



AFRL-AFOSR-VA-TR-2023-0245

New Materials Systems for Quantum Defects

**De Leon, Nathalie
TRUSTEES OF PRINCETON UNIVERSITY
1 NASSAU HALL
PRINCETON, NJ,
US**

**01/11/2023
Final Technical Report**

DISTRIBUTION A: Distribution approved for public release.

Air Force Research Laboratory
Air Force Office of Scientific Research
Arlington, Virginia 22203
Air Force Materiel Command

REPORT DOCUMENTATION PAGE

PLEASE DO NOT RETURN YOUR FORM TO THE ABOVE ORGANIZATION.

1. REPORT DATE 20230111	2. REPORT TYPE Final	3. DATES COVERED	
		START DATE 20180701	END DATE 20210630
4. TITLE AND SUBTITLE New Materials Systems for Quantum Defects			
5a. CONTRACT NUMBER	5b. GRANT NUMBER FA9550-18-1-0334	5c. PROGRAM ELEMENT NUMBER 61102F	
5d. PROJECT NUMBER	5e. TASK NUMBER	5f. WORK UNIT NUMBER	
6. AUTHOR(S) Nathalie De Leon			
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) TRUSTEES OF PRINCETON UNIVERSITY 1 NASSAU HALL PRINCETON, NJ US			8. PERFORMING ORGANIZATION REPORT NUMBER
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES) Air Force Office of Scientific Research 875 N. Randolph St. Room 3112 Arlington, VA 22203		10. SPONSOR/MONITOR'S ACRONYM(S) AFRL/AFOSR RTB1	11. SPONSOR/MONITOR'S REPORT NUMBER(S) AFRL-AFOSR-VA-TR-2023-0245
12. DISTRIBUTION/AVAILABILITY STATEMENT A Distribution Unlimited: PB Public Release			
13. SUPPLEMENTARY NOTES			
14. ABSTRACT Our work has focused on a systematic search aimed at the discovery of new solid state materials with defects that will act as bits for storing and processing quantum information. Although much progress has been made towards control of individual quantum defects in the solid state for applications in quantum information science and technology, prior work in this field has relied on the serendipitous, not the systematic, discovery of new materials and defects. In the first phase of this project, we leveraged our collaborative strengths in materials growth, defect engineering, spectroscopy, and confocal microscopy to create a screening pipeline for new quantum defects using complementary optical and spin spectroscopy techniques, combined with materials processing capabilities. This pipeline has allowed us to explore several new color centers in diamond, over a dozen new host materials for Er ³⁺ , and bulk and surface doping methods to stabilize particular charge states of quantum defects. This rate of exploration is orders of magnitude faster than the previous norm in the community, allowing us to uncover materials principles for new quantum defects, and filling out the "periodic table" of atomic defects relevant to quantum information science and technology. This work will enable emerging technologies in quantum information processing, nanoscale sensing, quantum metrology, and quantum communication.			
15. SUBJECT TERMS			
16. SECURITY CLASSIFICATION OF:		17. LIMITATION OF ABSTRACT	18. NUMBER OF PAGES
a. REPORT U	b. ABSTRACT U	c. THIS PAGE U	UU 9
19a. NAME OF RESPONSIBLE PERSON GRACE METCALFE			19b. PHONE NUMBER (Include area code) (703) 696-9740

Award Number/Project Number *Award number: FA9550-18-1-0334*

Report Type *Final*

Reporting Periods *1 July 2018 – 30 June 2021*

Distribution Statement *Distribution B – Not Approved For Public Release*

Program Officer Name *Dr. Grace Metcalfe, Dr. Ali Sayir*

Principal Investigator Name *Dr. Nathalie de Leon*

Project Title *New Materials Systems for Quantum Defects*

ABSTRACT

Our work has focused on a systematic search aimed at the discovery of new solid state materials with defects that will act as bits for storing and processing quantum information. Although much progress has been made towards control of individual quantum defects in the solid state for applications in quantum information science and technology, prior work in this field has relied on the serendipitous, not the systematic, discovery of new materials and defects. In the first phase of this project, we leveraged our collaborative strengths in materials growth, defect engineering, spectroscopy, and confocal microscopy to create a screening pipeline for new quantum defects using complementary optical and spin spectroscopy techniques, combined with materials processing capabilities. This pipeline has allowed us to explore several new color centers in diamond, over a dozen new host materials for Er^{3+} , and bulk and surface doping methods to stabilize particular charge states of quantum defects. This rate of exploration is orders of magnitude faster than the previous norm in the community, allowing us to uncover materials principles for new quantum defects, and filling out the “periodic table” of atomic defects relevant to quantum information science and technology. This work will enable emerging technologies in quantum information processing, nanoscale sensing, quantum metrology, and quantum communication.

Project Title: New Materials Systems for Quantum Defects
FA9550-18-1-0334

Principal Investigators: Nathalie de Leon, Jeff Thompson, Robert Cava, Stephen Lyon
Princeton University

Program Managers: Grace Metcalfe, Ali Sayir

Objective:

Our work has focused on a systematic search aimed at the discovery of new solid state materials with defects that will act as bits for storing and processing quantum information. Although much progress has been made towards control of individual quantum defects in the solid state for applications in quantum information science and technology, prior work in this field has relied on the serendipitous, not the systematic, discovery of new materials and defects. In the first phase of this project, we leveraged our collaborative strengths in materials growth, defect engineering, spectroscopy, and confocal microscopy to create a screening pipeline for new quantum defects using complementary optical and spin spectroscopy techniques, combined with materials processing capabilities. This pipeline has allowed us to explore several new color centers in diamond, over a dozen new host materials for Er^{3+} , and bulk and surface doping methods to stabilize particular charge states of quantum defects. This rate of exploration is orders of magnitude faster than the previous norm in the community, allowing us to uncover materials principles for new quantum defects, and filling out the “periodic table” of atomic defects relevant to quantum information science and technology. This work will enable emerging technologies in quantum information processing, nanoscale sensing, quantum metrology, and quantum communication.

Technical Approach:

Atomic and atom-like defects in solid-state hosts are one of the leading experimental platforms for quantum information processing, especially in the areas of quantum sensing and quantum networks.^{1,2} They offer excellent quantum coherence that enables long quantum bit (qubit) storage times, and efficient and stable optical transitions that can be used to manipulate and measure the qubit states. Several defects have been intensively studied over the last two decades, including the NV and SiV centers in diamond,^{1,3} damage centers in silicon,^{4,5} and rare earth ions.^{6,7} However, the space of possible defects is very large and mostly unexplored, and it is likely that much better candidates exist than those that are currently known.

The properties of a given defect are determined by the complex interaction of the defect with the host material through phonons, magnetic interactions with nuclei or paramagnetic impurities, charge transfer or charge noise, and Fermi level pinning by unwanted, co-occurring defects.⁸ This complexity makes the *ab initio* predictions of defects with desirable properties particularly difficult, motivating a broad experimental search. There are ongoing efforts in the community to look for new atom-like defects, especially in diamond and silicon carbide, primarily using single-atom fluorescence imaging techniques. This is a cumbersome approach for a broad search because it is slow, and requires that fluorescence from a defect is bright and stable enough to be observable by conventional means. Importantly, spin coherence is a critical parameter for any quantum system, and using fluorescence techniques to probe the spin

requires knowledge of the level structure and selection rules, which may themselves take years to work out.

Our plan, in contrast, is to pursue a novel approach in searching for new quantum defects by building an integrated pipeline for high-throughput characterization of hosts and defects, relying on rapid bulk characterization techniques, rather than on single-atom spectroscopy. The key elements of the pipeline are materials growth, implantation and annealing, and bulk spin and optical characterization. One major component of this bulk spectroscopy pipeline is a novel instrument that will allow spin and optical properties to be simultaneously characterized in bulk samples using a hybrid optical-electron spin resonance (ESR) approach. Finally, single-defect spin and optical characterization are applied to promising systems identified by the initial search.

Summary of work:

Over the past five years of this program, we have built a robust spectroscopic pipeline consisting of the following components:

1. Optical spectroscopy optimized for thin films of defects, including excitation-emission spectroscopy to disentangle different sites and subpopulations
2. High-sensitivity electron spin resonance and optical-ESR spectroscopy for associating optical lines and spin transitions
3. Materials processing (high vacuum and atmospheric annealing) and materials characterization for activating defects and mitigating ion implantation damage
4. Magneto-optic spectroscopy at high fields with a 6-1-1 vector magnet system (currently in development)

This pipeline has produced several hits for further study using single center spectroscopy: three new color centers in diamond, and three new host materials for Er. We have also learned many new materials processing and design rules for exploring more of the quantum defect parameter space. Here we describe two new spectroscopy tools that were developed as part of this research program.

First, we developed an optical-ESR spectrometer to definitively associate optical and spin transitions (Figure 1A). This is a new tool that we developed to characterize many sites and defects in complex samples, in contrast to the standard technique for associating spectroscopic lines seen in ESR and PL data—to look for correlated variations across samples or sample treatments that might change the density of a given defect. Such a process is extremely laborious and yields only indirect evidence. For example, in a case where 10 spin transitions and 10 optical transitions are observed in the same sample, there are around 3.6 million possible ways to pair them, so covariances of the 20 line intensities across a few samples can never yield enough information for an unambiguous association to be made. In our method, the ESR spectrometer is tuned to one of the spin transitions, while a laser illuminating the sample is scanned across one of the optical transitions identified in PL. If the optical transition belongs to the same defect as the spin transition, the amplitude of the ESR signal will change. Optical spin polarization and depolarization is a generic feature of atomic systems, so this technique is broadly applicable to quantum defects at low temperature.

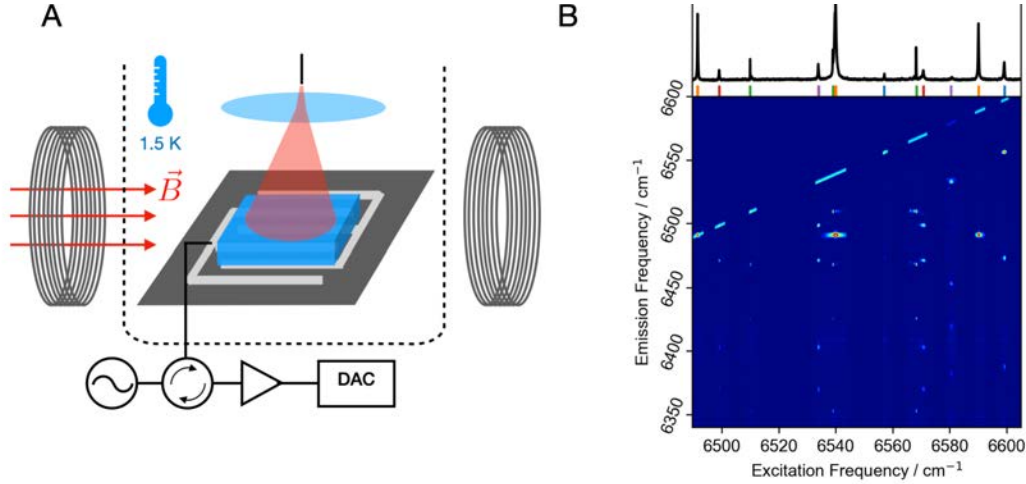


Figure 1: Quantum defect spectroscopy pipeline. (A) Hybrid optical-ESR spectrometer built for this project with the aim of rapidly associating optical and spin transitions. (B) Excitation-emission photoluminescence spectroscopy of $\text{Er}^{3+}:\text{MgO}$ showing several different incorporation sites, adapted from [17].

Second, we built an optical cryostat optimized for thin, implanted layers of defects to probe 2D excitation-emission spectroscopy (Figure 1B). By taking the PL spectra while scanning the excitation wavelength, we are able to readily group the optical transitions into particular sites, and then assign the spectral transitions for a given site. This rapid technique also gives us fast feedback on thermal annealing and materials processing, as it allows us to rapidly quantify conversion efficiencies into different sites.

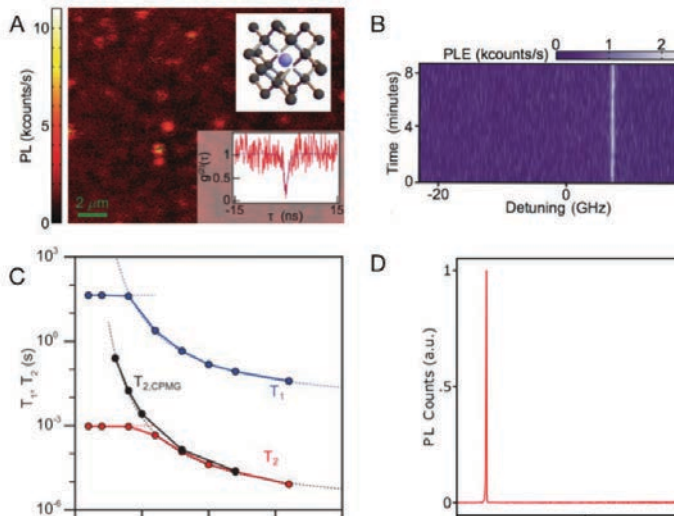


Figure 2: Coherence properties of SiV^0 (reproduced from [9]). (A) Single SiV^0 centers isolated by scanning confocal microscopy. (Insets) Atomistic structure of SiV^0 , Second order correlation spectra from a single center show antibunching. (B) Photoluminescence excitation (PLE) spectroscopy shows a near-transform-limited optical transition that is stable over time. (C) Spin T_1 , T_2 , Hahn echo, and T_2 , showing long relaxation and coherence times at low temperature. Photoluminescence (PL) spectroscopy shows that over 90% of optical emission is in the zero-phonon line.

A. Neutral silicon vacancy (SiV^0) centers in diamond

A major result from this systematic search has been the discovery of a new color center in diamond, the neutral charge state of the silicon vacancy (SiV^0) center, the first color center that has been demonstrated to have long spin coherence times ($T_1 \sim 1$ min, $T_2 \sim 1$ s) and near-transform-limited optical linewidths (Figure 2).⁹ Specifically, we achieved the stabilization and characterization of SiV^0 by deliberate engineering of the diamond host to tune the Fermi level below the $\text{SiV}^{0/-}$ transition point.

After establishing the optical and spin coherence properties of SiV^0 at low temperatures, we then turned to detailed characterization of the ground state spin dynamics at high temperatures.¹⁰ We then

established optically detected magnetic resonance (ODMR) in this new defect.¹¹ Specifically, using optical-ESR spectroscopy, we were able to identify an additional, previously unknown, excited state in SiV^0 at 952 nm, as well as several other higher lying transitions at 890, 850, and 835 nm that we have identified as Rydberg-like bound exciton transitions. These higher lying states allow for efficient bulk spin polarization, comparable to what is observed for resonant excitation at the zero-phonon line. This highly efficient spin polarization enabled the observation of ODMR for the first time.

Finally, we have recently shown that SiV^0 can be formed efficiently by two alternative methods. First, we showed that chemical control of the diamond surface could be used to achieve surface transfer doping, pulling the Fermi level below the $\text{SiV}^{0/-}$ transition point.¹² Specifically, we used low-damage chemical processing and annealing in a hydrogen environment to realize reversible and highly stable charge state tuning in undoped diamond. Second, we showed that SiV^0 centers can be efficiently stabilized by photoactivated itinerant carriers.¹³ Even in this nonequilibrium configuration, the resulting SiV^0 centers are stable enough to allow for resonant optical excitation and optically detected magnetic resonance. Both results pave the way towards broader technological deployment of SiV^0 because they obviate the need for precisely bulk-doped, high purity substrates.

B. Identification of the first telecom band emitter in diamond

We have also recently discovered the first telecom band emitter in diamond.¹⁴ The center has a zero-phonon line (ZPL) of ~ 1221 nm in the telecom O-band, which matches calculations of the $\text{SiV}_2:\text{H}^-$ center.¹⁵ We observe this center in multiple samples with 850 nm excitation at low temperature. This includes bulk-doped diamonds as well as Si-doped high-pressure high-temperature (HPHT) diamond nanoparticles (Figure 3). We have also studied the population dynamics of the defect using transient absorption spectroscopy, where we observe the same set of transitions and an excited state lifetime of around 230 ps. We assign the additional emission bands as phonon replicas, and we find that the intensity of the higher energy vibronic line (1170 nm) increases exponentially with temperature accordingly. The inferred vibrational energy is consistent with the motion of the Si-atom along the [111] axis of the center assigned as $\text{SiV}_2:\text{H}^-$.

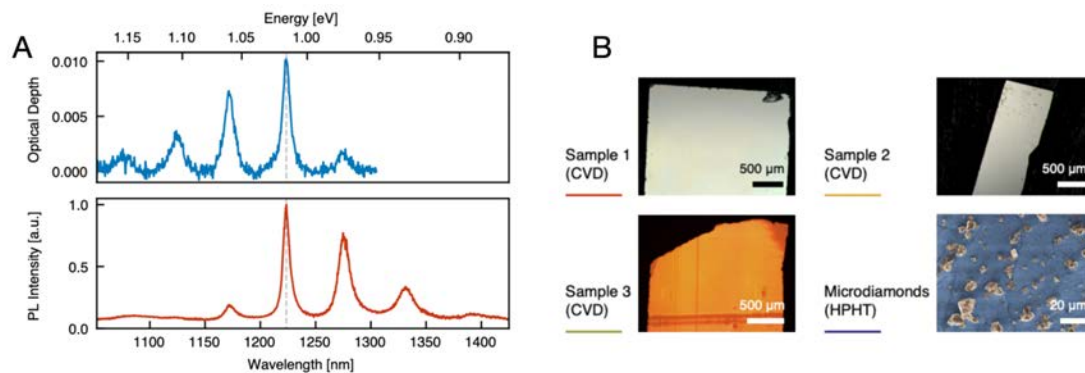


Figure 3: Telecom O-band emitter in diamond, adapted from [14]. (A) Absorption (top) and photoluminescence (bottom) spectra show mirror symmetry around a zero-phonon line at 1221 nm, allowing for spectroscopic identification. (B) This center has been observed in several samples with different synthesis conditions and form factors.

We are now working to isolate single centers and perform magneto-optic spectroscopy to study spin dynamics. Although this center is predicted to be a ground state singlet, we are interested in accessing the excited triplet state. Indeed, there is some indirect evidence in our transient absorption data for a long-lived triplet state, which could exhibit long spin lifetimes.

C. Preliminary characterization of oxygen vacancy (OV^0) centers in diamond

In the third year of our program, we expanded our optical spectroscopy capability to the visible range with a newly built broadband PL setup. The first system we studied was an oxygen related defect in diamond that we tentatively assign as OV^0 . In a bulk doped sample provided by Element Six, we observe spin transitions associated with a $S=1$ defect, close to the zero-field splitting of NV^- , which has been previously assigned as the WAR5 center [Cann]. Time-dependent measurements show that $T_1 = 25s$, $T_2 \sim 700$ us, and PL spectroscopy reveals an emission band at 543 nm. A separate, high purity sample was implanted with oxygen and annealed, and this sample also shows an emission band at 543 nm. Based on this correspondence, we believe that the WAR5 spin center is the neutral oxygen vacancy center. We have recently purchased a new laser to perform resonant PLE and optical-ESR measurements at 543 nm in order to definitively associate the optical and spin transitions. Our subsequent steps will be to explore sample fabrication and Fermi level stabilization to prepare high purity, well-controlled samples to study the limits of spin coherence.

D. Identification of several nuclear-spin free host materials for Er^{3+}

A major goal of our project is to develop new host material systems for Er^{3+} . Rare earth ions are an attractive class of atomic defects that feature narrow spin and optical transitions that are isolated from the host crystal, allowing incorporation into a wide range of materials. Er^{3+} in particular features a telecom band optical transition that is attractive for long distance quantum networks. However, the realization of long electronic spin coherence times is hampered by magnetic noise from abundant nuclear spins in the most widely studied host crystals, such as Y_2SiO_5 , $Y_3Al_5O_{12}$, $YLiF_4$ and Y_2O_3 . Our strategy is to identify commercially available, high purity, single crystal, bulk substrates and controllably introduce defects using ion implantation. Compared to bulk doping by synthetic methods, ion implantation enables much more rapid screening of defect-host combinations, which would otherwise require developing new synthetic procedures for each material system. We can constrain our search for good host materials for quantum defects to materials that satisfy the following: (1) the materials should be diamagnetic and composed of atoms with stable nuclear-spin-zero isotopes, (2) they should possess a suitable band gap to be optically transparent in the defect emission band, and (3) they should be of sufficient purity to allow for a low noise environment. Using these criteria, we applied a data mining and manual screening search to the Materials Project database and identified 541 possible hosts, 90 of which were unary or binary compounds.¹⁶

The first criterion confines us to even columns of the periodic table, and the second requires materials with optical band gaps greater than 0.8 eV. In our initial search we have focused on diamond, Si, SiC, SiO_2 , MgO, $PbWO_4$, $CaWO_4$, $SrTiO_3$, TiO_2 , MoO_3 , TeO_2 , ZnO, and ZnS. In addition to satisfying criteria (1) and (2), these materials were all commercially widely available as bulk single crystals with nominally high purity. The list includes covalent and ionic solids, as well as polar and nonpolar sites, allowing us to explore a wide range of material parameters in our search.

We performed 2D excitation-emission spectroscopy of Er^{3+} implanted into this list of host materials.¹⁷ Fast thermalization of the Er^{3+} excited states relative to the millisecond radiative lifetime results in identical emission spectra for each Er^{3+} site regardless of which transition was excited; thus when Er^{3+} can occupy multiple different sites the emission spectrum can be used to group excitation

peaks originating from Er^{3+} ions occupying the same site. With site-specific excitation and emission frequencies, we can extract the ground and excited state energies for each site. For each site we also measure the excited state lifetime, which we can compare to the expected magnetic dipole decay rate. This allows us to estimate the role of forced electric dipole moments in the decay rate, which are only allowed in sites without inversion symmetry.

By probing both the inhomogeneous linewidths and implantation yield for particular sites we are able to gain insight into both the static disorder in the crystal and the sensitivity of Er^{3+} to noise sources in the crystal. With this screen, we identified MgO , PbWO_4 , CaWO_4 , TiO_2 , and ZnO as particularly promising candidates, each showing narrow inhomogeneous linewidths for near-surface ions. In several material systems we have also demonstrated the importance of post-implantation thermal processing.

TiO₂

With our initial search, we demonstrated that Er^{3+} ions can be introduced via ion implantation into rutile TiO_2 , a host crystal that has not been studied extensively for rare earth ions and has a low natural abundance of nuclear spins.¹⁸ We observed efficient incorporation of the implanted Er^{3+} into the Ti^{4+} site (>50% yield) and measured narrow inhomogeneous spin and optical line widths (20 and 460 MHz, respectively) that are comparable to bulk-doped crystalline hosts for Er^{3+} . These narrow inhomogeneous linewidths arise from the nonpolar site symmetry, which guarantees a vanishing permanent dipole moment for the ion. Although this material system showed promising Er^{3+} optical properties, we were unable to address single Er^{3+} ions because silicon photonic crystals transferred to TiO_2 displayed low optical quality factors. Absorption in the telecom band in TiO_2 likely arises from other defects such as oxygen vacancies, and may be mitigated by other growth methods. We are currently working with materials synthesis collaborators (Charles Ahn at Yale, Tyrel McQueen at JHU) to study higher purity substrates.

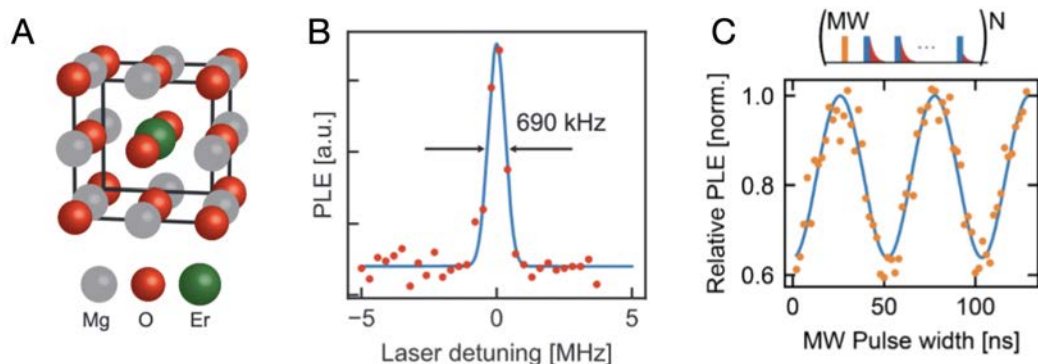


Figure 4: $\text{Er}^{3+}:\text{MgO}$ (A) Ball and stick model showing Er substitution in a Mg site. (B) PLE spectroscopy of a single Er^{3+} ion, showing sub-MHz linewidth. (C) Integration with silicon nanophotonics allows for spin readout.

MgO

We introduced Er^{3+} into MgO , a material with O_h symmetry, and we used optical spectroscopy to identify at least six different sites (Figure 4). Utilizing a cubic crystal-field model with an axial distortion yielded a good agreement between the observed level structure of one of the sites, which we attribute to the Mg^{2+} substitutional site with O_h symmetry previously identified in ESR. We used silicon photonic crystals transferred to the MgO surface to characterize single ions in the $\text{Er}^{3+}:\text{MgO}$ system, and obtained

single ion linewidths as narrow as 690 kHz. We demonstrated that these single ions can be coupled to photonic crystal cavities with a Purcell factor in excess of 1000 and exhibit little spectral diffusion on the timescale of hours.

E. Indistinguishable photons from $Er^{3+}:CaWO_4$

Most recently, we have demonstrated indistinguishable single photon emission from a single Er^{3+} ion coupled to a nanophotonic optical cavity. This is enabled by shallow ion implantation of Er^{3+} into $CaWO_4$, a host material satisfying the above criteria and for which long electron spin coherence has already been demonstrated in Er^{3+} ensembles.¹⁹ By coupling the ions to silicon nanophotonic circuits, we observe individual ions with optical linewidths of 150 kHz, and emission rate enhancement by a factor of $P = 850$ via the Purcell effect. In a Hong-Ou-Mandel experiment, we demonstrate that successive photons from a single ion have an

indistinguishability greater than 80%, by delaying one of the photons by $175 \mu s$ in a 36 km fiber spool. We also demonstrate spin initialization and single-shot readout with $F = 0.98$, and the preservation of electron spin coherence for more than $200 \mu s$, limited by paramagnetic impurities in the sample. This demonstration is a key step for the development of quantum repeaters based on single rare earth ions, and Er in particular.

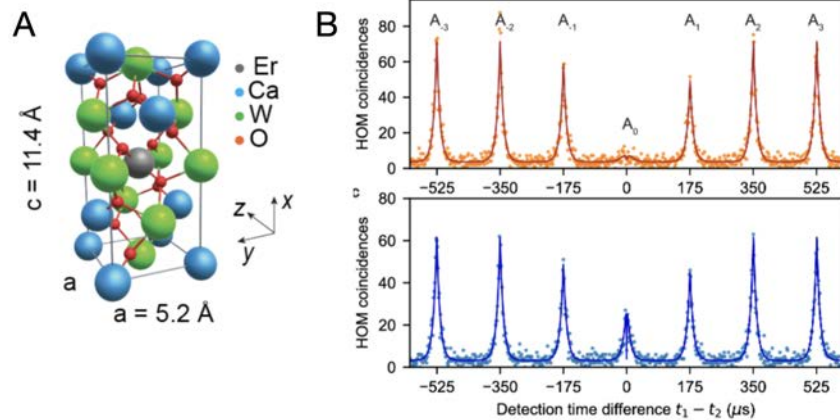


Figure 5: $Er^{3+}:CaWO_4$ (A) Ball and stick model showing Er^{3+} substitution into a Ca^{2+} site. (B) Indistinguishable photon emission from a single Er^{3+} ion shows excellent visibility. (Top) Indistinguishable case, (Bottom) distinguishable case with added phase noise on one path of the interferometer.

Future directions

In addition to the immediate next steps outlined above, there are several future directions for this research program. First, we plan to pursue higher purity synthesis and materials processing in the materials with promising Er^{3+} properties (MgO , TiO_2 , ZnO , $CaWO_4$) by thermal processing, zone refining, and homoepitaxy. Second, we would like to study the spin coherence in detail at lower temperatures, which would require building ESR capabilities in a dilution refrigerator. Third, we plan to pursue ultrafast spectroscopy methods to characterize detailed optical dynamics. Fourth, we will work iteratively with theorists to interpret our spectroscopic data and help refine their numerical methods.

Having developed our methodology with Er^{3+} , we also plan to broaden the scope to other defects, such as transition metals and molecular defects. One target of interest is to define the design parameters for high temperature operation in order to discover new quantum sensors.

Finally, we will explore the new device functionality allowed by some of these new host materials. For example, ZnO is a piezoelectric material, which may enable in situ tuning. MgO is known

to form high quality nanocrystals, which may enable heterogeneous integration into other material systems and devices.

References cited

1. Rodgers, L. V. H. *et al.* Materials challenges for quantum technologies based on color centers in diamond. *MRS Bull.* **46**, 623–633 (2021).
2. Chatterjee, A. *et al.* Semiconductor qubits in practice. *Nat. Rev. Phys.* **3**, 157–177 (2021).
3. Atatüre, M., Englund, D., Vamivakas, N., Lee, S.-Y. & Wrachtrup, J. Material platforms for spin-based photonic quantum technologies. *Nat. Rev. Mater.* **3**, 38–51 (2018).
4. Higginbottom, D. B. *et al.* Optical observation of single spins in silicon. *Nature* **607**, 266–270 (2022).
5. Komza, L. *et al.* Indistinguishable photons from an artificial atom in silicon photonics. Preprint at <https://doi.org/10.48550/arXiv.2211.09305> (2022).
6. Chen, S., Raha, M., Phenicie, C. M., Ourari, S. & Thompson, J. D. Parallel single-shot measurement and coherent control of solid-state spins below the diffraction limit. *Science* **370**, 592–595 (2020).
7. Gritsch, A., Weiss, L., Früh, J., Rinner, S. & Reiserer, A. Narrow Optical Transitions in Erbium-Implanted Silicon Waveguides. *Phys. Rev. X* **12**, 041009 (2022).
8. de Leon, N. P. *et al.* Materials challenges and opportunities for quantum computing hardware. *Science* **372**, eabb2823 (2021).
9. Rose, B. C. *et al.* Observation of an environmentally insensitive solid-state spin defect in diamond. *Science* **361**, 60–63 (2018).
10. Rose, B. C. *et al.* Strongly anisotropic spin relaxation in the neutral silicon vacancy center in diamond. *Phys. Rev. B* **98**, 235140 (2018).
11. Zhang, Z.-H. *et al.* Optically Detected Magnetic Resonance in Neutral Silicon Vacancy Centers in Diamond via Bound Exciton States. *Phys. Rev. Lett.* **125**, 237402 (2020).
12. Zhang, Z.-H. *et al.* Neutral silicon vacancy centers in undoped diamond via surface control. Preprint at <https://doi.org/10.48550/arXiv.2206.13698> (2022).
13. Zhang, Z.-H., Edmonds, A. M., Palmer, N., Markham, M. L. & de Leon, N. P. Neutral Silicon Vacancy Centers in Diamond via Photoactivated Itinerant Carriers. Preprint at <https://doi.org/10.48550/arXiv.2209.08710> (2022).
14. Mukherjee, S. *et al.* A telecom O-band emitter in diamond. Preprint at <https://doi.org/10.48550/arXiv.2211.05969> (2022).
15. Thiering, G. & Gali, A. Complexes of silicon, vacancy, and hydrogen in diamond: A density functional study. *Phys. Rev. B* **92**, 165203 (2015).
16. Ferrenti, A. M., de Leon, N. P., Thompson, J. D. & Cava, R. J. Identifying candidate hosts for quantum defects via data mining. *Npj Comput. Mater.* **6**, 1–6 (2020).
17. Stevenson, P. *et al.* Erbium-implanted materials for quantum communication applications. *Phys. Rev. B* **105**, 224106 (2022).
18. Phenicie, C. M. *et al.* Narrow Optical Line Widths in Erbium Implanted in TiO₂. *Nano Lett.* **19**, 8928–8933 (2019).
19. Le Dantec, M. *et al.* Twenty-three-millisecond electron spin coherence of erbium ions in a natural-abundance crystal. *Sci. Adv.* **7**, eabj9786 (2021).