, 83–87.
5. Miyazono, E.; Zhong, T.; Craiciu, I.; Kindem, J. M.; Faraon, A. Coupling of erbium dopants to yttrium orthosilicate photonic crystal cavities for on-chip optical quantum memories. Appl. Phys. Lett. 2016, 108, 011111.
6. Craiciu, I.; Lei, M.; Roehman, J.; Bartholomew, J. G.; Miyazono, E.; Zhong, T.; Sinclair, N.; Faraon, A. Nanophotonic quantum storage at telecommunications wavelength. Physical Review Applied 2019, 12, 024062.
7. Rancic, M.; Hedges, M. P.; Ahlefeldt, R. L.; Sellsar, M. J. Coherence time of over a second in a telecom-compatible quantum memory storage material. Nat. Phys. 2018, 14, 50–54.
8. Dibos, A. M.; Raha, M.; Phenicie, C. M.; Thompson, J. D. Atomic Source of Single Photons in the Telecom Band. Phys. Rev. Lett. 2018, 120, 243601.
9. Zhong, T.; Goldner, P. Emerging rare-earth doped material platforms for quantum nanophotonics. Nanophotonics 2019, 8, 2003–2015.
10. Raha, M.; Chen, S.; Phenicie, C. M.; Ourari, S.; Dibos, A. M.; Thompson, J. D. Optical quantum nondemolition measurement of a single rare earth ion qubit. Nat. Commun. 2020, 11, 1605.
(11) Martini, F. D.; Innocenti, G.; Jacobovitz, G. R.; Mataloni, P. Anomalous Spontaneous Emission Time in a Microscopic Optical Cavity. Phys. Rev. Lett. 1987, 59, 2955–2958.

(12) Chen, S.; Ourai, S.; Raha, M.; Phenicie, C. M.; Uysal, M. T.; Thompson, J. D. Hybrid microwave-optical scanning probe for addressing solid-state spins in nanophotonic cavities. Opt. Express 2021, 29, 4902.

(13) Zhong, T.; Kindem, J. M.; Bartholomew, J. G.; Rochman, J.; Craiciu, I.; Miyazono, E.; Bettinelli, M.; Cavalli, E.; Verma, V.; Nam, S. W.; Marsili, F.; Shaw, M. D.; Beyer, A. D.; Faraon, A. Nanophotonic rare-earth quantum memory with optically controlled retrieval. Science 2017, 357, 1392–1395.

(14) Craiciu, I.; Lei, M.; Rochman, J.; Bartholomew, J. G.; Faraon, A. Multifunctional on-chip storage at telecommunication wavelength for quantum networks. Optica 2021, 8, 114.

(15) Kindem, J. M.; Ruskuc, A.; Bartholomew, J. G.; Rochman, J.; Huan, Y. Q.; Faraon, A. Control and single-shot readout of an ion embedded in a nanophotonic cavity. Nature 2020, 580, 201–204.

(16) Merkel, B.; Ulanowski, A.; Reiserer, A. Coherent and Purcell-Enhanced Emission from Erbium Dopants in a Cryogenic High-Q Resonator. Physical Review X 2020, 10, 41025.

(17) Casabone, B.; Deshmukh, C.; Liu, S.; Serrano, D.; Ferrier, A.; Hümmer, T.; Goldner, P.; Hunger, D.; de Riedmatten, H. Dynamic control of Purcell enhanced emission of ion ensembles in nanoparticles. Nat. Commun. 2021, 12, 5370.

(18) Xie, T.; Rochman, J.; Bartholomew, J. G.; Ruskuc, A.; Kindem, J. M.; Craiciu, I.; Thiel, C. W.; Cone, R. L.; Faraon, A. Characterization of Er$^4+$:YVO$_4$ for microwave to optical transduction. Phys. Rev. B 2021, 104, 054111.

(19) Bertaina, S.; Gambarello, S.; Tkachuk, A.; Kurkin, I. N.; Malkin, B.; Step'anov, A.; Barbara, B. Rare-earth solid-state qubits. Nat. Nanotechnol. 2007, 2, 39–42.

(20) Le Dancte, M.; Rančić, M.; Lin, S.; Billaud, E.; Ranjan, V.; Flanagan, D.; Bertaina, S.; Chandelier, T.; Goldner, P.; Erb, A.; Liu, R. B.; Esteve, D.; Vion, D.; Flurin, E.; Bertet, P. Twenty-three-millisecond electron spin coherence of erbium ions in a natural-abundance crystal. Science Advances 2021, 7, No. eabj9786.

(21) Chen, S.; Raha, M.; Phenicie, C. M.; Ourai, S.; Thompson, J. D. Parallel single-shot measurement and coherent control of solid-state spins below the diffraction limit. Science 2020, 370, 592–595.

(22) Muralidharan, S.; Kim, J.; Lütkenhaus, N.; Lukin, M. D.; Jiang, L. Ultrafast and fault-tolerant quantum communication across long distances. Phys. Rev. Lett. 2014, 112, 250501.

(23) Moody, G.; et al. 2022 Roadmap on integrated quantum photonics. Journal of Physics: Photonics 2022, 4, 012501.

(24) Weiss, L.; Gritsch, A.; Merkel, B.; Reiserer, A. Erbium dopants in nanophotonic silicon waveguides. Optica 2021, 8, 40.

(25) Berkman, I. R.; Lysata, A.; de Boo, G. G.; Bartholomew, J. G.; Johnson, B. C.; McCallum, J. C.; Xu, B.-B.; Xie, S.; Ahlefeldt, R. L.; Sellars, M. J.; Yin, C.; Rogge, S. Sub-megahertz homogeneous linewidth for Er in Si via in situ single photon detection. arXiv 2021, DOI: 10.48550/arXiv.2108.07090.

(26) Hu, G.; Ahlefeldt, R. L.; de Boo, G. G.; Lysata, A.; Johnson, B. C.; McCallum, J. C.; Sellars, M. J.; Yin, C.; Rogge, S. Single site optical spectroscopy of coupled Er$^3+$ ion pairs in silicon. Quantum Science and Technology 2022, 7, 025019.

(27) Ferrenti, A. M.; de Leon, N. P.; Thompson, J. D.; Cava, R. J. Identifying candidate hosts for quantum defects via data mining. npj Computational Materials 2020, 6, 126.

(28) Stevenson, P.; Phenicie, C. M.; Gray, L.; Horvath, S. P.; Welinski, S.; Ferrenti, A. M.; Ferrier, A.; Goldner, P.; Das, S.; Ramesh, R.; Cava, R. J.; de Leon, N. P.; Thompson, J. D. Erbium-implanted materials for quantum communication applications. Phys. Rev. B 2022, 105, 224106.

(29) Kanai, S.; Heremans, F. J.; Seo, H.; Wolfovicz, G.; Anderson, C. P.; Sullivan, S. E.; Onizhuk, M.; Galli, G.; Awschalom, D. D.; Ohno, H. Generalized scaling of spin qubit coherence in over 12,000 host materials. Proc. Natl. Acad. Sci. U. S. A. 2022, 119, No. e2112808119.