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**Quantum Technologies with Optically Interfaced Solid-State Spins**

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**Spins of impurities in solids provide a unique architecture to realize quantum technologies. A quantum register of electron and nearby nuclear spins in the lattice encompasses high-fidelity state manipulation and readout, long-lived quantum memory, and long-distance transmission of quantum states by optical transitions that coherently connect spins and photons. These features, combined with solid-state device engineering, establish impurity spins as promising resources for quantum networks, information processing, and sensing. Focusing on optical methods for the access and connectivity of single spins, we review recent progress in impurity systems such as color centers in diamond and silicon carbide, rare-earth ions in solids, and donors in silicon. We project a possible path to chip-scale quantum technologies through sustained advances in nanofabrication, quantum control, and materials engineering.**

Driven by the quest for efficiency, modern technologies developed through persistent miniaturization. Devices such as transistors, magnetic memories, and lasers advanced by reducing the number of electrons used per gate, bit, or output photon. This progression's arrival at the quantum limit now inspires a new class of information processing hardware that starts with the quantum coherence of single charges, spins, or photons and grows by harnessing the inseparable connections among them. This reversal from scaling down to building up lies at the

34  heart of radical technologies that promise breakthroughs in computational power,
35  communications security, and sensor detection limit.

36  Solid-state spins are a promising platform for realizing these quantum advantages
37  because of their robustness to decoherence and compatibility with scalable device engineering[1].
38  In particular, this review focuses on optically addressed electron and nuclear spins at impurities
39  in crystals. In recent years, pioneering experiments have isolated single spins at these atomic-
40  scale impurities and demonstrated high-fidelity initialization, manipulation, and readout of their
41  quantum states[2]. These advances at the single-qubit level establish a critical foundation, but the
42  connectivity among multiple qubits is required to unlock their full potential. We highlight the
43  capacity of hybrid quantum registers formed by an electron spin coupled to multiple nuclear spins
44  in its proximity. Electron spins readily sense and interface to the outside environment, while
45  nuclear spins provide well-isolated quantum memories. These complementary functionalities,
46  accessed through the generation of entangled states, enable an array of applications, including
47  photonic memories[3], quantum repeaters[4], error-correction[5], and enhanced quantum sensing[6].

48  We concentrate on solid-state spins that utilize optical electronic transitions to fulfill several
49  of the DiVincenzo criteria for quantum information processing[7]. Optical pumping can directly
50  initialize the electron spin and its coupled nuclear spins, or alternatively, coherent manipulations
51  can transfer optically generated electron spin polarization to nuclear memories[8,9]. Additionally,
52  spin-dependent optical cycles correlate spin information to photon emission, enabling sensitive
53  readout of spin states. Such remarkable optical properties of defect systems have been combined
54  with techniques adapted from atomic physics and magnetic resonance to empower experiments
55  on single electron and nuclear spins at ambient conditions, surpassing limitations in the original
56  fields. Moreover, spin-selective optical transitions, accessed at cryogenic temperatures,
57  coherently map between the quantum states of local spins and propagating photons[10,11]. This
58  light-matter interface establishes each electron as a quantum gateway to distribute and process
59  entanglement between distant registers in a quantum network.

60  We aim to provide an introduction and broad update on optically-active impurity systems,
61  emphasizing the partnership between electron and nuclear spins. We first describe the framework
62  for manipulating hybrid quantum registers in the context of the prototype defect system, the
63  nitrogen-vacancy (NV) center in diamond[8]. We briefly review the optical and coherence properties
64  of the NV electron spin, which provides access to the entire register. This discussion identifies
65  the nuclear spin bath as the dominant source of decoherence but leads to the opportunity to

66 control selected nuclear memories via their distinct hyperfine interaction. We then overview
67 emerging impurity systems, including alternative color centers in diamond and silicon carbide,
68 rare-earth ions in solids, and optically-active donors in silicon. These platforms offer unique
69 advantages, such as in their optical properties or integrability with electronic or photonic devices
70 and stand to benefit from techniques developed for the NV center. In a latter part, we focus on
71 technological applications of registers of quantum memories, ranging from quantum
72 communication, computing, and sensing. We conclude our review by looking ahead to future
73 challenges and progress with impurity spins in solids.

74 We remark that spins in self-assembled[12,13] and gate-defined quantum dots[14] share many
75 parallel directions with impurity spins, including achievement of extended coherence times and
76 enhanced light-matter coupling to enable multi-qubit scaling and single photon nonlinearities. The
77 rapidly advancing state-of-the-art in this field is however beyond the scope of our discussion.
78 Likewise, we will overlook two-dimensional material systems, such as transition metal
79 dichalcogenides and hexagonal boron nitride, that have recently emerged as hosts for single
80 quantum emitters[15–17]. For these materials, explorations toward using the valley or spin degree of
81 freedom of excitons or defect states as qubits are still in their infancy but could open functionalities
82 for quantum photonics, optoelectronics, and sensing unattainable in bulk materials.

83 ## **The NV center in diamond**

84 Consisting of a substitutional nitrogen impurity adjacent to a missing carbon atom, the negatively
85 charged NV center in diamond traps six electrons at localized atomic-like states, protected from
86 charge scattering by diamond's wide bandgap (Fig. 1a). NV centers display room-temperature
87 quantum coherence, spin-photon entanglement, and functionality inside engineered
88 nanostructures, establishing their versatility for quantum information processing and nanoscale
89 sensing.

90 **The electron spin and its optical interface.** The NV electron spin can be off-resonantly excited
91 from its spin-triplet ground state (GS) to a spin-triplet, orbital-doublet excited state (ES) via
92 phonon-assisted optical absorption[8]. Due to a nonradiative, spin-flip decay channel that
93 preferentially couples to the $m_s = \pm 1$ sublevels of the excited state, repeated optical cycling
94 initializes the electron spin into the $m_s = 0$ level (~90% polarization)[18]. Concurrently, off-resonant
95 excitation of $|m_s = \pm 1\rangle$ results in ~30% lower photoluminescence (PL) than $|m_s = 0\rangle$, allowing
96 optical determination of the spin state at room temperature[19]. Higher fidelity initialization and
97 readout are obtained by cooling diamond below 10 K, where distinct spin-selective, zero-phonon

98    optical transitions are resolved[18] (Fig. 1b). Resonant optical pumping of a spin-mixed transition

99    (e.g. $|m_s = \pm1\rangle \rightarrow |A_1\rangle$) fully initializes the NV into $|m_s = 0\rangle$ (>99.7% polarization)[18]. Alternatively,

100    by resonant excitation of a cycling transition ($|m_s = 0\rangle \rightarrow |E_x\rangle$ or $|E_y\rangle$) and optimizing photon

101    collection efficiency, the electron spin state can be determined without averaging multiple

102    preparations (>97% fidelity averaged for $|m_s = 0\rangle$ and $|m_s = \pm1\rangle$ )[20]. Such single-shot

103    measurements can be non-demolition to allow initialization of electron and nuclear spins by

104    projective measurement[18]. Moreover, these spin-dependent optical transitions and their

105    polarization selection rules form the basis for spin-photon entanglement[10,11].

106    Aided by diamond's high Debye temperature and low spin-orbit coupling, NV centers possess

107    long spin-lattice relaxation times $T_1$ that reach ~5 ms at room temperature and exceed hours at

108    cryogenic temperatures (~25 mK)[21,22]. In high quality samples grown by chemical vapor

109    deposition, low concentrations of paramagnetic impurities leave the bath of $^{13}$C nuclear spins

110    (1.1% natural abundance) as the dominant magnetic noise[19]. This dephasing can be mitigated in

111    isotopically enriched materials (>99.99% $^{12}$C). For single spins in isotopically purified samples,

112    the inhomogeneous dephasing time $T_2^*$, reflecting temporal magnetic fluctuations, exceeds 100

113    µs at room temperature[23]. Dynamical decoupling further filters the noise spectrum and extends

114    spin coherence to the homogeneous dephasing time $T_2$ of several milliseconds at room

115    temperature[24] and nearly seconds at low temperature, limited by direct lattice contributions to spin

116    dephasing ($T_2 \approx 0.5\, T_1$)[25] (Fig. 1c). These remarkable coherence times underpin the technological

117    promise of NV centers, extending the range of its access to nearby nuclear spins and enhancing

118    its sensitivity to environmental influences.

119    **Strongly-coupled nuclear spins.** While the nuclear bath represents the main contribution to

120    electron spin decoherence, individual nuclear spins with isolated interactions offer a resource for

121    quantum memories and multi-qubit entanglement[9,26,27]. Strongly-coupled nuclear spins, such as

122    the intrinsic N forming the NV center and proximal $^{13}$C atoms, possess hyperfine couplings larger

123    than the electron spin resonance (ESR) linewidth, set by the dephasing rate $1/T_2^*$ (Fig. 1a,d). For

124    samples with natural isotope abundance, strongly-coupled nuclei typically occur within 1 nm from

125    the electron and have hyperfine couplings from 300 kHz to 130 MHz, where the latter value

126    corresponds to a $^{13}$C on a nearest neighbor lattice site[28]. For these nuclei, narrowband microwave

127    (MW) pulses at the distinct ESR transition frequencies (Fig. 1d) perform rotations of the electron

128    spin conditional on the nuclear spin state (e.g., controlled NOT gate, $C_nNOT_e$). Moreover, radio-

129    frequency (RF) pulses can directly drive nuclear spin transitions ($\Delta m_I = \pm1$) conditional on the

130    electron spin manifold (e.g. $C_eNOT_n$)[29]. Nuclear rotations can alternatively be implemented by

131  nuclear Larmor precession for orthogonal nuclear quantization axes in different electron

132  manifolds[9]. Using tailored pulse sequences of controlled two-qubit operations, the electron

133  polarization can be transferred to the nuclear spin, after which the electron spin is reset by optical

134  pumping[9,30].

135  Readout of the nuclear spin is achieved by mapping its population and coherences onto the

136  electron spin using a combination of selective RF and MW pulses[26]. In Ref. [[31]], single-shot

137  readout of the nuclear spin at room temperature was demonstrated by repeated application of

138  electron-nuclear correlation ($C_nNOT_e$) and optical readout of the electron (Fig. 1e). This repetition

139  leverages the robustness of the nuclear spin to optical excitation to accumulate sufficient photon

140  statistics. Projective readout can initialize multiple strongly coupled nuclei if the $C_nNOT_e$ logic is

141  conditional on each nuclear spin state of the multi-qubit register[18].

142  The coherence times of nuclear spins and their sensitivity to optical illumination determine their

143  usefulness as quantum memories. Nuclear coherence times $T_{2n}$ exceed several milliseconds at

144  room temperature and are limited by the electron spin relaxation time $T_1$ and interactions with

145  other nuclei[9]. Control over the NV charge state, either by strong optical illumination[32] or

146  electrostatic gating[33], can effectively decouple or eliminate the electron spin, extending nuclear

147  $T_{2n}$ beyond several seconds by the former technique. For nuclear depolarization ($T_{1n}$), transverse

148  components of the hyperfine interaction drive electron-nuclear flip-flops. Since hyperfine

149  interactions may be stronger in the excited state, optical excitation can exacerbate depolarization,

150  requiring high magnetic fields to minimize this process for strongly-coupled nuclear spins[31]. In

151  addition, these transverse terms become dominant near the excited-state anti-crossing ($B \approx$

152  $500\,G$), enabling optical excitation to drive dynamic nuclear spin polarization (DNP)[34,35]. The DNP

153  efficiency can approach unity, depending on the strength and direction of the hyperfine

154  interaction[36]. For quantum networks and error-corrected quantum computing, the coherences of

155  nuclear spins must also be conserved under all operations performed with the electron spin[37].

156  Failed remote entanglement attempts due to photon loss or errors in the electron spin readout

157  randomize the electron spin, introducing an uncertainty to the nuclear memory's precession

158  frequency and accumulated phase that cannot be refocused. Hence, these considerations have

159  motivated the exploration of more distant nuclear memories that are less sensitive to electronic

160  interference.

161  **Weakly-coupled nuclear spins.** Access to nuclear spins with hyperfine couplings weaker than

162  $1/T_2^*$ greatly increases the available number of register qubits and improves the longevity of each

163    memory, at the tradeoff of slower control speeds. This objective was achieved using dynamical

164    decoupling (DD) sequences on the electron, thereby both extending its coherence and isolating

165    its interaction with a particular nuclear spin[23,38,39]. The precession axis and frequency of a nuclear

166    spin is conditional on the electron spin state and on its distinct hyperfine tensor (Fig. 1f). DD

167    pulses that invert the electron spin thus modulate the nuclear precession, such that when the

168    unique dynamics of a targeted nuclear spin is in resonance with the periodicity of the DD pulses,

169    the precession due to its transverse hyperfine component accumulates[38,40] (Fig. 1g). Meanwhile,

170    destructive interference occurs for nuclear spins with dynamics out-of-sync with the DD sequence.

171    Through this method, universal control[40] and single-shot readout[41] of weakly-coupled nuclear

172    spins were demonstrated (Fig. 1h). These capabilities, together with remote electron

173    entanglement[11,42], establish an architecture for distributed quantum communication and

174    computing based on error-corrected, few-qubit nodes. Moreover, the direct applicability of DD

175    methods to sensing nuclear species external to diamond has impacted progress in nanoscale

176    magnetic resonance[43,44].

## Emerging Systems for Spin-Light Interfaces

178    Although possessing long spin coherence, NV centers are hindered by their broad emission

179    spectrum, where only ~4% of the total photons are coherently emitted into the zero-phonon line

180    (ZPL) at 6 K[45]. Since entanglement protocols require indistinguishable photons from remote NVs,

181    this low percentage and the high scattering losses in optical fiber at the ZPL wavelength (637 nm)

182    have limited entanglement rates to 40 Hz[20]. Achieving higher bandwidth in NV-based quantum

183    networks will thus require photonic cavities to boost ZPL emission[45–48] and, for long-distance

184    networks, quantum frequency conversion into telecom wavelengths[49], together with the

185    advantages of a quantum repeater architecture. Promising for quantum network applications at

186    cryogenic temperatures, recent experiments using microcavities have shown emission into the

187    ZPL with 46% probabability[50], and conversion of single NV photons into telecom wavelength has

188    been reported with 17% efficiency[51]. A parallel approach that may alleviate these requirements is

189    the advancement of alternative color centers with more favorable intrinsic properties.

190    **Silicon- and Germanium-Vacancy Centers in Diamond.** The silicon-vacancy center (SiV) in

191    diamond, consisting of an interstitial silicon atom midway between two adjacent carbon vacancies,

192    has garnered interest as a higher efficiency source of indistinguishable photons. The negative

193    charge state $SiV^-$ features an orbital doublet in both the ground and excited states, with

194    degeneracies broken by the spin-orbit interaction[52,53] (Fig. 2a). Four zero-phonon optical

195  transitions between these orbital levels comprise over 70% of the total fluorescence emission,

196  with ~50% into the single line C at 4 K (Fig. 2b). Moreover, due to the defect's inversion symmetry,

197  SiV⁻ optical transitions are robust in linear-order to electric field and strain variations, resulting in

198  highly stable and homogeneous emission frequencies with lifetime-limited linewidths[53].

199  The S=1/2 electronic spin of SiV⁻ can be coherently controlled by either microwave or optical

200  driving, where the latter utilizes a GS-ES lambda ($\Lambda$) system tuned by an off-axis magnetic field.

201  However, coherent population trapping[54,55] and Ramsey[56] experiments indicate a short spin

202  dephasing time $T_2^* \sim$ 100 ns at 4 K. Phonon-mediated transitions between the two ground-state

203  orbital branches, separated by 50 GHz ($\approx$ 2.5 K), represent the dominant contribution to spin

204  dephasing, arising from slightly different spin splitting in each orbital. Recently, by cooling SiV⁻ to

205  100 mK[57,58], spin coherence was extended to $T_2^* \approx$ 10 µs and $T_2 \approx$ 13 ms for an isotopically-

206  purified sample[58], clearing the way for coherent manipulation of coupled nuclear memories.

207  Additionally, SiV⁻'s efficient ZPL emission, predominantly polarized into a single transition dipole,

208  makes it attractive for coupling to nanophotonic structures[59]. Through precision placement of Si⁺

209  ions into a nanophotonic cavity, the entanglement signature between two emitters was observed[60]

210  (Fig. 2c). Possessing similar spin and optical properties as SiV⁻, the germanium-vacancy (GeV⁻)

211  in diamond may offer stronger atom-light coupling due to its higher quantum efficiency[61,62].

212  Furthermore, the neutral charge state of SiV[63,64] and the tin-vacancy center[65,66] have recently been

213  investigated to potentially combine a robust optical interface with long spin coherence at moderate

214  cryogenic temperatures, although optical spin readout for these systems has yet to be

215  demonstrated.

216  **Divacancy and Silicon-Vacancy in Silicon Carbide.** Silicon carbide (SiC) presents an

217  opportunity to integrate color centers into a technologically mature platform capable of wafer-

218  scale growth, fabrication, and doping[67]. Unlike diamond, SiC exists in over 250 polytypes[68]

219  (different stacking sequences of a silicon-carbon bilayer unit) to allow customizable material

220  properties and heterojunction devices, boosting the potential for interacting its hosted color center

221  spins with electrical or mechanical degrees of freedom[69,70]. Among color centers investigated in

222  SiC, the divacancy and silicon vacancy defects have fulfilled key requirements for quantum

223  applications, including optical electron and nuclear spin addressability[35,71], long spin coherence[72–

224  74], and single defect isolation[72,75].

225  The neutral divacancy ($V_C$-$V_{Si}$; missing C atom next to missing Si atom) possesses the same $C_{3v}$

226  symmetry and number of active electrons as the NV center in diamond, leading to analogous spin

227 and optical structures[67] (Fig. 2d). Its spin-triplet GS exhibits electron coherence times $T_2$ that can

228 exceed 1 ms in samples with natural isotope abundance[72]. This extended coherence despite

229 higher fractions of nuclear spins than diamond stems from the larger lattice spacing in SiC and

230 the suppression of heteronuclear flip-flops between $^{29}$Si and $^{13}$C at moderate magnetic fields ($T_2 >$

231 1 ms for $B > 15$ mT)[76,77]. Crucially, divacancies have ZPLs at near-infrared wavelengths (~1100

232 nm) where significantly lower attenuation in optical fiber (~1 dB/km vs ~8 db/km at 637 nm) will

233 facilitate entanglement generation over long distances. Recently, single divacancies, isolated in

234 the 4H and 3C polytypes, were shown to display well-resolved spin-dependent optical transitions

235 below 20 K, similar to the NV center (Fig. 2e)[78]. Consequently, high-fidelity resonant readout[78] of

236 the divacancy spin (Fig. 2f) and Stark tuning of its optical transition frequencies[79] were

237 demonstrated at cryogenic temperatures, establishing critical elements for the implementation of

238 entanglement protocols[11]. Harnessing the advantages of divacancies for long-range quantum

239 networks will also require improving their photon collection efficiency and natural ZPL emission

240 fraction (5-7%)[78]. Towards this, 3C-SiC, which can be epitaxially grown as thin films on silicon,

241 presents an advanced platform for nanophotonic engineering[80,81].

242 The negatively charged silicon vacancy ($V_{Si}$; single missing Si atom) diverges from the well-

243 studied NV-type level structure due to its odd number of active electrons that gives rise to a S =

244 3/2 ground-state and a complex set of optical transitions[82]. Notably, its spin properties and optical

245 readout at room temperature provide unique capabilities for thermometry[83] and vector

246 magnetometry[84]. Single $V_{Si}$ at both inequivalent lattice sites (V1 and V2 centers) in 4H-SiC have

247 been isolated, with both centers possessing coherence of order milliseconds under dynamical

248 decoupling[73,74] and the V1 center emitting 40% of its photons into the ZPL at 4 K[74] (Fig. 2d,g). A

249 crucial step will be demonstration of the predicted excited state fine structure[82] that enables

250 schemes for spin-photon entanglement.

251 **Rare-Earth Ions in Solids.** Due to shielding provided by their filled outer electronic shells, rare-

252 earth ions in solid-state crystals display *4f*-electronic optical and spin levels with narrow intrinsic

253 linewidths and high spectral stability, reminiscent of free atoms. This isolation from environmental

254 noise has been exploited in rare-earth ensembles for photonic quantum memories, where the

255 quantum state of single photons are stored and retrieved via the collective optical and spin-wave

256 excitations of the ensemble[3,85]. Moreover, rare earth impurities provide exceptionally long-lived

257 nuclear spins with relaxation and coherence times measured in units of days[86] and hours[87],

258 respectively. While these properties motivate rare-earth ensemble systems as leading platforms

259 for quantum repeaters and transducers[88,89], extending control to single rare-earth ions would
260 expand capabilities for programmable quantum circuits.

261 Recently, several groups have optically addressed single rare-earth ions, overcoming challenges
262 in signal-to-noise due to their weak optical emission and high background fluorescence in low
263 purity samples. These groups implemented techniques such as spectral isolation in micro-
264 crystallites[90,91], customized optics to minimize the confocal volume[92], and fluorescence up-
265 conversion to levels with shorter optical lifetimes[93]. Coherent control over single $Ce^{3+}$ electron
266 spins in yttrium aluminum garnet (YAG) was demonstrated[94]. Additionally, the hyperfine levels of
267 single $Pr^{3+}$ ions in yttrium orthosilicate (YSO)[90,91], $LaF_3$[92], and YAG[95] were spectroscopically
268 resolved, allowing nuclear spins to be initialized and read-out by resonant optical excitation and
269 manipulated by RF fields (Fig. 3a,b). Coherence of single nuclear spins in Pr:YAG was limited at
270 4 K by electron-phonon coupling[95] ($T_{1n} \sim 4\,\mu s$), but can exceed seconds for Pr:YSO based on
271 ensemble measurements[96]. Notably, an alternative detection route for single rare earths relies on
272 charge sensing of resonant photoionization, demonstrated for single $Er^{3+}$ ions in a silicon single-
273 electron transistor (SET)[97].

274 The robustness of rare-earth systems to surface perturbations motivates nanophotonic
275 engineering as a promising route to ameliorate their low photon count rate. Recently, an atomic
276 frequency comb quantum memory with optical control over the storage time was demonstrated
277 for an small ensemble of $Nd^{3+}$ ions in a nanophotonic cavity[98]. Moreover, several groups reported
278 addressing single rare-earth ions exploiting cavity enhancement of their spontaneous emission
279 rate (Purcell factors >100)[99,100]. Cavities can be fabricated directly into glassy rare-earth host
280 materials by focused ion beam milling[98,99]. Additionally, they can be fabricated externally into a
281 material such as silicon and then transferred onto the rare-earth host for evanescent coupling[100].
282 These platforms set the stage for advances in optical control and remote entanglement of rare-
283 earth electrons and nuclei, as well as quantum routing of single photons, in integrated photonic
284 devices.

285 **Optically Active Donors in Silicon.** The miniaturization of silicon electronics to the nanoscale
286 has naturally broached the prospect for quantum systems in this technologically ubiquitous
287 platform. In recent years, the electron and nuclear spin of single $^{31}P$ donors in silicon have been
288 read out by spin-dependent tunneling in an SET, demonstrating state-of-the-art coherence times
289 ($T_{2e} > 0.5$ s at 100 mK) and control fidelities (>99.99% for a single $^{31}P$ nuclear spin)[101]. However,
290 coupling between multiple donor qubits in silicon remains an unsolved challenge, motivating

291 various proposals that span nanometer[1] to macroscopic length scales[102,103]. Schemes requiring
292 strong coupling of individual donor spins to a common microwave resonator[102] are difficult to
293 realize due to their small magnetic dipole moment. Recently, strong coupling between microwave
294 cavity photons and a single electron spin in a silicon double quantum dot was achieved only after
295 hybridizing the spin with its electronic wavefunction in the presence of an inhomogeneous
296 magnetic field[104,105]. Alternatively, optical photon interconnects[103] could realize longer distance
297 couplings.

298 For group V shallow donors, optical transitions to excited orbital states occur in the far-infrared, a
299 technologically difficult regime. Instead, efforts have focused on transitions between the neutral
300 donor ($D^0$) and its donor-bound exciton ($D^0X$) with energies at 1.15 eV, near bandgap. In
301 isotopically-enriched $^{28}Si$ samples, these transitions are sufficiently narrow to resolve the
302 hyperfine structure of $^{31}P$ donors, allowing optical pumping through an Auger de-excitation to
303 hyperpolarize the electron and nuclear spin[106,107]. This Auger decay occurs with near-unit
304 probability in indirect bandgap Si and enables electrical readout of long-lasting nuclear spin
305 coherence that exceeds 39 minutes at room temperature for ensembles of ionized donors[106,107].
306 However, the nonradiative nature of this process precludes optical detection or coupling of single
307 donors.

308 Recently, singly-ionized, chalcogen double donors (e.g., $S^+$, $Se^+$) have begun to be investigated
309 for their stronger binding energies (~600 mV/2.9 µm) that permit donor optical transitions in the
310 mid-infrared (Fig. 3c)[103]. Optical measurements on $^{77}Se^+$ ensembles at 1.2 K demonstrate
311 competitive electron spin coherence times ($T_{2,Hahn} = 2$ s for a qubit based on a clock transition)
312 and hyperfine-resolved optical transitions with narrow homogeneous linewidths (<29 MHz).
313 Isolation of single chalcogen donors in the future could enable the use of silicon-on-insulator
314 photonics for cavity-enhanced readout and coupling[103].

315 **Applications of Quantum Registers and Memories**

316 In this section, we highlight recent advances that leverage the collective functionality of electron
317 and nuclear spins in solid-state defect systems for quantum applications.

318 **Quantum Networks.** Quantum networks that utilize shared entanglement between spatially
319 separated nodes represent the essential infrastructure for quantum key distribution and quantum
320 cloud computing, possessing security independent of the trustworthiness of the communicating
321 devices[108] or computing server[109]. Although photons are ideal long-distance carriers of quantum

322    states, their conversion to long-lived stationary qubits is necessary for flexible timing of network

323    tasks and efficient propagation of entanglement over long distances[4]. These requirements are

324    fulfilled by the NV center in diamond, which combines a spin-light interface with control over local

325    nuclear memories. In 2013, a breakthrough experiment established entanglement between two

326    NV spins separated by three meters[11]. Using the Barrett-Kok (BK) protocol, each NV center was

327    prepared to allow probabilistic emission of a photon correlated with its spin state. If the NV centers

328    emit indistinguishable photons (requiring the ZPL), overlapping their emitted modes on a beam-

329    splitter erases which-path information and causes detection of a single photon to project the NV

330    centers into an entangled state. Due to possible photon loss, detection of a second single photon

331    in an ensuing trial where both NV spins are flipped is required to confirm the entangled state,

332    leading to a low success probability $P_E \approx 10^{-7}$, proportional to the square of the photon collection

333    efficiency. Nevertheless, the high fidelity of the heralded entangled state (~.92), as well as its fast

334    tomography by single-shot readout, enabled the first loophole-free Bell test, using two NV centers

335    separated by 1.3 km[110] (Fig. 4a,b).

336    Incorporating nuclear memories with remote electron entanglement empowers key network

337    primitives, such as teleportation, entanglement purification, and photonic quantum memory. The

338    state of the nitrogen nuclear spin associated with one NV center of an entangled pair was

339    unconditionally teleported (i.e., each attempt is successful) onto the electron spin of the other NV

340    center[111]. Recently, entanglement purification, an essential capability of quantum repeaters that

341    "distills" a single high-fidelity entangled state from multiple lower fidelity copies, was achieved[112]

342    (Fig. 4c,d). Here, a low-quality entangled state is first generated between two NV spins by

343    detecting only a single output photon at the beam-splitter, forgoing the second trial of the BK

344    protocol[11,110]. The entangled electron states are then swapped onto $^{13}$C nuclear spins at both

345    nodes, freeing the communication qubits for additional entanglement generation. Critically, the

346    weakly-coupled nuclear memories used are robust to several hundred optical cycles of the

347    electron[37], allowing the second entanglement generation to be attempted until success. The

348    memory qubits are finally projected onto a pure entangled state depending on the outcome of

349    local operations on the two entangled pairs[112]. Since the purification protocol does not require

350    simultaneous detection of single photons in two consecutive entanglement attempts, it achieves

351    faster entanglement rates that scale linearly in photon detection probability.

352    While the above applications leverage the emission of spin-entangled photons, the NV's optical

353    interface additionally permits quantum storage of incident photons and absorption-based remote

354    entanglement[113]. Raman quantum memories for photons similar to those for rare-earth and atomic

355 systems were proposed for ensembles of NV centers[114,115], but have been difficult to realize due

356 to the crowded NV excited state structure and larger optical inhomogeneous broadening.

357 Alternatively, using a single NV center, photon polarization states were coherently transferred

358 onto the intrinsic nitrogen nuclear spin[113] (Fig. 4e,f). This recent demonstration prepares the

359 electron and nitrogen nuclear spins in an entangled Bell state, and then utilizes the entangled

360 absorption of a photon in a degenerate optical $\Lambda$ system[116] to teleport the photon polarization onto

361 the nuclear spin. This approach paves the way for entanglement distribution between network

362 nodes through the absorption of entangled photon pairs at the NV resonant energy.

363 **Quantum Computing.** Overcoming the challenge of scaling a quantum computer to the large

364 number of qubits required to outperform classical algorithms hinges on correcting the inevitable

365 errors that arise due to the delicate, analogue nature of quantum states. While a potential

366 resolution lies in a monolithic architecture employing a large qubit array stabilized by topological

367 codes, a distributed or modular architecture that utilizes photonically linked nodes, each with only

368 a small number of qubits, could provide efficiencies due to its reconfigurable connections and

369 non-local quantum gates[117]. This distributed architecture is particularly applicable to defect-based

370 quantum registers of electron and nuclear spins, which satisfy the requirements for inter-node

371 photonic entanglement, as well as intra-node universal control and non-demolition readout.

372 Encouragingly, a distributed error correction approach using only four qubits per node was shown

373 to possess modest threshold error rates for the entangling links (~10%) and local operations

374 (~1%) that would allow a large-scale implementation to be fault-tolerant[118].

375 Experimental efforts have so far focused on error correction within individual nodes. In 2014, two

376 groups[30,40] demonstrated a majority-vote error correction protocol, encoding a quantum bit in a

377 logical qubit of three spins (Fig. 4g). These experiments leveraged advanced electron-nuclear

378 initialization sequences and quantum gates to correct a single bit-flip error automatically during

379 the decoding of the three-qubit register, avoiding direct measurement of error syndromes. More

380 recently, by utilizing non-demolition, single-shot readout at cryogenic temperatures, a stabilizer-

381 based approach[119] involving active error detection and real-time feedback was implemented for a

382 logical qubit of three weakly-coupled $^{13}C$ nuclei. This latter experiment demonstrated a

383 continuously phase-corrected logical qubit with coherence exceeding its best component qubit.

384 Future prospects include extending error correction to larger qubit registers and applying more

385 sophisticated codes to repair arbitrary single-qubit errors, or combining it with remote electron

386 entanglement for networked information processing. In addition, the potential to coherently

387 interface shallow NV centers with chemically-assembled nuclear spin arrays on the surface of

388     diamond could enable large-scale quantum simulations[120]. Towards simulations of quantum

389     chemistry and condensed matter phases, proof-of-principle demonstrations with NV quantum

390     registers internal to diamond have deduced the energy structure of a HeH$^+$ cation[121] and a

391     topological wire coupled to a superconductor[122].

392     **Quantum Sensing.** The atomic-scale dimensions, fast dynamics, and acute sensitivity of

393     quantum systems make them exceptional probes of their environment, combining high spatial

394     resolution, bandwidth, and precision. The challenge of quantum sensing is to isolate maximum

395     information about a target variable from its effect on the dynamics of the quantum sensor, which

396     simultaneously experiences decoherence and a multitude of competing influences[6]. The room-

397     temperature coherence of NV centers and its diverse deployment inside wide-field arrays[123],

398     nanoparticles[124], and scanning probes[125–127] have precipitated their development as a

399     transformative technology for sensing magnetic fields and other external perturbations (Fig. 5a-

400     d). Wide-ranging efforts have leveraged the NV sensor for detection and characterization of

401     intracellular and biological processes[123,124,128], nanoscale nuclear magnetic resonance

402     (NMR)[43,44,129,130], magnetism in condensed matter systems[125–127,131], and device performance of

403     nanotechnologies[132,133].

404     In particular, NV detection of NMR spectroscopy significantly enhances the sensitivity and spatial

405     resolution of this powerful technique, promising chemical structure identification with single

406     molecule sensitivity and subcellular resolution (Fig. 5e,f). However, improving the frequency

407     resolution of NV detection to distinguish the few-hertz changes in NMR frequencies due to

408     chemical shifts and spin-spin couplings represents an ongoing pursuit. Pioneering works applied

409     DD sequences to single shallow NV centers to sense the magnetic field fluctuations from a

410     statistical polarization of ~$10^2$ external protons[43,44]. These experiments demonstrated ~10 kHz

411     NMR linewidths, limited by $T_2$ of the NV sensor and diffusion of the target nuclei through the

412     nanoscale sensing volume. Correlation spectroscopy extends the phase acquisition time to the

413     electron $T_1$ (~ms), improving spectral resolution to several hundred hertz[134,135]. Here, the phase

414     accumulated during an initial DD segment is stored in the polarization of the electron spin and

415     then correlated to the phase from a second DD segment after free evolution of the target nuclei.

416     Recently, the intrinsic nitrogen nuclear spin was leveraged as a longer-term memory for the initial

417     phase measured in correlation spectroscopy, extending the interrogation time to $T_{1n}$ (>minutes

418     for tesla-scale magnetic fields)[136–138]. This approach naturally integrates repetitive, non-demolition

419     nuclear state readout[31,130] and also frees the sensor electron spin so that its backaction on the

420　target nuclei, which shortens their correlation time, can be decoupled[137]. Although sample

421　diffusion still limited the sensing duration (~5 ms), the nuclear memory approach resolved part-

422　per-million level chemical shifts in liquid state samples[139] (Fig. 5f). Alternatively, a recent NMR

423　scheme[140] utilized an NV ensemble and a synchronous readout technique[141,142] that enables

424　frequency resolution independent of the sensor coherence time. Here, the larger sensing volume

425　mitigated the effects of diffusion and enabled access to the thermal, rather than statistical, nuclear

426　polarization, culminating in ~3 Hz spectral resolution to resolve chemical shifts and nuclear spin-

427　spin couplings[140].

428　**Challenges and Outlook**

429　Even with the enormous progress in impurity systems, the potential for improvement across the

430　spectrum of applications is limitless. Realizing kilohertz entanglement rates across metropolitan

431　distances, distributed logical qubits in a fault-tolerant network, or real-time intracellular NMR

432　spectroscopy, among other far-reaching goals, will require concerted advances in photonic and

433　device engineering, quantum control, and materials science. Moreover, while it may be enticing

434　to regard solid-state spins as a self-sufficient platform, exploration of their connection to diverse

435　quantum systems may lead to hybrid devices with optimized components. Recent advances in

436　interacting NV spins with microwave[143], acoustic[144,145], or magnonic excitations[146,147] open new

437　routes toward on-chip state transfer and entanglement between impurity spins or hybrid systems.

438　While the solid-state environment poses challenges to impurity spins via phonon broadening and

439　total internal reflection of their emission, it also provides them their greatest asset, amenability to

440　fabrication and device integration. Coupling of optical emitters to photonic cavities with high

441　quality factors and small mode volumes concentrates their emission into the ZPL and into a single

442　cavity mode for efficient collection. A recent Fabry-Pérot microcavity design[50] directs 46% of the

443　NV center's emission into the ZPL, offering *in-situ* resonance tuning and reduced processing-

444　induced broadening of NV linewidths as advantages over previous monolithic diamond

445　approaches[46] (Fig. 6a). In parallel, novel defect platforms with robust ZPLs[60] or advanced

446　heteroepitaxy and selective etching techniques[78] will accelerate photonic cavity development (Fig.

447　6b). These advances will enable significantly higher rates of indistinguishable photons for long-

448　distance entanglement, as well as ultimately near-deterministic light-matter interactions in the

449　strong-coupling regime of cavity quantum electrodynamics. Towards continental-scale, fiber-

450　based quantum networks, defect systems with near-telecom wavelength emission, such as

451 divacancies in SiC or various rare earths, may offer additional avenues to create efficient
452 entanglement between solid-state nodes.

453 At a more local level, a promising direction lies in integrating impurity spins with on-chip photonics,
454 thereby creating field-deployable quantum sensors and compact information processing devices.
455 An integrated package using microfabricated photonic elements, including waveguides, couplers,
456 metalenses, and superconducting nanowire detectors, could crucially reduce optical losses and
457 eliminate bulky free-space optics[48,148,149] (Fig. 6c). In these integrated photonic circuits, impurity
458 spins would provide powerful functionalities as single photon sources, photonic memories, and
459 nonlinear single-photon switches, establishing a scalable architecture for quantum processors
460 based on either photonic or modular matter-based qubits. Moreover, engineering of lab-on-a-chip
461 devices[150] and deployable probes, such as nanodiamonds attached directly to optical fibers[151],
462 will proliferate the reach of quantum sensing. For example, diamond devices with surface-
463 structured microfluidic grooves realize both enhanced sensor-analyte contact for NMR
464 applications and increased photon collection efficiency by optical waveguiding[152].

465 Advances in quantum control and materials science will naturally be fundamental to continued
466 progress. Encoding quantum memories in decoherence-free subspaces of two nuclear spins
467 would allow the use of more strongly-coupled nuclear spins while maintaining robustness to
468 optical excitation[37]. This would increase speeds for interfacing quantum memories and enable
469 high-throughput, multiplexed versions of entanglement protocols, where entanglement is
470 continuously attempted using multiple memories before successes are heralded[153]. Time-
471 dependent Hamiltonian engineering, such as optimal control[154] and shortcuts to adiabacity[155],
472 could be extended for fast and robust gates despite densely spaced electron-nuclear energy
473 levels. Furthermore, the convergence of quantum error correction and quantum sensing could
474 improve sensitivity by extending qubit coherence regardless of the noise spectrum, in contrast to
475 dynamical decoupling. Pioneering demonstrations[156,157] leveraged a single ancilla nuclear spin
476 that is robust against noise to correct the NV electron sensor. Finally, improvements in materials
477 engineering could overcome current limits to many applications, as well as open new
478 opportunities. Important challenges include the creation of high-density, near-surface defect
479 centers with long coherence times and a single orientation[158–160], control over defect charge states
480 by Fermi level tuning, and customized surface functionalization for targeted sensing tasks. In
481 addition, the discovery of novel impurity systems with superlative optical, spin, and host material
482 properties may aid in bringing solid-state spins closer to widely impactful quantum technologies.

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**References**

1. Kane, B. E. A silicon-based nuclear spin quantum computer. *Nature* **393,** 133–137 (1998).

2. Hanson, R. & Awschalom, D. D. Coherent manipulation of single spins in semiconductors. *Nature* **453,** 1043–1049 (2008).

3. de Riedmatten, H. & Afzelius, M. in *Engineering the Atom-Photon Interaction* (eds. Predojević, A. & Mitchell, M. W.) 241–273 (Springer, 2015).

4. Munro, W. J., Azuma, K., Tamaki, K. & Nemoto, K. Inside Quantum Repeaters. *IEEE J. Sel. Top. Quantum Electron.* **21,** 78–90 (2015).

5. Terhal, B. M. Quantum error correction for quantum memories. *Rev. Mod. Phys.* **87,** 307–346 (2015).

6. Degen, C. L., Reinhard, F. & Cappellaro, P. Quantum sensing. *Rev. Mod. Phys.* **89,** 035002 (2017).

7. DiVincenzo, D. P. The Physical Implementation of Quantum Computation. *Fortschritte Phys.* **48,** 771–783 (2000).

8. Doherty, M. W. *et al.* The nitrogen-vacancy colour centre in diamond. *Phys. Rep.* **528,** 1–45 (2013).

9. Dutt, M. V. G. *et al.* Quantum Register Based on Individual Electronic and Nuclear Spin Qubits in Diamond. *Science* **316,** 1312–1316 (2007).

10. Togan, E. *et al.* Quantum entanglement between an optical photon and a solid-state spin qubit. *Nature* **466,** 730–736 (2010).

11. Bernien, H. *et al.* Heralded entanglement between solid-state qubits separated by three metres. *Nature* **497,** 86–90 (2013).

12. Warburton, R. J. Single spins in self-assembled quantum dots. *Nat. Mater.* **12,** 483–493 (2013).

13. Lodahl, P. Quantum-dot based photonic quantum networks. *Quantum Sci. Technol.* **3,** 013001 (2018).

14. Vandersypen, L. M. K. *et al.* Interfacing spin qubits in quantum dots and donors—hot, dense, and coherent. *npj Quantum Inf.* **3,** 34 (2017).

15. Aharonovich, I., Englund, D. & Toth, M. Solid-state single-photon emitters. *Nat. Photon.* **10,** 631–641 (2016).

16. Shiue, R. J. *et al.* Active 2D materials for on-chip nanophotonics and quantum optics. *Nanophotonics* **6,** 1329–1342 (2017).

17. 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).

18. Robledo, L. *et al.* High-fidelity projective read-out of a solid-state spin quantum register.

530     *Nature* **477,** 574–578 (2011).

531     19.     Childress, L. *et al.* Coherent Dynamics of Coupled Electron and Nuclear Spin Qubits in
532             Diamond. *Science* **314,** 281–285 (2006).

533     20.     Humphreys, P. C. *et al.* Deterministic delivery of remote entanglement on a quantum
534             network. *Nature* **558,** 268–273 (2018).

535     21.     Jarmola, A., Acosta, V. M., Jensen, K., Chemerisov, S. & Budker, D. Temperature- and
536             Magnetic-Field-Dependent Longitudinal Spin Relaxation in Nitrogen-Vacancy Ensembles
537             in Diamond. *Phys. Rev. Lett.* **108,** 197601 (2012).

538     22.     Astner, T. *et al.* Solid-state electron spin lifetime limited by phononic vacuum modes. *Nat.*
539             *Mater.* **17,** 313–317 (2018).

540     23.     Zhao, N. *et al.* Sensing single remote nuclear spins. *Nat. Nanotech.* **7,** 657–662 (2012).

541     24.     Balasubramanian, G. *et al.* Ultralong spin coherence time in isotopically engineered
542             diamond. *Nat. Mater.* **8,** 383–387 (2009).

543     25.     Bar-Gill, N., Pham, L. M., Jarmola, A., Budker, D. & Walsworth, R. L. Solid-state
544             electronic spin coherence time approaching one second. *Nat. Commun.* **4,** 1743 (2012).

545     26.     Neumann, P. *et al.* Multipartite entanglement among single spins in diamond. *Science*
546             **320,** 1326–1329 (2008).

547     27.     Fuchs, G. D., Burkard, G., Klimov, P. V. & Awschalom, D. D. A quantum memory intrinsic
548             to single nitrogen–vacancy centres in diamond. *Nat. Phys.* **7,** 789–793 (2011).

549     28.     Dréau, A., Maze, J. R., Lesik, M., Roch, J. F. & Jacques, V. High-resolution spectroscopy
550             of single NV defects coupled with nearby 13C nuclear spins in diamond. *Phys. Rev. B* **85,**
551             134107 (2012).

552     29.     Jelezko, F. *et al.* Observation of Coherent Oscillation of a Single Nuclear Spin and
553             Realization of a Two-Qubit Conditional Quantum Gate. *Phys. Rev. Lett.* **93,** 130501
554             (2004).

555     30.     Waldherr, G. *et al.* Quantum error correction in a solid-state hybrid spin register. *Nature*
556             **506,** 204–207 (2014).

557     31.     Neumann, P. *et al.* Single-Shot Readout of a Single Nuclear Spin. *Science* **329,** 542–544
558             (2010).

559     32.     Maurer, P. C. *et al.* Room-Temperature Quantum Bit Memory Exceeding One Second.
560             *Science* **336,** 1283–1286 (2012).

561     33.     Pfender, M. *et al.* Protecting a Diamond Quantum Memory by Charge State Control.
562             *Nano Lett.* **17,** 5931–5937 (2017).

563     34.     Jacques, V. *et al.* Dynamic Polarization of Single Nuclear Spins by Optical Pumping of
564             Nitrogen-Vacancy Color Centers in Diamond at Room Temperature. *Phys. Rev. Lett.* **102,**
565             057403 (2009).

566     35.     Falk, A. L. *et al.* Optical Polarization of Nuclear Spins in Silicon Carbide. *Phys. Rev. Lett.*

567 **114,** 247603 (2015).

568 36. Ivády, V. *et al.* Theoretical model of dynamic spin polarization of nuclei coupled to
569 paramagnetic point defects in diamond and silicon carbide. *Phys. Rev. B* **92,** 115206
570 (2015).

571 37. Reiserer, A. *et al.* Robust Quantum-Network Memory Using Decoherence-Protected
572 Subspaces of Nuclear Spins. *Phys. Rev. X* **6,** 021040 (2016).

573 38. Kolkowitz, S., Unterreithmeier, Q. P., Bennett, S. D. & Lukin, M. D. Sensing Distant
574 Nuclear Spins with a Single Electron Spin. *Phys. Rev. Lett.* **109,** 137601 (2012).

575 39. Taminiau, T. H. *et al.* Detection and control of individual nuclear spins using a weakly
576 coupled electron spin. *Phys. Rev. Lett.* **109,** 137602 (2012).

577 40. Taminiau, T. H., Cramer, J., van der Sar, T., Dobrovitski, V. V & Hanson, R. Universal
578 control and error correction in multi-qubit spin registers in diamond. *Nat. Nanotech.* **9,**
579 171–176 (2014).

580 41. Liu, G.-Q. *et al.* Single-Shot Readout of a Nuclear Spin Weakly Coupled to a Nitrogen-
581 Vacancy Center at Room Temperature. *Phys. Rev. Lett.* **118,** 150504 (2017).

582 42. Dolde, F. *et al.* Room-temperature entanglement between single defect spins in diamond.
583 *Nat. Phys.* **9,** 139–143 (2013).

584 43. Staudacher, T. *et al.* Nuclear magnetic resonance spectroscopy on a (5-nanometer)3
585 sample volume. *Science* **339,** 561–563 (2013).

586 44. Mamin, H. J. *et al.* Nanoscale Nuclear Magnetic Resonance with a Nitrogen-Vacancy
587 Spin Sensor. *Science* **339,** 557–560 (2013).

588 45. Barclay, P. E., Fu, K.-M. C., Santori, C., Faraon, A. & Beausoleil, R. G. Hybrid Nanocavity
589 Resonant Enhancement of Color Center Emission in Diamond. *Phys. Rev. X* **1,** 011007
590 (2011).

591 46. Faraon, A., Santori, C., Huang, Z., Acosta, V. M. & Beausoleil, R. G. Coupling of
592 Nitrogen-Vacancy Centers to Photonic Crystal Cavities in Monocrystalline Diamond.
593 *Phys. Rev. Lett.* **109,** 033604 (2012).

594 47. Aharonovich, I. & Neu, E. Diamond nanophotonics. *Adv. Opt. Mater.* **2,** 911–928 (2014).

595 48. Schröder, T. *et al.* Quantum nanophotonics in diamond. *J. Opt. Soc. Am. B* **33,** B65–B83
596 (2016).

597 49. Zaske, S. *et al.* Visible-to-Telecom Quantum Frequency Conversion of Light from a
598 Single Quantum Emitter. *Phys. Rev. Lett.* **109,** 147404 (2012).

599 50. Riedel, D. *et al.* Deterministic Enhancement of Coherent Photon Generation from a
600 Nitrogen-Vacancy Center in Ultrapure Diamond. *Phys. Rev. X* **7,** 031040 (2017).

601 51. Dréau, A., Tcheborateva, A., Mahdaoui, A. El, Bonato, C. & Hanson, R. Quantum
602 frequency conversion to telecom of single photons from a nitrogen-vacancy center in
603 diamond. Preprint at http://arxiv.org/abs/1801.03304 (2018).

604   52.   Hepp, C. *et al.* Electronic Structure of the Silicon Vacancy Color Center in Diamond.
605         *Phys. Rev. Lett.* **112,** 036405 (2014).

606   53.   Rogers, L. J. *et al.* Multiple intrinsically identical single-photon emitters in the solid state.
607         *Nat. Commun.* **5,** 4739 (2014).

608   54.   Pingault, B. *et al.* All-Optical Formation of Coherent Dark States of Silicon-Vacancy Spins
609         in Diamond. *Phys. Rev. Lett.* **113,** 263601 (2014).

610   55.   Rogers, L. J. *et al.* All-Optical Initialization, Readout, and Coherent Preparation of Single
611         Silicon-Vacancy Spins in Diamond. *Phys. Rev. Lett.* **113,** 263602 (2014).

612   56.   Pingault, B. *et al.* Coherent control of the silicon-vacancy spin in diamond. *Nat. Commun.*
613         **8,** 15579 (2017).

614   57.   Becker, J. N. *et al.* All-Optical Control of the Silicon-Vacancy Spin in Diamond at
615         Millikelvin Temperatures. *Phys. Rev. Lett.* **120,** 053603 (2018).

616   58.   Sukachev, D. D. *et al.* Silicon-Vacancy Spin Qubit in Diamond: A Quantum Memory
617         Exceeding 10 ms with Single-Shot State Readout. *Phys. Rev. Lett.* **119,** 223602 (2017).

618   59.   Rogers, L. J. *et al.* Electronic structure of the negatively charged silicon-vacancy center in
619         diamond. *Phys. Rev. B* **89,** 235101 (2014).

620   60.   Sipahigil, A. *et al.* An integrated diamond nanophotonics platform for quantum-optical
621         networks. *Science* **354,** 847–850 (2016).

622   61.   Bhaskar, M. K. *et al.* Quantum Nonlinear Optics with a Germanium-Vacancy Color Center
623         in a Nanoscale Diamond Waveguide. *Phys. Rev. Lett.* **118,** 223603 (2017).

624   62.   Siyushev, P. *et al.* Optical and microwave control of germanium-vacancy center spins in
625         diamond. *Phys. Rev. B* **96,** 081201 (2017).

626   63.   Green, B. L. *et al.* Neutral Silicon-Vacancy Center in Diamond: Spin Polarization and
627         Lifetimes. *Phys. Rev. Lett.* **119,** 096402 (2017).

628   64.   Rose, B. C. *et al.* Observation of an environmentally insensitive solid state spin defect in
629         diamond. Preprint at https://arxiv.org/abs/1706.01555 (2017).

630   65.   Tchernij, S. D. *et al.* Single-Photon-Emitting Optical Centers in Diamond Fabricated upon
631         Sn Implantation. *ACS Photonics* **4,** 2580–2586 (2017).

632   66.   Iwasaki, T. *et al.* Tin-Vacancy Quantum Emitters in Diamond. *Phys. Rev. Lett.* **119,**
633         253601 (2017).

634   67.   Koehl, W. F., Buckley, B. B., Heremans, F. J., Calusine, G. & Awschalom, D. D. Room
635         temperature coherent control of defect spin qubits in silicon carbide. *Nature* **479,** 84–87
636         (2011).

637   68.   Falk, A. L. *et al.* Polytype control of spin qubits in silicon carbide. *Nat. Commun.* **4,** 1819
638         (2013).

639   69.   Falk, A. L. *et al.* Electrically and Mechanically Tunable Electron Spins in Silicon Carbide
640         Color Centers. *Phys. Rev. Lett.* **112,** 187601 (2014).

641  70.  Whiteley, S. J. *et al.* Coherent Control of Spins with Gaussian Acoustics. Preprint at
642      https://arxiv.org/abs/1804.10996 (2018).

643  71.  Klimov, P. V, Falk, A. L., Christle, D. J., Dobrovitski, V. V & Awschalom, D. D. Quantum
644      entanglement at ambient conditions in a macroscopic solid-state spin ensemble. *Sci. Adv.*
645      **1,** e1501015 (2015).

646  72.  Christle, D. J. *et al.* Isolated electron spins in silicon carbide with millisecond coherence
647      times. *Nat. Mater.* **14,** 160–163 (2014).

648  73.  Simin, D. *et al.* Locking of electron spin coherence above 20 ms in natural silicon carbide.
649      *Phys. Rev. B* **95,** 161201 (2017).

650  74.  Nagy, R. *et al.* Quantum Properties of Dichroic Silicon Vacancies in Silicon Carbide.
651      *Phys. Rev. Appl.* **9,** 034022 (2018).

652  75.  Widmann, M. *et al.* Coherent control of single spins in silicon carbide at room
653      temperature. *Nat. Mater.* **14,** 164–168 (2014).

654  76.  Yang, L.-P. *et al.* Electron spin decoherence in silicon carbide nuclear spin bath. *Phys.*
655      *Rev. B* **90,** 241203 (2014).

656  77.  Seo, H. *et al.* Quantum decoherence dynamics of divacancy spins in silicon carbide. *Nat.*
657      *Commun.* **7,** 12935 (2016).

658  78.  Christle, D. J. *et al.* Isolated Spin Qubits in SiC with a High-Fidelity Infrared Spin-to-
659      Photon Interface. *Phys. Rev. X* **7,** 021046 (2017).

660  79.  de las Casas, C. F. *et al.* Stark tuning and electrical charge state control of single
661      divacancies in silicon carbide. *Appl. Phys. Lett.* **111,** 262403 (2017).

662  80.  Radulaski, M. *et al.* Photonic crystal cavities in cubic (3C) polytype silicon carbide films.
663      *Opt. Express* **21,** 32623–32629 (2013).

664  81.  Calusine, G., Politi, A. & Awschalom, D. D. Cavity-Enhanced Measurements of Defect
665      Spins in Silicon Carbide. *Phys. Rev. Appl.* **6,** 014019 (2016).

666  82.  Soykal, O., Dev, P. & Economou, S. E. Silicon vacancy center in 4H -SiC: Electronic
667      structure and spin-photon interfaces. *Phys. Rev. B* **93,** 081207(R) (2016).

668  83.  Kraus, H. *et al.* Magnetic field and temperature sensing with atomic-scale spin defects in
669      silicon carbide. *Sci. Rep.* **4,** 5303 (2014).

670  84.  Niethammer, M. *et al.* Vector Magnetometry Using Silicon Vacancies in 4H-SiC Under
671      Ambient Conditions. *Phys. Rev. Appl.* **6,** 034001 (2016).

672  85.  Heshami, K. *et al.* Quantum memories: emerging applications and recent advances. *J.*
673      *Mod. Opt.* **63,** 2005–2028 (2016).

674  86.  Könz, F. *et al.* Temperature and concentration dependence of optical dephasing,
675      spectral-hole lifetime, and anisotropic absorption in Eu3+:Y2SiO5. *Phys. Rev. B* **68,**
676      085109 (2003).

677  87.  Zhong, M. *et al.* Optically addressable nuclear spins in a solid with a six-hour coherence

678        time. *Nature* **517,** 177–180 (2015).

679   88.   O'Brien, C., Lauk, N., Blum, S., Morigi, G. & Fleischhauer, M. Interfacing
680        Superconducting Qubits and Telecom Photons via a Rare-Earth-Doped Crystal. *Phys.*
681        *Rev. Lett.* **113,** 063603 (2014).

682   89.   Williamson, L. A., Chen, Y.-H. & Longdell, J. J. Magneto-Optic Modulator with Unit
683        Quantum Efficiency. *Phys. Rev. Lett.* **113,** 203601 (2014).

684   90.   Utikal, T. *et al.* Spectroscopic detection and state preparation of a single praseodymium
685        ion in a crystal. *Nat. Commun.* **5,** 3627 (2014).

686   91.   Eichhammer, E., Utikal, T., Götzinger, S. & Sandoghdar, V. Spectroscopic detection of
687        single $Pr^{3+}$ ions on the $^3H_4 - {}^1D_2$ transition. *New J. Phys.* **17,** 083018 (2015).

688   92.   Nakamura, I., Yoshihiro, T., Inagawa, H., Fujiyoshi, S. & Matsushita, M. Spectroscopy of
689        single Pr3+ ion in LaF3 crystal at 1.5 K. *Sci. Rep.* **4,** 7364 (2015).

690   93.   Kolesov, R. *et al.* Optical detection of a single rare-earth ion in a crystal. *Nat. Commun.* **3,**
691        1029 (2012).

692   94.   Siyushev, P. *et al.* Coherent properties of single rare-earth spin qubits. *Nat. Commun.* **5,**
693        3895 (2014).

694   95.   Xia, K. *et al.* Optical and spin properties of a single praseodymium ion in a crystal.
695        Preprint at http://arxiv.org/abs/1706.08736 (2017).

696   96.   Fraval, E., Sellars, M. J. & Longdell, J. J. Dynamic Decoherence Control of a Solid-State
697        Nuclear-Quadrupole Qubit. *Phys. Rev. Lett.* **95,** 030506 (2005).

698   97.   Yin, C. *et al.* Optical addressing of an individual erbium ion in silicon. *Nature* **497,** 91–94
699        (2013).

700   98.   Zhong, T. *et al.* Nanophotonic rare-earth quantum memory with optically controlled
701        retrieval. *Science* **357,** 1392–1395 (2017).

702   99.   Zhong, T. *et al.* Optically addressing single rare-earth ions in a nanophotonic cavity.
703        Preprint at http://arxiv.org/abs/1803.07520 (2018).

704   100.  Dibos, A., Raha, M., Phenicie, C. & Thompson, J. Isolating and enhancing the emission
705        of single erbium ions using a silicon nanophotonic cavity. Preprint at
706        http://arxiv.org/abs/1711.10368 (2017).

707   101.  Muhonen, J. T. *et al.* Storing quantum information for 30 seconds in a nanoelectronic
708        device. *Nat. Nanotech.* **9,** 986–991 (2014).

709   102.  Tosi, G., Mohiyaddin, F. A., Huebl, H. & Morello, A. Circuit-quantum electrodynamics with
710        direct magnetic coupling to single-atom spin qubits in isotopically enriched 28 Si. *AIP*
711        *Adv.* **4,** 087122 (2014).

712   103.  Morse, K. J. *et al.* A photonic platform for donor spin qubits in silicon. *Sci. Adv.* **3,**
713        e1700930 (2017).

714   104.  Mi, X. *et al.* A coherent spin–photon interface in silicon. *Nature* **555,** 599–603 (2018).

715　105. Samkharadze, N. *et al.* Strong spin-photon coupling in silicon. *Science* **359,** 1123–1127
716　　　 (2018).

717　106. Steger, M. *et al.* Quantum Information Storage for over 180 s Using Donor Spins in a
718　　　 28Si 'Semiconductor Vacuum'. *Science* **336,** 1280–1283 (2012).

719　107. Saeedi, K. *et al.* Room-Temperature Quantum Bit Storage Exceeding 39 Minutes Using
720　　　 Ionized Donors in Silicon-28. *Science* **342,** 830–833 (2013).

721　108. Vazirani, U. & Vidick, T. Fully Device-Independent Quantum Key Distribution. *Phys. Rev.*
722　　　 *Lett.* **113,** 140501 (2014).

723　109. Barz, S. *et al.* Demonstration of Blind Quantum Computing. *Science* **335,** 303–308
724　　　 (2012).

725　110. Hensen, B. *et al.* Loophole-free Bell inequality violation using electron spins separated by
726　　　 1.3 kilometres. *Nature* **526,** 682–686 (2015).

727　111. Pfaff, W. *et al.* Unconditional quantum teleportation between distant solid-state quantum
728　　　 bits. *Science* **345,** 532–535 (2014).

729　112. Kalb, N. *et al.* Entanglement distillation between solid-state quantum network nodes.
730　　　 *Science* **356,** 928–932 (2017).

731　113. Yang, S. *et al.* High-fidelity transfer and storage of photon states in a single nuclear spin.
732　　　 *Nat. Photon.* **10,** 507–511 (2016).

733　114. Heshami, K. *et al.* Raman quantum memory based on an ensemble of nitrogen-vacancy
734　　　 centers coupled to a microcavity. *Phys. Rev. A* **89,** 040301 (2014).

735　115. Poem, E. *et al.* Broadband noise-free optical quantum memory with neutral nitrogen-
736　　　 vacancy centers in diamond. *Phys. Rev. B* **91,** 205108 (2015).

737　116. Kosaka, H. & Niikura, N. Entangled Absorption of a Single Photon with a Single Spin in
738　　　 Diamond. *Phys. Rev. Lett.* **114,** 053603 (2015).

739　117. Monroe, C. *et al.* Large-scale modular quantum-computer architecture with atomic
740　　　 memory and photonic interconnects. *Phys. Rev. A* **89,** 022317 (2014).

741　118. Nickerson, N. H., Li, Y. & Benjamin, S. C. Topological quantum computing with a very
742　　　 noisy network and local error rates approaching one percent. *Nat. Commun.* **4,** 1756
743　　　 (2013).

744　119. Cramer, J. *et al.* Repeated quantum error correction on a continuously encoded qubit by
745　　　 real-time feedback. *Nat. Commun.* **7,** 11526 (2016).

746　120. Cai, J., Retzker, A., Jelezko, F. & Plenio, M. B. A large-scale quantum simulator on a
747　　　 diamond surface at room temperature. *Nat. Phys.* **9,** 168–173 (2013).

748　121. Wang, Y. *et al.* Quantum Simulation of Helium Hydride Cation in a Solid-State Spin
749　　　 Register. *ACS Nano* **9,** 7769–7774 (2015).

750　122. Kong, F. *et al.* Direct Measurement of Topological Numbers with Spins in Diamond. *Phys.*
751　　　 *Rev. Lett.* **117,** 060503 (2016).

752  123.  Le Sage, D. *et al.* Optical magnetic imaging of living cells. *Nature* **496,** 486–489 (2013).

753  124.  Simpson, D. A. *et al.* Non-Neurotoxic Nanodiamond Probes for Intraneuronal
754        Temperature Mapping. *ACS Nano* **11,** 12077–12086 (2017).

755  125.  Thiel, L. *et al.* Quantitative nanoscale vortex imaging using a cryogenic quantum
756        magnetometer. *Nat. Nanotech.* **11,** 677–681 (2016).

757  126.  Pelliccione, M. *et al.* Scanned probe imaging of nanoscale magnetism at cryogenic
758        temperatures with a single-spin quantum sensor. *Nat. Nanotech.* **11,** 700–705 (2016).

759  127.  Gross, I. *et al.* Real-space imaging of non-collinear antiferromagnetic order with a single-
760        spin magnetometer. *Nature* **549,** 252–256 (2017).

761  128.  Barry, J. F. *et al.* Optical magnetic detection of single-neuron action potentials using
762        quantum defects in diamond. *Proc. Natl. Acad. Sci.* **113,** 14133–14138 (2016).

763  129.  Shi, F. *et al.* Single-protein spin resonance spectroscopy under ambient conditions.
764        *Science* **347,** 1135–1138 (2015).

765  130.  Lovchinsky, I. *et al.* Nuclear magnetic resonance detection and spectroscopy of single
766        proteins using quantum logic. *Science* **351,** 836–841 (2016).

767  131.  Casola, F., van der Sar, T. & Yacoby, A. Probing condensed matter physics with
768        magnetometry based on nitrogen-vacancy centres in diamond. *Nat. Rev. Mater.* **3,** 17088
769        (2018).

770  132.  Appel, P., Ganzhorn, M., Neu, E. & Maletinsky, P. Nanoscale microwave imaging with a
771        single electron spin in diamond. *New J. Phys.* **17,** 112001 (2015).

772  133.  Jakobi, I. *et al.* Measuring broadband magnetic fields on the nanoscale using a hybrid
773        quantum register. *Nat. Nanotech.* **12,** 67–72 (2016).

774  134.  Laraoui, A. *et al.* High-resolution correlation spectroscopy of (1)(3)C spins near a
775        nitrogen-vacancy centre in diamond. *Nat. Commun.* **4,** 1651 (2013).

776  135.  Kong, X., Stark, A., Du, J., McGuinness, L. P. & Jelezko, F. Towards Chemical Structure
777        Resolution with Nanoscale Nuclear Magnetic Resonance Spectroscopy. *Phys. Rev. Appl.*
778        **4,** 024004 (2015).

779  136.  Zaiser, S. *et al.* Enhancing quantum sensing sensitivity by a quantum memory. *Nat.*
780        *Commun.* **7,** 12279 (2016).

781  137.  Pfender, M. *et al.* Nonvolatile nuclear spin memory enables sensor-unlimited nanoscale
782        spectroscopy of small spin clusters. *Nat. Commun.* **8,** 834 (2017).

783  138.  Rosskopf, T., Zopes, J., Boss, J. M. & Degen, C. L. A quantum spectrum analyzer
784        enhanced by a nuclear spin memory. *npj Quantum Inf.* **3,** 33 (2017).

785  139.  Aslam, N. *et al.* Nanoscale nuclear magnetic resonance with chemical resolution. *Science*
786        **357,** 67–71 (2017).

787  140.  Glenn, D. R. *et al.* High-resolution magnetic resonance spectroscopy using a solid-state
788        spin sensor. *Nature* **555,** 351–354 (2018).

789  141.  Boss, J. M., Cujia, K. S., Zopes, J. & Degen, C. L. Quantum sensing with arbitrary
790        frequency resolution. *Science* **356,** 837–840 (2017).

791  142.  Schmitt, S. *et al.* Submillihertz magnetic spectroscopy performed with a nanoscale
792        quantum sensor. *Science* **356,** 832–837 (2017).

793  143.  Astner, T. *et al.* Coherent Coupling of Remote Spin Ensembles via a Cavity Bus. *Phys.*
794        *Rev. Lett.* **118,** 140502 (2017).

795  144.  Barfuss, A., Teissier, J., Neu, E., Nunnenkamp, A. & Maletinsky, P. Strong mechanical
796        driving of a single electron spin. *Nat. Phys.* **11,** 820–824 (2015).

797  145.  Lee, D., Lee, K. W., Cady, J. V., Ovartchaiyapong, P. & Jayich, A. C. B. Topical review:
798        spins and mechanics in diamond. *J. Opt.* **19,** 033001 (2017).

799  146.  Wolfe, C. S. *et al.* Off-resonant manipulation of spins in diamond via precessing
800        magnetization of a proximal ferromagnet. *Phys. Rev. B* **89,** 180406 (2014).

801  147.  Andrich, P. *et al.* Long-range spin wave mediated control of defect qubits in
802        nanodiamonds. *npj Quantum Inf.* **3,** 28 (2017).

803  148.  Najafi, F. *et al.* On-chip detection of non-classical light by scalable integration of single-
804        photon detectors. *Nat. Commun.* **6,** 5873 (2015).

805  149.  Grote, R. R. *et al.* Imaging a Nitrogen-Vacancy Center with a Diamond Immersion
806        Metalens.

807  Preprint at http://arxiv.org/abs/1711.00901 (2017).

808  150.  Steinert, S. *et al.* Magnetic spin imaging under ambient conditions with sub-cellular
809        resolution. *Nat. Commun.* **4,** (2013).

810  151.  Fedotov, I. V. *et al.* Electron spin manipulation and readout through an optical fiber. *Sci.*
811        *Rep.* **4,** 5362 (2015).

812  152.  Kehayias, P. *et al.* Solution nuclear magnetic resonance spectroscopy on a
813        nanostructured diamond chip. *Nat. Commun.* **8,** 188 (2017).

814  153.  van Dam, S. B., Humphreys, P. C., Rozpędek, F., Wehner, S. & Hanson, R. Multiplexed
815        entanglement generation over quantum networks using multi-qubit nodes. *Quantum Sci.*
816        *Technol.* **2,** 034002 (2017).

817  154.  Geng, J. *et al.* Experimental Time-Optimal Universal Control of Spin Qubits in Solids.
818        *Phys. Rev. Lett.* **117,** 170501 (2016).

819  155.  Zhou, B. B. *et al.* Accelerated quantum control using superadiabatic dynamics in a solid-
820        state lambda system. *Nat. Phys.* **13,** 330–334 (2017).

821  156.  Hirose, M. & Cappellaro, P. Coherent feedback control of a single qubit in diamond.
822        *Nature* **532,** 77–80 (2016).

823  157.  Unden, T. *et al.* Quantum Metrology Enhanced by Repetitive Quantum Error Correction.
824        *Phys. Rev. Lett.* **116,** 230502 (2016).

825  158.  Lesik, M. *et al.* Perfect preferential orientation of nitrogen-vacancy defects in a synthetic
826       diamond sample. *Appl. Phys. Lett.* **104,** 113107 (2014).

827  159.  Michl, J. *et al.* Perfect alignment and preferential orientation of nitrogen-vacancy centers
828       during chemical vapor deposition diamond growth on (111) surfaces. *Appl. Phys. Lett.*
829       **104,** 102407 (2014).

830  160.  Fukui, T. *et al.* Perfect selective alignment of nitrogen-vacancy centers in diamond. *Appl.*
831       *Phys. Express* **7,** 055201 (2014).

832

**Figure 1 | The NV center in diamond and nearby nuclear spins.**

**a)** Schematic of an NV⁻ center in diamond. The NV electron spin is coupled to proximal nuclear spins, such as its intrinsic $^{14}$N and lattice $^{13}$C. **b)** PL into the phonon sideband (PSB) as the detuning of a laser is scanned across the NV center ZPL (637.2 nm, $T = 7$ K). Resonant transitions to six spin-orbit excited states follow spin-selection rules and enable optical spin initialization, readout, and entanglement. Cycling transitions are labeled in green, while non-cycling transitions are labeled in orange. **c)** Coherence time $T_2$ of an NV center ensemble in isotopically-purified diamond (0.01% $^{13}$C) as a function of the number of pulses $n$ in a dynamical decoupling sequence, for various temperatures. **d)** Optically detected magnetic resonance (ODMR) spectrum taken by sweeping the frequency of a microwave field and measuring the NV center PL. The NV electronic transition $|m_s = 0\rangle$ to $|-1\rangle$ is split due to interaction with three strongly-coupled nuclear spins (hyperfine couplings: $^{14}$N, 2.16 MHz; $^{13}$C$_1$, 413 kHz; $^{13}$C$_2$, 89 kHz). **e)** (left) Single-shot readout protocol for strongly-coupled nuclear spins. The following block is repeated: the nuclear spin state (here for $^{13}$C$_1$) is correlated to the electron spin state by a controlled-not operation and then the electron state is read out. (right) Fluorescence time trace showing quantum jumps between nuclear spin states. **f)** Illustration of hyperfine coupling. If the electron spin is in $|m_s = 0\rangle$, the nuclear spin precesses with frequency $\omega_0$ about the applied magnetic field $\vec{B}$, while in $|m_s = -1\rangle$, the nuclear precession frequency $\omega_{-1}$ and axis are modified by the hyperfine components $A_{\parallel}$ and $A_{\perp}$, which depend on relative position between the nuclear and electron spin. **g)** Schematic for the evolution of a nuclear spin when the electron spin is repeatedly flipped by a dynamical decoupling sequence. When the spacing between the electronic $\pi$-pulses is in resonance with a half-period of the nuclear Larmor precession, the nuclear spin effectively rotates in opposite directions depending on the initial electron spin state. **h)** Data showing conditional control over a weakly-coupled nuclear spin by a dynamical decoupling sequence. The electron spin state determines the nuclear rotation direction, while the number of pulses $N$ determines the total rotation angle.

Figures reproduced from: a) Ref. [23]; b) Ref. [10]; c) Ref. [25]; d) e) Ref. [30]; f) Ref. [37]; g) Ref. [38]; h) Ref. [40].

862 **Figure 2 | The silicon vacancy center in diamond and color centers in silicon carbide.**

863 **a)** Orbital structure of the SiV⁻ center, showing doublets in both the ground and excited state that
864 are split by spin-orbit coupling. **b)** PL spectrum of the SiV⁻ center in diamond, displaying four zero-
865 phonon lines corresponding to transitions labeled in a) (linewidths shown are limited by
866 spectrometer resolution). The SiV⁻ center's high Debye-Waller factor (~70% of total emission into
867 ZPLs at 4 K), lifetime-limited optical linewidths, and robustness to spectral diffusion make it an
868 attractive source for indistinguishable photons. **c)** Scanning electron micrograph (SEM) of
869 diamond nanophotonic cavities with SiV centers incorporated by precision ion implantation
870 (upper). The orbital states of two SiV centers can be entangled by the emission of a single
871 indistinguishable Raman photon into a nanophotonic waveguide (lower). This entangled state is
872 observed by its enhanced photon scattering statistics. **d)** Structure of 4H-SiC, showing two non-
873 equivalent atomic sites for the Si and C atoms (h – quasihexagonal, k – quasicubic). There are
874 two inequivalent forms of the silicon vacancy ($V_{si}$) and four inequivalent forms of the divacancy
875 ($V_C$-$V_{Si}$) in 4H-SiC. **e)** PL excitation measurement by scanning the detuning of a laser about the
876 ZPL of a single divacancy (hh) in 4H-SiC (1132 nm, $T$ = 8 K). The excited state fine structure and
877 optical spin-selection rules are analogous to the NV center in diamond, but involve photons with
878 near-telecom wavelengths. **f)** Rabi oscillation of a single divacancy spin. The high readout
879 contrast of 94% is enabled by resonant excitation of the $|0\rangle \rightarrow |E_y\rangle$ cycling transition. **g)** Room-
880 temperature Rabi oscillations of a single silicon vacancy spin (V2 center) in 4H-SiC as a function
881 of microwave power.

882 Figures reproduced from: a) b) Ref. [53]; c) Ref. [60]; d) Ref. [72]; e) f) Ref. [78]; g) Ref. [75].

883

884     **Figure 3 | Rare-earth ions in crystals and optically-active donors in silicon.**

885     **a)** Fluorescence excitation spectrum of a single $Pr^{3+}$ ion in a YSO microparticle, resolving the
886     hyperfine structure of the $^3P_0$ excited state ($T$ = 4 K). **b)** ODMR for the ground state hyperfine
887     sublevels of a single $Pr^{3+}$ ion in bulk YAG. Readout is achieved by a two-photon upconversion
888     process. The relatively broad ODMR linewidth ($\sim 2\pi \cdot 1.5$ MHz) indicates that the coherence of the
889     $Pr^{3+}$ nuclear spin in YAG is limited by spin-lattice relaxation at 4 K. **c)** ODMR between the electron-
890     nuclear spin singlet ($S_0$) and triplet ($T_-, T_0, T_+$) states of an ensemble of singly-ionized $^{77}Se$ double
891     donors in silicon. Data are taken by first optically pumping the ensemble into the $S_0$ state and then
892     measuring the relative absorption of the $T \leftrightarrow \Gamma_7$ transition (2902 nm) at 2 K and in Earth's
893     magnetic field (70 $\mu T$).

894     Figures reproduced from: a) Ref. [91]; c) Ref. [95]; b) Ref. [103].

895

**Figure 4 | Quantum registers for quantum networks and computing.**

**a)** Photograph showing three lab stations A, B, C at the Delft University of Technology. An entangled state between two NV centers separated by 1.3 km (stations A and B) is generated by sending spin-entangled photons to station C. **b)** Experimental schematic for remote entanglement. Indistinguishable ZPL photons from NV centers at stations A and B are overlapped onto a beam-splitter at station C, where a particular sequence of single photon detections heralds the generation of an entangled state. High-fidelity and fast single-shot spin readout enables a loophole-free test of the Bell inequality. **c)** Principle of entanglement distillation. Two lower quality remote entangled states can produce a single higher fidelity entangled state through local operations and classical communication. **d)** Experimental implementation of entanglement distillation using an NV center electron spin (communication qubit) and a weakly coupled $^{13}$C nuclear spin (memory qubit) at each node. The success of the distillation protocol, resulting in a higher fidelity entangled state between the memory qubits, is probabilistic, but is heralded by a particular measurement outcome on the communication qubits. **e)** Scheme for storage of a photon polarization state by the $^{14}$N nuclear spin of the NV center. The NV center is first prepared in an electron-nuclear Bell state. At zero magnetic field, the electronic levels $|\pm 1\rangle_e$ and $|A_1\rangle$ form a degenerate $\Lambda$ system, and the entangled absorption of a resonant photon transfers the photon polarization onto the phase of the nuclear superposition state. Successful absorption is heralded by single-shot spin readout. **f)** Data showing the faithful mapping of the photon polarization onto the nuclear superposition state. **g)** Quantum error correction using a register of three strongly-coupled nuclear spins. This code can surpass the fidelity of no error correction (blue circles) when a single error occurs of one of the qubits (green triangles) or when errors occur with equal probability on all three qubits (red circles).

Figures reproduced from: a) b) Ref. [110]; c) d) Ref. [112]; e) f) Ref. [113]; g) Ref. [30].

921 **Figure 5 | Diverse modalities and applications of quantum sensing with impurity spins.**

922 **a)**. Wide-field magnetic imaging of magnetotactic bacteria on top of a dense array of shallow NV
923 centers. The colorbar depicts the magnetic field projection along the [111] NV axis. The inset
924 shows the bright-field image for the same region. **b)** Intracellular temperature map of primary
925 cortical neurons using nanodiamonds (ND) incorporated by cellular uptake. The color code
926 denotes the change in measured temperature for each ND in response to a reduction in
927 environmental temperature. **c)** An atomic force microscope using a diamond scanning tip with a
928 single NV center at its apex enables magnetic field imaging with a spatial resolution of tens of
929 nanometers, determined by the NV to sample distance. **d)** Magnetic stray field image capturing
930 the non-collinear magnetic order of a multiferroic $BeFeO_3$ thin film. The colorbar is related to the
931 change in local magnetic field from a reference value. **e)** Geometry for NMR on nanoscale sample
932 volumes with a near surface NV magnetic sensor. **f)** NMR chemical shift for $^{19}F$ nuclei in PFPE
933 (fomblin) detected by an NV spin. The high spectral resolution of this experiment is achieved by
934 using a long-lived nuclear memory to extend the phase acquisition time.

935 Figures reproduced from: a) Ref. [123]; b) Ref. [124]; c) d) Ref. [127]; e) f) Ref. [139].

936

937    **Figure 6 | Photonic engineering for solid-state spins.**

938    **a)** Miniaturized Fabry-Pérot microcavity containing a thin diamond membrane. Nano-positioning
939    a moveable distributed Bragg reflector (DBR) mirror allows *in situ* tuning of the cavity resonance
940    frequency and antinode location. Emission of the NV center into the ZPL is boosted to 46% at 4
941    K. **b)** SEM image of a photonic crystal cavity in 3C-SiC. The starting material is grown as a
942    heteroepitaxial thin film on silicon, allowing for effective substrate removal. Collected PL from a
943    divacancy ensemble in the cavity is enhanced by a factor of 30. **c)** Implementation of a photonic
944    integrated circuit featuring on-chip waveguides and superconducting nanowire single photon
945    detectors. This architecture could provide a route toward scalable, on-chip entanglement of
946    impurity spins.

947    Figures reproduced from: a) Ref. [50]; c) Ref. [81]; b) Ref. [148].

**a)** 

**b)**

PSB fluorescence (a.u.)

$|E_{1,2}\rangle$   $|E_y\rangle$   $|E_x\rangle$   $|A_1\rangle$   $|A_2\rangle$

$|{\pm}1\rangle$   $|0\rangle$   $|0\rangle$   $|{\pm}1\rangle$   $|{\pm}1\rangle$

Laser Detuning (GHz)

**c)**

Coherence time $T_2$ (ms)

$T_2^{max}(T_1) = 0.53\,T_1$

$T_2^{max}$ (ms)

$T_1$ (ms)

300 K
240 K
190 K
160 K
120 K
77 K

$T_2^{max} = 580$ ms

$T_1 = 506$ ms
$T_1 = 77$ ms
$T_1 = 30$ ms
$T_1 = 12$ ms
$T_1 = 6$ ms

Number of pulses $n$

**d)**

$^{14}$N

$^{13}$C$_1$

$^{13}$C$_2$

Signal

Microwave Frequency (GHz)

0.5 MHz

**e)**

$n$

NV $|0\rangle$

$^{14}$N

$^{13}$C$_1$

$^{14}$N

Fluorescence

$|0,-1\rangle$

$|+1\rangle$

0.05 s

$^{13}$C$_1$

Fluorescence

$|-\frac{1}{2}\rangle$

$|+\frac{1}{2}\rangle$

0.5 s

Time

**f)**

$A_\perp$

$A_\parallel$

$\omega_0$   $\omega_{-1}$

$n$

$\vec{B}$

**g)**

$\pi/2$ pulse   $\pi$ pulse   $\pi$ pulse

$N=1$   $\mathbf{B}_0$   $\mathbf{B}_1$   $N=1$   $N=3$

$|0\rangle$   $|{\uparrow}\rangle_{nuc}$

$m_s = 0$   $m_s = 1$

$|\psi\rangle_{el}$

$|1\rangle$   $\mathbf{B}_1$   $\mathbf{B}_0$   $N=1$   $N=3$

$|{\uparrow}\rangle_{nuc}$

$m_s = 1$   $m_s = 0$

**h)**

$\langle Y \rangle$

$m_s = -1$

$m_s = 0$

Conditional $R^{N}_{\pm X}{}^{\varphi}$

Number of pulses $N$

**a)** Pr$^{3+}$:YSO

Fluorescence (counts/s) vs Laser Frequency Detuning (MHz)

$^3P_0$

$^3H_4$

5.65 MHz   2.96 MHz

**b)** 4f5d

$^3P_0$   ±1/2   ±3/2   ±5/2

$^3H_4$ (1)   ±1/2   ±3/2   ±5/2

RF

Pr$^{3+}$:YAG

Fluorescence Intensity (a.u.) vs RF frequency (MHz)

**c)** $^{28}$Si:$^{77}$Se$^+$

Transmittance vs $f$ (MHz)

$S_0 \Leftrightarrow T_-$   $S_0 \Leftrightarrow T_0$   $S_0 \Leftrightarrow T_+$

Γs

1s:$T_2$   2.27 meV   Γ7

Spin-valley

427 meV

1s:$A$   6.87 μeV   Γs

Hyperfine   $T$   $S_0$

a) 

b)

c) Entanglement distillation

d)
1. Generate remote entangled state
Communication qubit (NV e⁻ spin)
Memory qubit ($^{13}$C nuclear spin)

2. Swap to memories

3. Generate another remote entangled state

4. Distillation via local operations

e) $|A_1\rangle$

$|E_x\rangle$

$\sigma_+$ $\sigma_-$

$|-1\rangle_e |+1\rangle_n$ $|+1\rangle_e |-1\rangle_n$

$|-1\rangle_e |0\rangle_n$ $|+1\rangle_e |0\rangle_n$

$|0\rangle_e |+1\rangle_n$ $|0\rangle_e |-1\rangle_n$

$|0\rangle_e |0\rangle_n$

f)

Stored phase in nuclear state (°)

Input photon polarization phase (°)

g)

Process fidelity, $f$

Error probability, $p$

Measured, no QEC
Ideal, no QEC
Measured, single qubit error, with QEC
Measured, multiple qubit errors, with QEC
Ideal, multiple qubit errors, with QEC

NV
Hyperfine
$^{13}C_1$ $^{14}N$ $^{13}C_2$

a)



b)



c) Microscope objective

Diamond tip

RF

NV e-spin

BiFeO₃

d)



e)

diamond

30 nm

f)

$^{19}$F

F₃CO

CF3

×10 +0.5

×5 +0.2

normalized |FFT|²

chemical shift (ppm)

**a)**
DBR
SiO₂
Diamond
0 - 3 µm
DBR
1 µm
SiO₂
Nanopositioners
z
y
x

**b)**
3C-SiC
Thin film
Cavity
Photonic Crystal
3 µm

**c)**
Waveguide-detectors
B2
B1
Input B
Fibre
Input A
A2
A1
To RF readout
I II III IV V VI