

Diamond integrated quantum photonics

Diamond is a leading contender as the material of choice for the quantum computer industry. This potential arises mainly from the quantum properties of color centers in diamond. However, before diamond can realize its full potential, the technology to fabricate and sculpt diamond as well as, if not better than, silicon must be developed. A comprehensive processing capability for diamond that will allow the fabrication of qubits and their associated photonic structures is required. Here we describe the remarkable properties of diamond color centers, and the techniques being developed to engineer qubits and sculpt monolithic structures around them. Finally we outline some of the new proposals that use engineered diamond to realize tasks not possible with existing technologies.

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To complete the quantum revolution that began with transistors and integrated circuits in the 20th century requires the development of devices that exploit all the properties of quantum mechanics. Foremost among the challenges are devices that work in the quantum regime, i.e. one photon, one atom, or a few quantized modes of oscillation. Although some truly quantum devices have been demonstrated in laboratory settings, there are very few examples of practical commercial devices. This, then, is a major challenge for 21st century technology: the construction of platforms and architectures to harness the full power of quantum mechanics through coherent control for practical applications. For solid-state implementations of quantum information processing,

the challenges (at the few qubit level) are essentially ones of materials and control. Materials challenges are also vital for what are traditionally considered *non-material* implementations – e.g. the miniaturization of linear optical quantum computing requires the construction of ultra-low-loss integrated optical circuits, which is a serious materials challenge¹. Amongst the many potential platforms for practical solid-state quantum devices, diamond is rapidly becoming a material of choice.

Every property of matter has an extremum, one material with properties that exceed all others. Often, that material is diamond. Diamond is the hardest material, has the highest Young's modulus, the greatest thermal conductivity, the highest number density of atoms,

and the widest optical bandgap. Additionally it is biocompatible², chemically inert, and can be doped to alter its electrical and optical properties. Why, then, is diamond not routinely being used in computers, windows, solar cells, and lenses? In fact, diamond is increasingly finding its place in such applications, but mainly as a niche product. There are good reasons why diamond is not utilized more; these include cost, quality, workability, and the constant advances in rival materials. However, researchers worldwide are eroding these constraints. In this review we concentrate in particular on the emerging applications based on the coherent control of quantum effects.

The desirable quantum properties of diamond originate in part from the fact that the three common sources of decoherence are minimized in diamond. These sources of decoherence are nuclear spins (limited by low ¹³C concentration and impurities), low electron concentrations (because diamond is a wide bandgap insulator), and low phonon scattering at room temperature³. In addition, it has large dipole photostable color centers, which add an extra dimension not shared by, for example, silicon. One in particular, the negatively charged nitrogen-vacancy color center (NV⁻), has attracted significant interest. Our discussion starts with a summary of some of the properties and quantum demonstrations arising from this center. But NV⁻ is by no means the only color center in diamond, and probably not the best one for most applications. We also discuss some of the other promising contenders (especially Ni- and Si-related centers), and how they can be formed.

Demonstrations of quantum effects in diamond have concentrated mainly on native optical centers which occur fortuitously and are explored using single-center confocal microscopy and related techniques. Going beyond such proof-of-concept demonstrations requires a toolkit for building integrated optoelectronic structures, preferably monolithically. There are several approaches to realizing such a toolkit, which we discuss along with the challenges and opportunities they present. Structures that have been realized in diamond include waveguides, photonic bandgap structures, and resonators, and we will review some of the progress to date, highlighting the requirements for more ambitious future devices.

Finally, we describe the new quantum technologies that diamond promises, highlighting some of the proposals that use diamond-containing color centers as fundamental building blocks in quantum machines. These proposals include sources of nonclassical light, quantum computers, and quantum simulators.

Color centers in diamond

Diamond is the widest bandgap semiconductor known, with a bandgap of around 5.5 eV stretching from the ultraviolet to the far infrared. Because of this, diamond is transparent in the visible region, and any colors observed arise from defects – impurities, dislocations, vacancies, and complexes – which create electronic energy levels within the bandgap. Because of their importance to gemology, many such defects

have been catalogued (well over 500⁴). However, despite such studies, there has been surprisingly little work to completely characterize the quantum properties of these centers and comparatively little work that extends beyond the visible spectrum. Because of the growing importance of color centers we expect this situation to change dramatically in the coming years.

One of the most intensively studied color centers in diamond is the NV color center. Nitrogen is the most common impurity in diamond⁵. The NV color center shows at least three remarkable properties: it is extremely bright and a single center can be seen at room temperature; it can be spin polarized by shining white light on it; and its spin state can be read out by using the difference in fluorescence between the bright spin zero ground state and the relatively dark spin ± 1 ground state. The NV color center comprises a substitutional N next to a vacancy in the lattice. It occurs in at least two forms, neutral and negatively charged. Here we will only refer to the NV⁻ version as it is in this form that it is most useful for quantum information purposes. The first published spectrum of the NV⁻ center was by Clark and Norris⁶ in 1971, and Davies and Hamer⁷ identified this transition as being likely due to a NV complex in 1976. Remarkably, the electronic structure of the center has still not been definitively determined, although considerable progress has been made in that regard (see, for example, Manson *et al.*⁸ for perhaps the most complete description), and there is ongoing work into *ab initio* modeling of its structure^{9–12}.

Ab initio calculations showing the electron densities of the lowest unoccupied and highest occupied molecular orbitals (LUMO/HOMO) are shown in Fig. 1a and b, respectively. The C_{3v} symmetry of the center can be seen in both configurations, and the electronic configurations appear to be localized in the center. The electron densities of HOMO/LUMO orbitals are equally and symmetrically distributed among three C atoms neighboring the vacancy; however, the electron density around the N is much higher for the HOMO than the LUMO, giving rise to the observed dipole moment. The NV⁻ center has a zero phonon line at 637 nm (1.945 eV), which is the

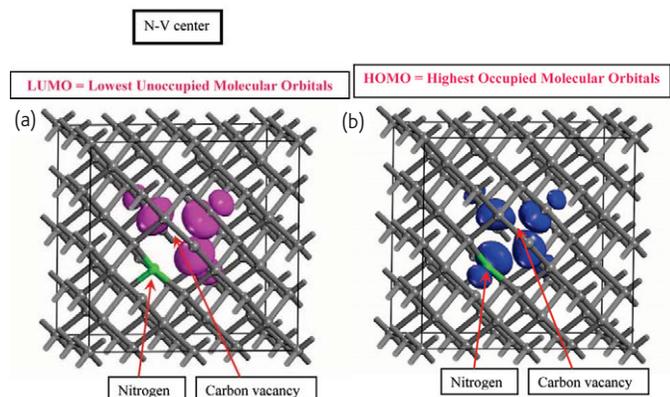


Fig. 1 Orbital isosurfaces for NV⁻ center in diamond: (a) lowest unoccupied molecular orbital (LUMO); (b) highest occupied molecular orbital (HOMO). (Adapted from¹³.)

HOMO/LUMO energy gap, with pronounced phonon sidebands. It has a large dipole moment which gives rise to a relatively rapid spontaneous emission rate (12 ns) and pronounced Stark shift^{14,15}, which has been also observed in spectrally resolved centers^{16,17}. An energy level diagram of some of the latest models of the ground and excited state structures is shown in Fig. 2a, with a low-temperature fluorescence spectrum in Fig. 2b.

One of the most dramatic features of the NV⁻ center is its spin polarization properties. The center is a six-electron paramagnetic system, which can be treated as a two-hole system to form a closed shell that requires eight electrons. The system therefore has an integer spin of $S = 1$ with trigonal (C_{3v}) symmetry. The optical excitation study of Loubser and van Wyk¹⁸ detected electron spin resonance (ESR) signals of a spin polarized triplet ($S=1$), confirming this picture. At zero magnetic field, the ground state degeneracy is lifted by a crystal field interaction, which gives rise to a 2.88 GHz separation between the $m_s = 0$ and $m_s = \pm 1$ states. Under green or white illumination, an optical pumping cycle gives rise to a greatly enhanced population in the electronic spin $m_s = 0$ ground state at the expense of the $m_s = \pm 1$ states. This effect was characterized in a series of beautiful Raman heterodyne measurements performed by He *et al.*^{19,20} and can be likened to 'laser cooling' of the NV⁻ far below the temperature of the surrounding lattice. It is because the transitions in NV⁻ are decoupled from the phonon modes of diamond that this optical pumping is so effective, even at room temperature, enabling the beautiful demonstrations of spin coherence in room-temperature samples.

Before 2004, there was steady progress uncovering the properties of NV⁻ centers, however the scene changed dramatically with the publication of a landmark study by the group of Wrachtrup²¹ which demonstrated single center readout, spin polarization (initialization), and coherent oscillations of a single center *at room temperature*. The

sample used contained diamond nanocrystals (produced by standard high-pressure/high-temperature methods) embedded into poly(methyl methacrylate) and spun onto a surface. Using confocal microscopy, the sample is scanned until a suitable center is found, which is then studied. The formation and use of diamond nanoparticles was reviewed by Krueger²². Such fortuitous centers can lead to dramatic results that open a new field, as this study assuredly did; however, they do not inform as to what the properties of 'typical' centers are.

One of the most important parameters for quantum information processing is the coherence time, and we may understand this parameter by exploring the meaning of coherence in the quantum setting. Just as the fundamental unit of classical information is the bit, there is a fundamental unit of quantum information, the qubit. One distinction between bits and qubits is that a qubit can be placed in a superposition of states, i.e. a bit can be either 0 or 1, but a qubit can be in a superposition state, $|\Psi\rangle = \alpha|0\rangle + \beta|1\rangle$, where the kets are the two 'values' that can be taken by the qubit, and α and β represent the (in general complex) coefficients describing the superposition. The coherence time is the characteristic time taken for the fragile quantum information to leak into the environment, or alternatively can be seen as the time for a quantum superposition state to become a mixed state, i.e. a state with a classical probability of being found in either state, but without a well-defined quantum phase. In our example above, decoherence takes the superposition state $|\Psi\rangle$ to a mixed state which has the probability $|\alpha|^2$ of being found in $|0\rangle$ and $|\beta|^2$ of being found in $|1\rangle$. Decoherence erodes the complex phases, and much of the power of quantum computing.

If coherence times are longer than some threshold (which depends precisely on the architecture and the form of error correction used), then active quantum error correction can be used to protect the information for arbitrarily long times, at the expense of increased

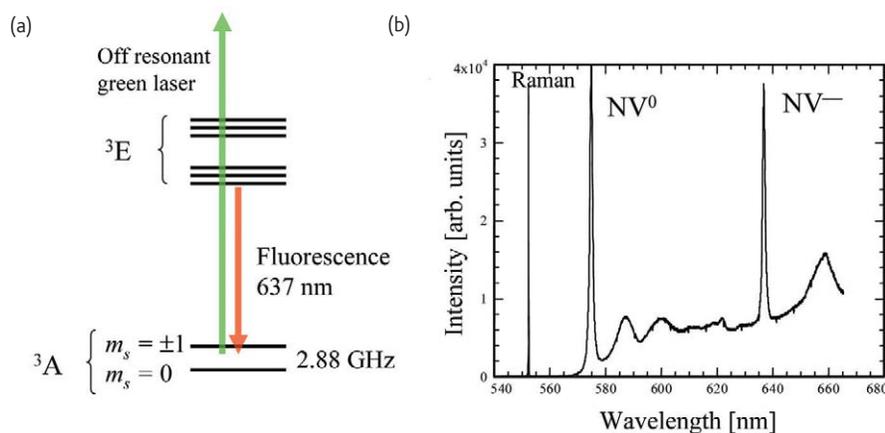


Fig. 2 (a) Energy level structure of the NV⁻ center showing ground and excited states, showing green, off-resonant excitation pulse used for readout, and the fluorescence at 637 nm. The green laser excites the center from the ³A ground state to some phononic sideband of the excited state, which de-excites nonradiatively to the ³E manifold. These transitions are predominantly spin conserving at zero magnetic field and zero strain, and the $m_s = 0$ state is slightly brighter than the $m_s = \pm 1$ state, leading to effective spin readout. Slight spin nonconserving transitions are also observed which lead to the spin polarization in the $m_s = 0$ state under broad band illumination. (Adapted from⁴².) (b) Typical fluorescence spectrum taken at low temperature of the NV⁻ center at 637 nm with first phonon sideband, under 514 nm illumination. Also shown are the first order Raman line at 550 nm and the pronounced NV⁰ emission at 575 nm with phonon sidebands.

number of qubits. A simple target threshold is that 10^4 operations can be performed before decoherence for quantum error correction to be possible²³. In 2004, the measured coherence time of $2 \mu\text{s}$ ²¹ was at that time a record for solid-state coherence of a single particle; however, by 2003 Kennedy *et al.*³ had demonstrated a coherence time of $58 \mu\text{s}$ in an ensemble of NV^- centers, also at room temperature, and showed that these coherence times were sample dependent.

Recent results in implanted samples have been even more impressive, with room-temperature coherence times of 0.35 ms reported²⁴ in ultra-low-N synthetic diamond using pulse-echo techniques. Given the fundamental single qubit rotation time of around 30 ns , this already meets the required precision for single qubit operations, if scalable two-particle interactions were possible. Childress *et al.*²⁵ have demonstrated that ^{13}C nuclear spins limit the NV^- coherence times. In certain applications, however, the ^{13}C can be used as a resource^{26–28} and this is discussed later. The key to longer coherence times is in materials control, especially obtaining synthetic diamond samples with very low impurity (especially N) concentrations and ideally pure ^{12}C .

Many quantum protocols require two-photon interference. Interference between two photons is an entirely nonclassical effect that manifests in the Hong–Ou–Mandel dip²⁹. The importance of nonclassical interference will be discussed later; however, it is important to realize that for two photons to interfere, they must be identical, i.e. every degree of freedom (energy, polarization, direction, etc.) must be the same. When an NV^- center decays, it can do so by emitting a single photon, the zero-phonon line (ZPL), or it can emit a photon and several phonons, giving rise to phonon sidebands. In such protocols the phononic sidebands can be highly problematic, with a NV^- Debye–Waller factor of only a few percent³⁰, as seen in Fig. 2b. This factor measures the ratio of intensity in the ZPL to that in phonon sidebands. An equivalent measure of the ratio of sideband to ZPL transitions is the Huang–Rhys factor, and these are related by $D = \exp(-H)$, where D is the Debye–Waller factor and H the Huang–Rhys factor. These phonons carry away energy and also information about the emitted photons, which means that two-photon interference cannot occur with perfect fidelity. Ways of overcoming this limitation are discussed below.

The large dipole moment also introduces limitations. It means that the center is extremely sensitive to strain and mobile charges, which give rise to spectral diffusion (especially in polycrystalline samples) of single centers. These two factors contribute to the large accepted value for the inhomogeneous linewidth of NV^- centers in diamond of 750 GHz . However, there is evidence that this linewidth is not a fundamental limit. Spectra obtained from N implanted into high-quality synthetic diamonds have shown reduced NV^- inhomogeneous linewidths, below 200 GHz . The ultimate limit has yet to be reached in ensembles of centers³¹. Stable individual centers with little or no inhomogeneous broadening have been identified in cryogenic

experiments (e.g. Santori *et al.*³²), although at room temperature, spectral diffusion of individual centers gives rise to large effective inhomogeneous broadening.

Demonstrations of quantum coherence to date have mainly used the ground spin states. Unlike the excited states, the ground state properties are very reproducible from center to center, which is evidenced by the raft of ensemble work done on NV^- diamond. Their reproducibility comes about as they are magnetic dipoles in a rigidly defined, nonmagnetic material. Quantum coherence is preserved because phonons do not readily couple to the centers, with decoherence predominantly arising due to proximal nuclear spins (especially ^{13}C). Because of this well-defined ground state splitting, there are possible applications for NV^- diamond in ultra-sensitive magnetometry³³.

One of the first quantum applications of NV^- diamond has been in single-photon sources for quantum key distribution. This derives from the large dipole moment of NV^- in diamond, and the ability to find isolated centers or implant N at low densities. The first demonstration of single-photon emission in NV^- diamond was by Kurtsiefer *et al.*³⁴ in 2000, which has been followed by demonstrations of quantum key distribution (e.g. by Beveratos *et al.*³⁵). One novel possibility for diamond-based single-photon sources is to combine nanocrystals containing NV^- with optical fibers, either by adhesion³⁶ or growth³⁷. Indeed, the first diamond-based commercial single-photon source is now becoming available³⁸.

By embedding the NV^- within a cavity in the strong Purcell regime, it is, in principle, possible to reduce the contribution from the phonon sidebands and also effectively decrease the lifetime of the center into the tens of picoseconds regime³⁹. Such developments are important for the continued development of NV^- centers for quantum key distribution and long-range coupling between centers (see below).

NV^- is currently the most important center for quantum information applications. As yet, no other diamond color centers have demonstrated optical coherent effects such as electromagnetically induced transparency (EIT)^{40,41} and coherent population trapping^{32,42}. However, there are now several demonstrated single-photon emitters in diamond. For reasons of space we will only discuss two of the most immediately promising, the Ni-related NE8 and Si-vacancy (Si-V) centers, both of which seem to be superior single-photon sources for quantum key distribution and standards purposes⁴³.

Ni is a common impurity in synthetic diamond as it acts as a catalyst for growth. Accordingly, Ni-related centers are also found in diamond, and the NE8 center in particular has proven especially interesting for single-photon purposes. NE8 was first identified in 1999 by Nadolinny *et al.*⁴⁴ in a high-pressure, high-temperature (HPHT) sample, although they report its existence in natural diamonds also. The NE8 center comprises a single Ni atom in either substitutional or split-vacancy interstitial position, with four neighboring N atoms, as seen in the *ab initio* calculations reproduced in Fig. 3. As with the

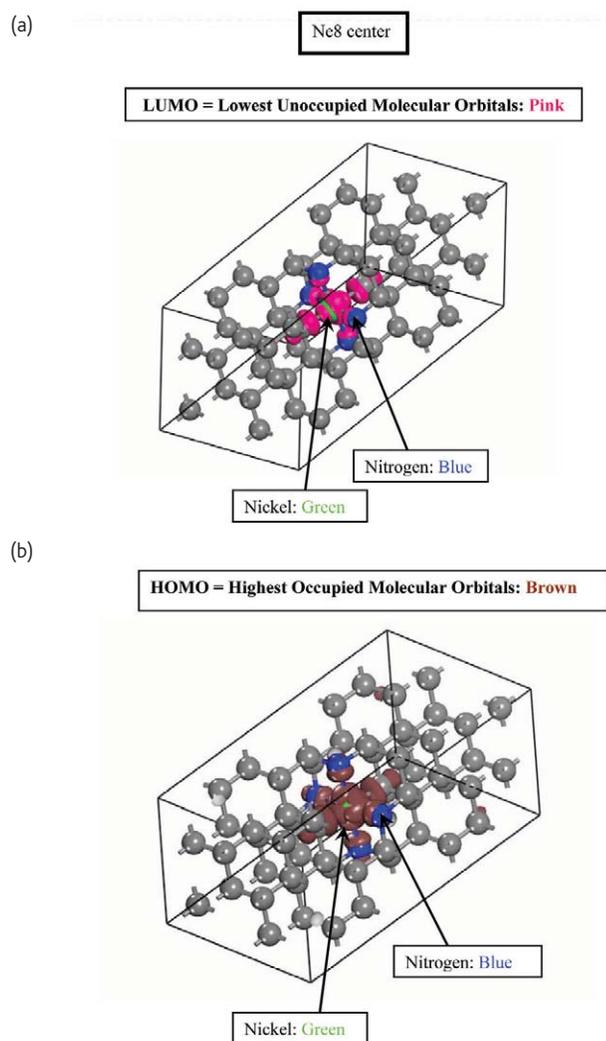


Fig. 3 Orbital isosurfaces for Ni-4N complex (NE8) center in diamond: (a) lowest unoccupied molecular orbital (LUMO); (b) highest occupied molecular orbital (HOMO). (Adapted from¹³.)

NV⁻ center, there is a large dipole moment arising from the charge separation of the HOMO and LUMO. The short lifetime of around 2 ns gives rise to a source with potentially higher bit rate than for NV⁻; more important factors are the greatly increased Debye–Waller factor of 0.7, an emission bandwidth of 1.2 nm, and fluorescence shifted to ~800 nm, which make this a far more desirable single-photon source for quantum key distribution purposes. The potential of NE8 for single-photon work was first demonstrated by Gaebel *et al.*⁴⁵, and subsequently a triggered source for quantum key distribution (QKD) was realized^{46,47}. Deliberate fabrication of NE8 centers has proven far more difficult than for NV, although it has been formed in chemical vapor deposition (CVD) samples by adding Ni to the initial slurry used to nucleate diamond growth⁴⁸.

The final center that we will briefly mention is the Si-V center. The advantage of Si-V over NE8 is that it can be made via ion implantation

(not yet achieved for individual Ni-related centers), which gives rise to the possibility of placing centers in engineered structures. The ZPL emission wavelength of 738 nm, lifetime of 1.8 ns and emission bandwidth of 5 nm make this center a good candidate for practical applications. The center was identified from investigations of synthetic diamonds grown from melts containing Si⁴⁹, and later demonstrated as a room-temperature single-photon source by Wang *et al.*⁵⁰. Wang observed a discrepancy between their detected fluorescence and that expected from an emitter with the measured lifetime. Following the results of Feng and Schwartz⁵¹, they attributed the reduction in fluorescence to nonradiative transitions from the excited to the ground state. If so, then the applicability of this source to practical quantum applications will be seriously limited. Nevertheless, the motivation to explore new centers is strong given the outstanding results to date. We have barely begun to study the range of centers available.

Controlled creation of NV and other optical centers in diamond

Much of the fundamental work to date has been performed on native color centers in either single-crystal or nanodiamond particles. Ion implantation offers the possibility of direct placement of NV centers in diamond with nanometer-scale precision using technologies developed for the placement of single P donors in Si⁵². However, unlike the case of Si, diamond offers two alternative routes for the creation of individual NV centers. The first involves implantation which can be used to create vacancies in N-rich (intrinsic) type Ib diamond. Annealing above 600°C initiates migration of the vacancies to the intrinsic substitutional N sites to form NV⁰ and NV⁻ centers³¹. Implantation of Ga, He, C and other ions have been employed for NV⁻ formation with this method, as has irradiation with neutrons and electrons. It is important to emphasize that in this case the native N is already distributed randomly in the diamond crystal, and therefore only limited control over NV⁻ center formation can be achieved. Also residual damage within the implanted volume may limit the applicability of this method. This method seems to be particularly well suited for the creation of ensembles of NV centers for use in, for example, EIT-based applications.

The second method involves direct implantation of N ions into type IIa diamond followed by thermal annealing^{53,54}. This method performs well on the availability of diamond materials with extremely low (<0.1 ppm) N concentration. This is because each implanted N ion creates many tens to thousands of vacancies along the ion track. Any native N that lies within the cylinder of damage created by an impinging ion can potentially be converted into a NV center upon annealing. The deeper the implantation, the higher the level of purity required to ensure that it is the implanted, rather than the native, N which is converted into the NV center. Native N can be distinguished from implanted N by implanting ¹⁵N rather than the naturally occurring ¹⁴N⁵⁵. Fortunately, ultrapure diamond materials containing

parts per billion levels of N and B are now commercially available via Element Six⁵⁶, which will allow high-precision implanted N devices.

Although the broad parameters for the production of NV⁻ centers in diamond are known, the level of control required over the implantation process for quantum information processing (QIP) applications exceeds that for diamond doped by ion implantation for electronic applications⁵⁷. This is a multiparameter problem in which the N content of the starting material, the implantation dose and energy, the implantation temperature, and the annealing conditions all play important roles. Unlike the case of doping for electronic applications, for QIP the implantation process needs to be both understood and optimized at the *atomic*, rather than at the *ensemble*, level. For example, the role of the proximity of surface and its termination (H or O terminated) on the stability of NV⁻ centers has not been considered in detail yet. To illustrate the importance of atomic control of implants, it was recently demonstrated that a single pair of strongly coupled spins in diamond can be optically initialized and read out at room temperature²⁴. To effect such strong coupling, close proximity of the two spins is required, but large distances from other spins are required to avoid deleterious decoherence. The scheme developed to reconcile these competing requirements is one in which coupled pairs of spins are created by implantation of N₂ dimers instead of N atoms. The decoherence time for these coupled centers was measured to be 0.35 ms at room temperature.

Finally, although the NV center is the most studied of all diamond optical centers, other optical centers have also shown great promise, especially for use as single-photon sources, and so strategies for their creation are required. In particular Ni-related centers in diamond have been used as single-photon sources, and here, as with other color centers, the availability of pure samples is essential. For example, although most CVD diamonds do not contain Ni as impurities, Ni interacts strongly with N to form a series of complexes (e.g. Ni, NiN, NiN₂, NiN₄), each of which displays different optical transitions. Thus it is hard to establish the role of Ni in diamond *per se* unless ultrapure samples are used. The same will be true of other color centers which have been created by ion implantation (e.g. Si and Xe⁵⁸).

Nanofabrication of diamond

The challenges of high-efficiency photon collection and ultimately quantum information processing require a nanofabrication toolkit for diamond at least as good as that available for silicon processing. The principle aspects to address are the creation (or identification) of the optical centers and the incorporation of the center into a nano-optical structure. Cavities are vital in collecting emitted photons, and allow for strong atom–photon interactions at the one-photon level. Waveguides allow photons to be routed between cavities. In general, because the refractive index of diamond ($n = 2.4$) is very different from more conventional optical media (e.g. silica $n = 1.5$, and Si $n \approx 3.4$ in the near-infrared) monolithic structures are preferable to heterostructures.

For monolithic devices, the inertness and robustness of diamond become liabilities. However, a fabrication route is provided by another remarkable property of diamond – its conversion by damage from an sp^3 -bonded form of carbon to an sp^2 -bonded form upon ion bombardment.

To create a sacrificial layer in diamond for subsequent processing, one can use ion irradiation to create a buried layer of damage. The buried layer can then be subsequently removed by chemical etching. This technique is based on the fundamental observation that there exists for diamond a critical ion-induced damage threshold, D_c , above which diamond will graphitize upon annealing. In regions in which the damage level is below D_c , the damaged diamond is annealed back to diamond. This provides an efficient method for the creation of sacrificial layers for more complicated free-standing structures⁵⁹.

By combining the implantation process over small regions of the diamond sample, and using focused ion beam (FIB) milling, it is possible to write optical structures on thin layers of diamond, connected to the bulk at predetermined points^{60,61}. Using this method, free-standing waveguides (Fig. 4) and cantilever structures have been made on 3.5 μm thick layers of single-crystal diamond.

The method of single energy irradiation is limited, however, in that the swelling produced via the ion implantation requires pressure to prevent cracking, and so layers thinner than around 1 μm are problematic. There is an alternative, which is to use two irradiations of different energies. The idea is to create *two* graphitic layers, which have a thin diamond membrane 'sandwiched' between them. For example, by implanting with 2 and 1.88 MeV He ions, a diamond membrane approximately 200 nm thick, 3.3 μm below the surface has been achieved⁶². At such depth, the pressures due to the diamond cap layer approach 10 GPa⁶³, which leads to a D_c higher than that near the surface. Because the desired layer is buried, one can anneal samples to higher temperatures without being too concerned about surface damage and graphitization. The cap layer is removed by welding a

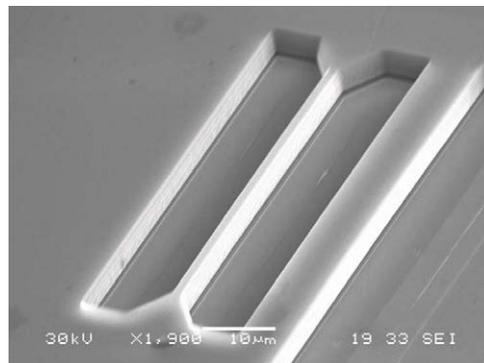


Fig. 4 Scanning electron microscope image of a diamond waveguide prepared from a 2 MeV He ion implant. The layer produced is 3.5 μm deep and the distance from mirror to mirror is $\sim 75 \mu\text{m}$. This waveguide was multimoded for visible light and was a significant step towards single-mode waveguides (Image courtesy of P. Olivero.)

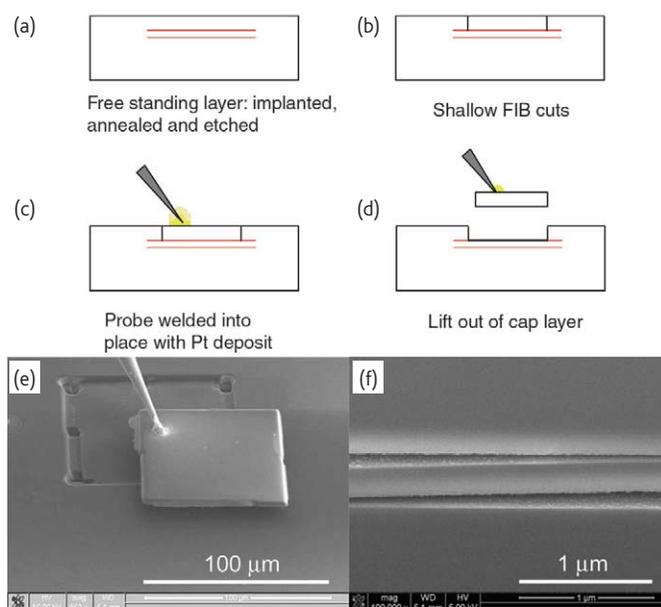


Fig. 5 Schematic representation of the fabrication steps required to expose an ultrathin layer of diamond. (a) Sample is implanted with He ions, and annealed. Etch holes are milled to allow etching to leave two air gaps at depth within the sample. (b) Shallow FIB cuts to partially free the cap layer. (c) Positioning of microprobe and welding to attach the probe to the layer. (d) The final cut on the cap is made and it can be lifted away from the sample. Scanning electron microscope images: (e) liftout of the cap layer, exposing a 330 nm layer for machining; (f) a 250 nm diamond layer in cross-section.

micromanipulator tip to the layer and post-processing continues as before. A schematic of the processing sequence is shown in Fig. 5, along with scanning electron microscopy images of the cap layer being removed and a test structure fabricated in the material.

Other methods for sculpting diamond, such as reactive ion etching (RIE), have also been used to fabricate microring resonators⁶⁴, photonic bandgap structures⁶⁵, and waveguides⁶⁶. One of the aims of all these processes is to create integrated quantum optical circuits – a diamond nano-optical workbench. These techniques are still in their infancy, and considerable effort is required to ensure that the exacting levels of perfection required for quantum information processing application are achieved.

The design of optical structures in diamond also faces fundamental challenges. These challenges arise from the fact that the refractive index of diamond is quite different from more traditional optical media (e.g. Si and silica), and also that the main operating wavelength of 637 nm is different from the standard telecoms bands. Despite these differences, rapid progress has been made. Tomljenovic-Hanic *et al.*⁶⁷ showed that high-Q photonic bandgap cavities ($Q \sim 10^4$) were possible in diamond, and subsequent analyses have extended these results⁶⁸. There is much scope to improve these quality factors, using results from other media with similar refractive indices⁶⁹, indicating that as diamond fabrication improves, there are designs ready to take advantage of these improvements.

Quantum information processing in diamond

Quantum information science is concerned with employing the full powers of quantum mechanics, especially quantum entanglement, to computational tasks. Combining information science and quantum mechanics has already proven extremely fruitful, and has resulted in the discovery of cryptographic protocols impossible classically, and algorithms that scale more favorably than their classical counterparts⁷⁰. Diamond is important to quantum information because it sits at the interface between solid-state and optical implementations of quantum computing. As indicated above, the color centers of diamond are the only solid-state photostable room-temperature single photon emitters known, and no other solid-state system has demonstrated individual qubit coherence times as long as NV⁻ centers (although there are longer coherence times measured in ensemble systems, especially P in Si at cryogenic temperatures⁷¹). To exploit the long coherence times, and obtain long-range qubit–qubit interactions, requires *controlled* fabrication of centers and optical structures to guide the light, precisely the nanofabrication toolkit described above. In this section we describe some of the ideas for engineering the quantum properties that will result from devices so created.

The potential for a diamond-based quantum computer was recognized very early with two proposals based on ensembles of NV⁻ centers coupled via switched resonant optical dipole–dipole coupling^{72,73}; see also reviews^{31,74}. This coupling mechanism also has proven popular in a number of other proposals, and has been observed in hole-burning studies of ensembles⁷⁵ (albeit without control of the number of centers coupled). However, there have been no demonstrations of this coupling in spectrally resolved systems of two NV⁻ centers, and spectrally selective coupling mechanisms^{72,73} would not appear to meet the requirements of a scalable architecture.

The coupling of NV⁻ centers to nearby spins has shown dramatic results and is a proven testbed for exploring few-qubit protocols. One of the most important features of such coupled systems is that the NV⁻ provides a clear readout for otherwise hidden nuclear and electronic spin dynamics, so called dark spins. Spin–spin dynamics observed include those between NV⁻ electronic spins and ¹³C nuclei^{25–28}, and NV⁻ and proximal N electronic spins^{24,76}. Such protocols are clearly not scalable, however, because the inter-qubit spacing is very small, of the order a few nanometers. However, such features could be useful in larger scalable quantum computers as small quantum registers²⁷, coupled via long-range photonic interactions mediated by the color centers. More sophisticated models for quantum information processing in diamond fall broadly into two styles: those where the NV⁻ are the qubits with optical coupling between qubits; and those where photons act as the qubits and the NV⁻ serve as a resource to entangle the qubits. We explain each of these styles in turn.

The NV⁻ center is an obvious candidate for a matter-based quantum architecture. Proven single-qubit operations and readout^{21,77,78}, long coherence times²⁴, and Stark shift control

centers^{16,17}, coupled with transform-limited single-photon generation⁷⁹ for flying qubit generation and the potential for photon-mediated qubit–qubit interactions, are all extremely promising for eventual diamond-based quantum processors. There are several options available for coupling qubits in a scalable fashion, though none have yet been demonstrated experimentally. It is important to realize that although very long decoherence times for NV⁻ have been observed at room temperature, optical coupling seems to require cryogenic temperatures.

The most obvious schemes for matter-based quantum processing in diamond take advantage of cavity quantum electrodynamics (QED) approaches⁸⁰. The general idea is to build cavities (preferably monolithic diamond cavities) with high Q/V (quality factor divided by volume) containing NV⁻ centers. The strong coupling between the NV⁻ center and the cavity mode allows quantum information in the center to be transferred to a photon, which can then be classically routed to another cavity. In so doing, a remote qubit–qubit interaction is effected. As discussed above, innovative cavity designs, Q-switches, and single-photon routing are all required to enable such schemes, and work on their development is ongoing.

The repeat-until-success schemes^{81,82} and related brokering schemes⁸³ seem ideally suited to NV⁻-based quantum computing and can innovatively take advantage of the center's properties. The essential idea underpinning all of these proposals is that when two centers are made to emit a single photon, information about which center has emitted the photon can be erased by suitable design of an interferometric detector (Fig. 6). Combining these approaches with heralding allows remote centers to be probabilistically entangled via the measurement process. Heralding is where a particular process

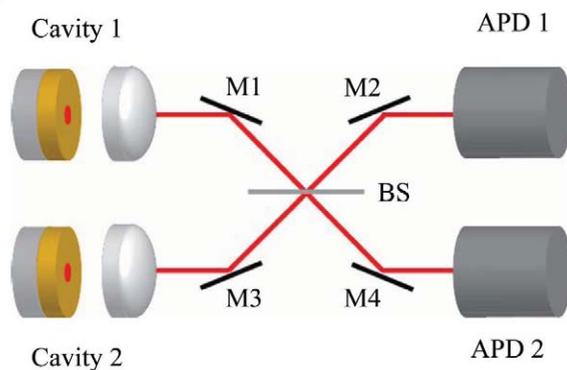


Fig. 6 Schematic depicting the experimental setup required to entangle two remote centers via measurement. In this scheme, each center (red dot) is embedded in a high-finesse optical cavity, so that the spontaneous emission from each center is predominantly into the cavity mode (strong Purcell regime). A weak excitation pulse applied to both centers will, on average, excite only one center. The interferometer formed by mirrors M1–M4 and the beamsplitter, BS, erases the which-path information, and detectors (avalanche photodiodes) APD 1 or APD 2 will collapse the centers onto one of the two Bell states of having emitted or not emitted. By using multiple ground states (accessed using radiofrequency pulses) and a heralding scheme⁸², it is possible to create remote entanglement between the electronic states of the centers, which can then be swapped to the more robust nuclear states (brokering)⁸³.

(e.g. the entangling interaction) only occurs probabilistically, but a clear signature of success is obtained. Brokering takes these ideas a step further by generating entanglement on a particular degree of freedom (e.g. the electronic spin), and when the entanglement operation succeeds, the entanglement is swapped for a more robust degree of freedom (e.g. the nuclear spin) to build up a large-scale entangled state that can be used as a resource for quantum protocols. Such schemes have also been discussed in the context of ion trap registers⁸⁴. These ideas also translate directly to operation of few-qubit registers, comprised, for example, of several proximal ¹³C nuclei²⁷. As there are few requirements in terms of the absolute positions of the optically coupled qubits, this approach is well suited to coupling fortuitous centers found by exhaustive search of the diamond sample; however, a more efficient approach would be to embed the registers in optical cavities to improve overall photon collection efficiencies and to narrow the linewidths to improve path-erasure fidelities.

The alternative to matter-based computing with mobile photons mediating long-range interactions is to place the focus on the photons as qubits, and use the NV⁻ centers to mediate interactions between them. Again, there are many different ways that such interactions can be achieved, and we can only discuss a few here. The first approach is based on weak interactions of nonlinear optics⁸⁵. In these schemes, a weak Kerr nonlinearity (intensity-dependent refractive index) is used to generate a conditional phase shift on a strong probe field. The weak Kerr nonlinearity must be generated via a lossless interaction, and electromagnetically induced transparency (EIT) appears to be the ideal process for generating this interaction⁸⁶. Importantly, the nonlinear phase shift *need not be* the full $\pi/2$ phase rotation required for a traditional entangling gate, but rather a more modest (and achievable) phase rotation of the order of 1° is acceptable when combined with a measurement of the probe field. In such a way, the Kerr medium can effect a quantum nondemolition measurement of the photons. This latter point is important as most photon measurements are performed *destructive*. The ability to measure a specific degree of freedom of a photon *without* destroying the photon constitutes a resource that can be used to generate entanglement and perform quantum computing. The strategy for a quantum computer would then be to have diamond waveguides carrying the single-photon signals, with ensembles of NV⁻ centers under conditions of EIT providing the nonlinear interactions.

Strong coupling and measurement provides another means for entangling photons, using so-called 'photonic modules'⁸⁷. The idea is that a series of initially unentangled photons pass through an atom-cavity system, which performs a quantum nondemolition measurement on the photons, similar to that achieved by the Kerr interaction. By careful preparation of the photons in superposition states and measurement of the state of the atom after the photons have passed through, an operator measurement on all the photons is effected. This is potentially very powerful and is a deterministic

process for building up multiparticle entanglement analogous to the probabilistic processes discussed above. The simple design of the chip also encourages a modular approach to design and integrated photonic chips, and it has been theoretically shown that parallel applications of the modules leads to a highly efficient ‘factory’ of two-dimensional cluster states⁸⁸, which are a universal resource for quantum computing when combined with arbitrary basis measurement. Nielsen⁸⁹ has discussed the use of cluster states in a photonic setting. The continuous generation of a two-dimensional cluster state would be extremely useful as it avoids many of the problems associated with probabilistic cluster state generation, including quantum memory, dynamic memory, and routing. The modular design allows for defect-tolerant approaches to large-scale integration to be pursued, by characterizing individual chips and discarding nonfunctioning elements, and incorporating working elements into the larger architecture.

Feynman’s⁹⁰ original vision of a quantum computer was for a device that simulated the precise mechanics of a given system of interest, but where the state of the computer was supposedly easier to read out and control than the unknown system. This is direct simulation by regime replication. The conventional circuit model for quantum computing has arguably moved far from this vision, but a startling new paradigm was recently discovered. In 2006, three groups independently showed that a system of coupled cavities (e.g. photonic bandgap cavities), each containing a single two- or four-state system, could be used to realize a photonic analog of the Bose–Hubbard system^{91–93}. The fundamental structures needed to build such a simulator are a source of two-level atoms, and an array of closely spaced resonant cavities (Fig. 7). Diamond seems the ideal medium in which to realize this, given the availability of large dipole moment optical centers, and the fabrication toolkit outlined above.

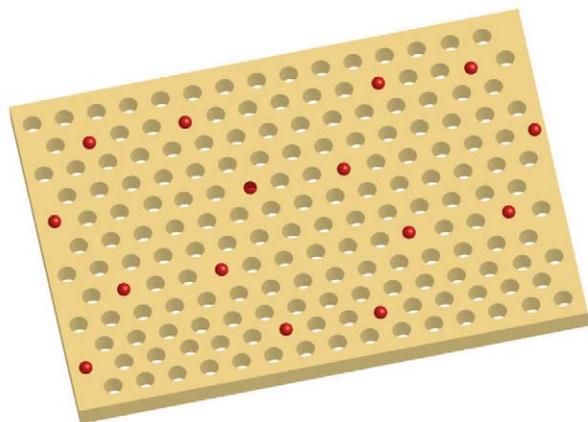


Fig. 7 Two-dimensional array of coupled photonic bandgap cavities, each containing a single two-level atom (red dots). Photons hop from cavity to cavity via evanescent tunneling, and an effective photon–photon interaction is generated by the resonant interactions with the atoms. Such a system is a photonic analog of the well-known Bose–Hubbard model and its realization will form a bridge between quantum optics and solid-state physics. (Adapted from⁹².)

The Hubbard model is a solid-state model that describes superconductivity and the metal–insulator quantum phase transition. The importance of demonstrating a photonic version, the Jaynes–Cummings–Hubbard model⁹⁴, is that it should be possible to build the structure of the Hamiltonian into the structure of the lattice of cavities and the local Hamiltonians of each atomic system. As we have described in this article, progress towards realizing this model in diamond is underway. Readout of individual elements is straightforward as each cavity, which will be separated on typical photonic length-scales (a few microns), can be individually interrogated. The ultimate challenge for such a device is not, of course, the demonstration of well-known models, but rather to map the physically accessible Hamiltonians to more complicated models that are not efficiently soluble. It is hoped that the ability to reconstruct any Hamiltonian will shed new light on the structure of chemicals and surfaces. The ideal solid-state platform for such a device is diamond because of the color centers. Regardless of the final designs for such coupled cavity systems, by uncovering this new connection between solid-state physics and quantum optics, new perspectives in both fields are being unearthed.

Conclusions and future outlook

Diamond’s long-standing appeal as a gemstone is being transformed by the understanding that diamond holds the key to unlocking practical devices exploiting quantum coherence. This strong statement is backed up by the proven room-temperature properties of color centers in diamond, namely single-photon generation, single-qubit operations at the level required for quantum error correction, and multi-qubit coupling. Coherent optical manipulations and coherent single-photon generation at cryogenic temperatures complete the picture. The importance of coherently generating photons is that there are many applications that can take advantage of nonclassical light before the information-processing advantages of quantum computers take hold.

The dramatic demonstrations of quantum coherence in diamond, however, mask the fact that despite the improvements in synthetic diamond growth, further improvements are required before large-scale diamond quantum information processors are possible. Such challenges are now in the realms of possibility for materials science and engineering, and every new advance in the development of diamond materials will be exploited by the quantum information community to realize ever more sophisticated devices. The quantum technologies which would be made available promise such large commercial payoffs that the cost–benefit ratio for developing diamond looks very good indeed. Such commercial cost–benefits are in addition to the tremendous scientific returns that we have already seen from studying diamond. Exploiting quantum coherence in diamond is the beginning of a journey into a new world, the world of massively entangled quantum systems: only now is it possible to begin to comprehend the scientific benefits of this journey. 

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