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Diamond photonics

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Diamond, a material marvelled for its strength, beauty and perfection, was first used to polish stone axes in Neolithic times. This most ancient of materials is now being touted by many as the ideal platform for quantum-age technologies. In this Review, we describe how the properties of diamond match the requirements of the 'second quantum revolution'. We also discuss recent progress in the development of diamond — and particularly diamond colour centres — for transforming quantum information science into practical quantum information technology.

Silicon and silica are the two most important materials for modern information and communication technology. Silicon chips power our computers, silica fibres transmit signals and silicon photonics provides integrated optical solutions. It may seem strange, therefore, to discuss a radical departure from silicon-based platforms that is potentially capable of surpassing silicon and silica in a new generation of photonic devices. The requirement for a new platform stems from difficulties in translating silicon to the quantum regime, where the properties of colour centres (optically active defects) in diamond are uniquely suited.

Diamond, in principle, is a material *par excellence*. It has the largest optical bandgap of any material, which provides it with many potential electronic, optical and thermal applications[1]. Furthermore, diamond is biocompatible and chemically robust. Although recent advances in the perfection of diamonds have been dramatic, arguably of more interest is the precise control and application of diamond's colour centres, which can be harnessed for quantum, sensing and labelling applications. Diamond has a unique combination of properties that is not found in any other material. New nanofabrication techniques and the availability of high-quality synthetic diamonds have together led to an explosion of interest in the application of diamond to important twenty-first century technological problems.

This Review discusses some of the emerging photonic applications of diamond and the material properties that underpin them. Such applications include classical and quantum applications, and particularly those enabled by optically active colour centres. Some of the most interesting properties of these colour centres derive from their spin properties — in particular those of negatively charged nitrogen–vacancy (NV) colour centres — but for this we refer the reader to the review of Awschalom *et al.*[2] Here we cover the less-reviewed areas of diamond in the fields of biomarking and nanoscopy, methods for enhancing the emission of colour centres using external structures, and the push towards the strong coupling regime of cavity quantum electrodynamics (QED).

The development of quantum information and communication technologies represents a grand scientific challenge. By designing information-processing platforms that exploit all the laws of physics — particularly quantum entanglement — it will be possible to efficiently solve ever more complex quantum mechanical problems, such as those related to the chemistry of proteins and other biomolecules or the physics of quantum phase transitions. Furthermore, quantum mechanics gives rise to effects that cannot be explained through classical treatments, and this provides enhanced efficiency for certain techniques (such as factoring) or protocols that are not possible classically (such as quantum key distribution).

The intense race for a platform based on quantum information and communication technologies has given rise to a plethora of candidate platforms and architectures. Among these, diamond has emerged not only as a room-temperature solid-state platform for applications such as quantum key distribution and quantum magnetometry, but also as a promising low-temperature platform for large-scale quantum computing. This promise stems from diamond's range of bright, high-dipole moment colour centres that exhibit emission from the visible to the near-infrared. One of these — the negatively charged NV centre — exhibits room-temperature spin polarization and single-spin read-out, and can be used to 'cool' nearby spins[3,4].

The biocompatibility and colour centre photostability of diamond also make it an important platform for biological applications, where it can be utilized in sub-diffraction imaging techniques. Material developments, including access to high-quality, low-birefringence diamond, has increased interest in diamond Raman lasers, which have wide applications across an unprecedented spectral range.

In this Review, we will first discuss the optical properties of diamond that enable many of its photonic applications. Then we will discuss techniques for fabricating both nanocrystal and large single-crystal diamonds, including post-fabrication micro- and nanosculpting. Finally we will cover classical applications such as biomarking and nanoscopy, before discussing the coupling of colour centres to resonant structures such as waveguides and cavities.

Optical properties of diamond

Utilizing the quantum states of light is one of the fundamental goals of photonics, but this imposes stringent requirements. In particular, ultralow-loss optical fibres and compact atom–cavity systems are required, together with few-quantum-bit quantum processors for operations such as quantum repeating[5]. Identifying appropriate platforms for such tasks is not a trivial matter.

Diamond exhibits an enviable list of properties for quantum applications. It is a wideband semiconductor (5.5 eV at room temperature) with a moderate refractive index of 2.4, and is optically transparent from the deep-ultraviolet to the infrared. Moreover, diamond is an excellent thermal conductor (2×10^3 W m^{-1} K^{-1}) that exhibits low thermal expansion, high breakdown voltage and high carrier mobility. With proper engineering and optimization, diamond will offer a promising platform for a new generation of technological devices.

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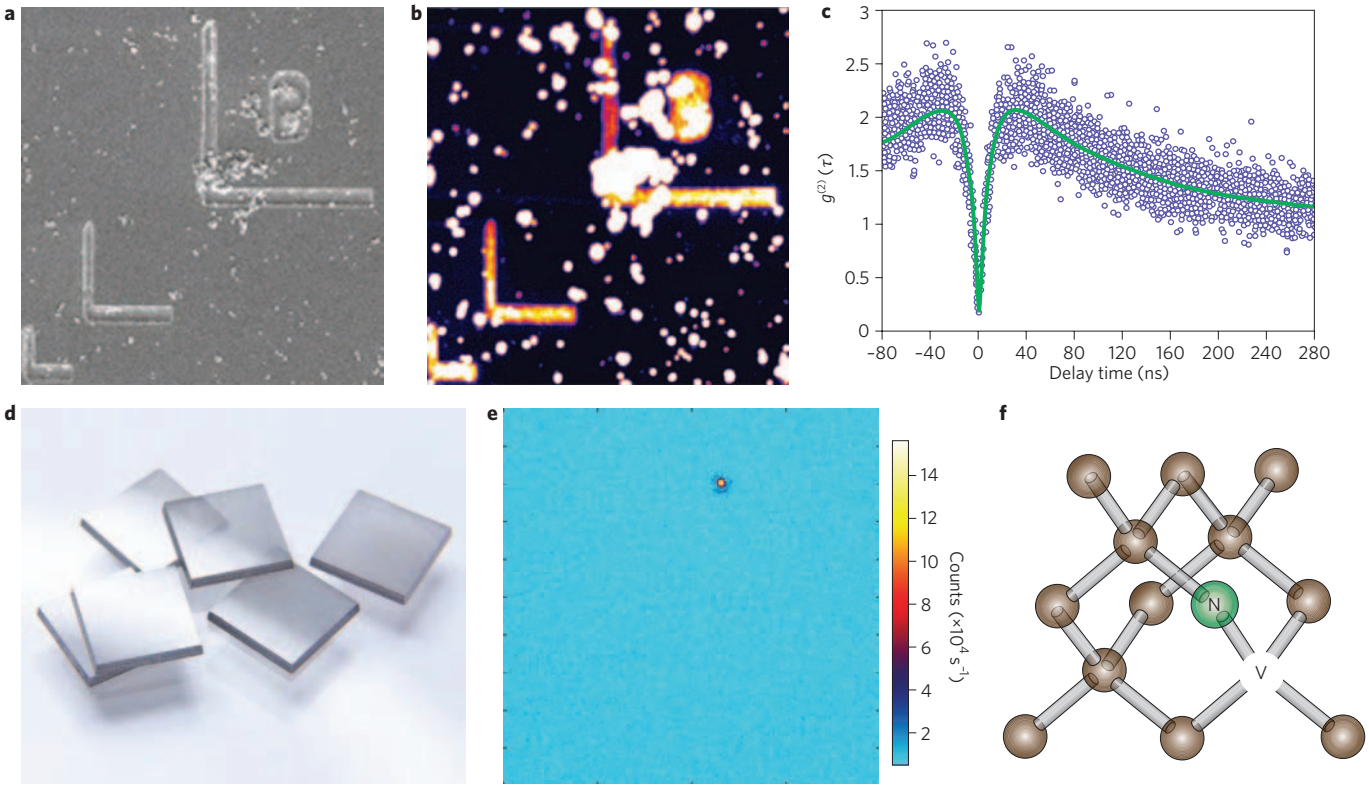

Figure 1 | NV centres in nanodiamond and single-crystal diamond. a,**b**, Scanning electron microscopy image (**a**) and corresponding confocal map (**b**) of deposited nanodiamonds. Some of the bright spots are single NV centres with count rates approaching 2.5×10^5 s^{-1}. **c**, Second-order correlation function demonstrating single-photon emission from an individual nanocrystal (marked by the red square in **b**). **d**, Commercially available single-crystal diamond CVD plates (image courtesy of Element Six). **e**, A 20 μm × 20 μm confocal map of such a crystal, showing an isolated NV centre. **f**, Crystallographic structure of the NV centre. Figure reproduced with permission from: **a**–**c**, ref. 38, © 2009 OSA; **d**, ref. 39, © 2008 Elsevier; **e**, ref. 40, © 2009 NPG.

Diamond, although indeed intriguing as a platform for photonics, has one feature that makes it particularly compelling for use in quantum devices: it hosts over 500 colour centres[6]. Owing to the large bandgap of diamond, emission from such colour centres can escape the crystal without absorption. Moreover, as will be discussed below, some of these centres function as bright single-photon sources (SPSs) at room temperature, thus providing atom-like systems in a solid-state matrix. The ability to exploit the properties of colour centres in a diamond photonic platform has caused an explosion of interest in what is now known as 'quantum diamond'.

So far, only a few colour centres in diamond have been identified as bright SPSs. Following the first demonstration of single-photon extraction from an NV centre[7], at least four new centres showing non-classical emission have been identified. The NV complex is the most studied of all colour centres, and it has already been exploited as a source of single photons in practical quantum key distribution systems[8,9]. NV centres exist in one of two charge states: NV^- and NV^0, which have zero-phonon lines of 637 nm and 575 nm, respectively. Other diamond-based SPSs include the silicon–vacancy (SiV) centre[10,11] (738 nm), a nickel nitrogen complex known as NE8[12–14] (793–802 nm), chromium-related emitters[15,16] (750–770 nm) and a carbon-related TR12 centre[17] (470 nm), among others[18–20]. It seems highly unlikely that these five centres are the totality of bright SPSs in diamond.

Most SPSs in diamond, including the SiV, NE8 and NV centres mentioned above, possess a three-level emission scheme. Thus, in addition to the desired optical transition, these centres have a parallel decay channel that may be either radiative or non-radiative. Most diamond SPSs identified so far have a transition to a metastable state, which is characterized by a long emission time, thereby reducing single-photon emission and altering the emission statistics. Some recently discovered chromium centres exhibit a pure two-level system[15,16,21]. This family of centres therefore achieves high emission rates, which will be advantageous for future metrological applications.

Diamond materials

Diamond for photonic applications can be broadly categorized into three groups: individual nano- and microcrystalline diamond, ultrananno- and polycrystalline diamond films, and single-crystal diamond plates. Each has its own advantages for photonic applications, although the properties of the hosted colour centres differ between platforms, particularly with the varying impurity concentrations within a given platform. In particular, the role of the crystal surface seems to be critical in determining the properties of many of these colour centres.

The choice for a specific type of diamond is often inherently determined by the application itself. For emerging applications such as biomarking, diamond nanocrystals such as single-digit nanodiamonds[22–25] (nanodiamonds with a crystal size below 10 nm) are the leading candidates. On the other hand, large single crystals of diamond are preferred for applications that require purity and spectral stability. There has also been success in the development of hybrid systems, which involve positioning pre-characterized nanodiamonds so as to interact with photonic structures formed in more conventional photonic media[26–31]. In this section, we first discuss nano- and microcrystalline diamond before covering large single-crystal diamond wafers.

Nano- and microcrystalline diamond

There are several existing methods for fabricating nanodiamonds, and the most efficient among these is the detonation process. In this technique, the explosive acts as the carbon source, and the required high

temperature and pressure result from the explosion[32,33]. Although a thorough purification process is required to clean the diamond particles from the soot, this technique allows for the mass-production of sub-10-nm nanodiamonds.

Particles of detonated diamond are commonly used as seeds for further nanodiamond growth through chemical vapour deposition (CVD)[33,34]. Figure 1a shows a scanning electron microscope of nanodiamonds grown using microwave-assisted CVD[35] from 4–6 nm diamond seeds. Figure 1b shows a room-temperature confocal map recorded from the same diamond nanocrystals. Bright spots show the emission from NV centres embedded in the nanocrystals. Some of the crystals host only a single NV centre, as can be seen from photon correlation measurements (Fig. 1c).

Nanodiamonds can alternatively be produced through the crushing or milling of synthetic or natural diamonds[36]. However, this method not only results in non-uniform crystal sizes and thus requires further steps to reach the nanoscale, but also has a significantly lower yield than that of the detonation process. Nevertheless, working with synthetic crystals permits the controlled introduction of optically active impurities such as nitrogen, which can then be incorporated into the nanodiamonds.

Diamond wafers

Monolithic single-crystal diamonds (typical size of 3 mm × 3 mm × 0.3 mm) are now grown by a number of commercial companies. Bulk crystals can be grown either through the high-pressure, high-temperature (HPHT) technique or by high-plasma-density CVD. The HPHT process is complicated, requires high-maintenance equipment and provides minimal flexibility for controlling dopants, thus making CVD the more common method for growing high-purity diamond crystals. CVD can also produce high-quality single crystals through homoepitaxial growth[1,37]. At the pressures used for the CVD growth technique, graphite — not diamond — is the stable phase of carbon. Growth is therefore driven by kinetics, not equilibrium thermodynamics. The main advantages of the CVD process over the HPHT technique is its simplicity, flexibility and control over the incorporation of impurities.

The extreme control provided by CVD has also led to the production of pure diamond with nitrogen and boron impurities at the parts-per-billion level. It is also possible to produce isotopically pure diamond in which the ^{13}C concentration is reduced from the native level of 1.2% to less than 0.3%. Furthermore, diamond plates have uniform and reproducible properties, which have allowed diamond to be used as an engineering material for the first time. For quantum applications, advances such as isotopic enrichment and low background impurity levels have led to reduced electron spin decoherence rates and narrower photoluminescence lines. The colour centre properties of single-crystal diamond can therefore be superior to comparable emitters in other forms of diamond (such as films or individual nanodiamonds). Figure 1d shows an optical image of typical high-purity CVD plates. Figure 1e shows a 20 μm^2 confocal map of such a plate with an isolated single NV centre, and Fig. 1f shows the crystallographic structure of the NV centre.

Another important development is the ability to grow low-birefringence diamonds, which has led to renewed interest in diamond lasers (discussed below). Birefringence arises because of dislocations and strain during the crystal growth process[37]. Typically, HPHT treatment reduces the amount of dislocations and aids the production of low-birefringence material[37].

Nanofabrication of single-crystal diamond structures

Readily available diamond wafers with reproducible properties open the possibility to fabricate practical monolithic diamond photonic structures. However, diamond is chemically inert, which makes accurate etching a significant challenge. Although a complete toolkit similar to that available for semiconductors such as silicon has not yet been developed, significant progress in the nanomachining of diamond has been demonstrated, including as ring-resonator structures (Fig. 2a)[41], waveguides (Fig. 2b)[42] and nanopillars (Fig. 2c,d)[43].

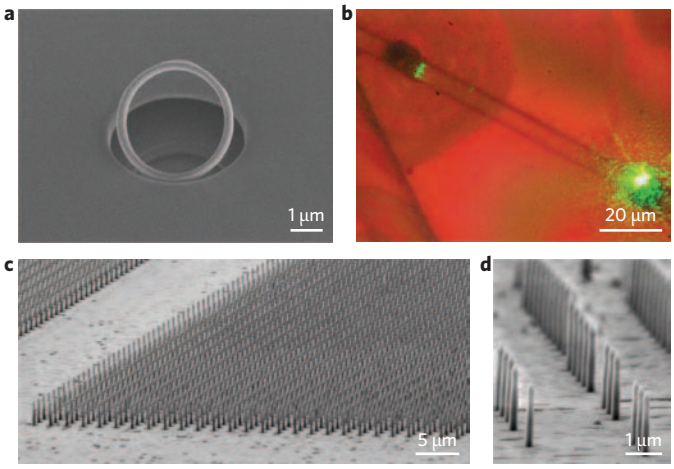

Figure 2 | Single-crystal diamond photonic structures. a, A diamond ring as a prototype for a whispering-gallery mode resonator. **b**, Diamond waveguide guiding green laser light. **c**, Diamond nanowires fabricated from single-crystal diamond. **d**, High-magnification image of three nanowires. Figure reproduced with permission from: **a**, ref. 41, © 2008 Wiley; **b**, ref. 42, © 2008 OSA; **c**,**d**, ref. 43, © 2010 NPG.

The principal step for many integrated optical or electronic circuits is to create a sacrificial layer. Although this is difficult to achieve in diamond, there is one approach that seems to work quite well. Subsurface sacrificial layers can be made using a modification of the 'lift-off' technique[44] commonly employed in the formation of synthetic diamonds from seed crystals. Ion implantation is then used to create a buried damage layer (or layers); when annealed this layer changes into a graphitic material that is susceptible to chemical etching[41,45]. This method can be used to make thin membranes of single-crystal diamond, which can be further processed to create structures[46] and then removed from the substrate for subsequent device integration.

Generating in-plane structures requires different methodologies from the high-energy implantation technique discussed above. Laser ablation can be employed to cut large areas[47], but this is usually too coarse for most optical structures. A useful method for creating single devices is focused ion-beam milling[41]. The simultaneous fabrication of multiple devices can be achieved using electron-beam-based patterning and reactive ion etching. Figure 2b shows a diamond waveguide etched from single-crystal diamond. A more sophisticated example is an array of diamond nanowires[43] etched from monolithic single-crystal diamond (Fig. 2c). The original diamond substrate shown here included nitrogen, and thus single NV centres are natively present in the diamond nanowires. Researchers achieved an order-of-magnitude enhancement in the fluorescence from certain NVs embedded in the nanowires, which clearly demonstrates the potential of nanostructured diamond devices. Furthermore, the zero-phonon line of such NV centres was recently coupled to modes of a diamond microring cavity etched directly from single-crystal diamond[48]. Quality factors of ~4,000 were measured from this device, which marks a great milestone towards integrated diamond photonics.

Applications

In this section we highlight several of diamond's applications. We also discuss the main advantages and challenges of each material as a diamond platform for classical and quantum applications.

Classical applications. Diamond has a number of niche applications that employ its unique optical, electrical and thermal properties.

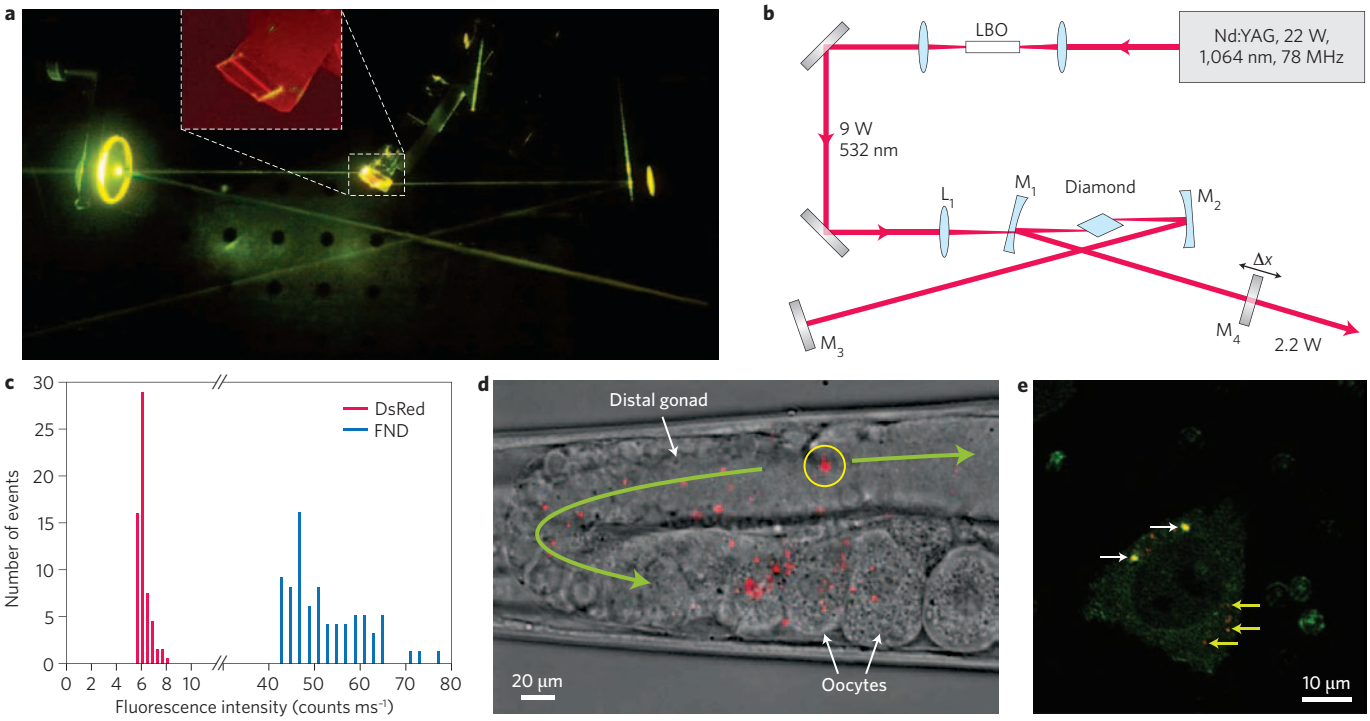

Figure 3 | Classical applications of diamond. a, Synchronously pumped diamond Raman laser operating at 573 nm, its first Stokes wavelength (image courtesy of Eduardo Granados). **b**, Schematic of the diamond Raman laser in ref. 61. The cavity comprised two curved mirrors (M1 and M2) and two plane mirrors (M3 and M4). The Nd:YAG laser was frequency doubled to 532 nm using a lithium triborate crystal (LBO). Mirrors M1, M2 and M3 were highly reflecting at the first Stokes wavelength of 573 nm, and the output coupler M4 had a transmission of 12%. **c**, Histogram showing the relative brightness of 19 nm fluorescent nanodiamonds (FNDs) compared with the chromophore DsRed. **d**, Nanodiamonds as fluorescent biomarkers. Epifluorescence/differential interference contrast merged image of an injected worm. Green arrows indicate bulk streaming of FNDs with cytoplasmic materials and the yellow circle indicates the site of injection. **e**, Overlay image of fluorescent nanodiamonds internalized in HeLa cells in a combined contrast of a backscattered excitation laser light (488 nm) and emission from an NV colour centre. White arrows identify nanoparticles that are observed in both backscattered and fluorescence modes, whereas yellow arrows show nanodiamonds that are detected only in the photoluminescence mode. Figure reproduced with permission from: **b**, ref. 61, © 2010 OSA; **c**, ref. 72, © 2009 Wiley; **d**, ref. 62, © 2010 ACS; **e**, ref. 66, © 2009 ACS.

For example, diamond windows were used on some of the Russian Venera probes to protect cameras from the corrosive, high-pressure environment on Venus, and diamond windows are often used for applications in which chemical resistance, robustness to large pressure gradients or low thermal expansion are required. A related development is the fabrication of diamond microlenses for optoelectronic devices[49,50]. Diamond also shows promise for high-energy particle detection[51,52], heat spreaders for thermal managment[53], photodetectors[54], ultraviolet LEDs[55] and near-infrared LEDs[56].

Diamond Raman lasers have emerged as an important platform for generating intense yellow light (573 nm)[57,58]. One of the main applications for such radiation is in the treatment of skin diseases, as this wavelength corresponds to the peak absorption of oxyhaemoglobin. Diamond's high thermal conductivity makes it ideal for high-power applications, and its high Raman gain coefficient means that only small crystals are required. Although the principal target wavelength is yellow, the wide optical transparency of diamond allows for tunable Raman sources across multiple spectral ranges[59–61]. In addition to their yellow laser, the Macquarie group also recently demonstrated a deep-ultraviolet laser. Figure 3 shows photographs of their single-crystal diamond Raman yellow laser and a schematic of the experimental set-up.

Most classical applications that employ single crystals of diamond do not require complex fabrication techniques, and commercially available HPHT diamond plates are usually sufficient. However, there is nonetheless a demand for the fabrication of large-size, low-cost diamond wafers approaching the size of commercial silicon wafers. Meeting this demand would allow all-diamond devices such as radiation detectors and high-power lasers to make the transition from niche devices to mainstream products. It is not clear at present if this goal can be reached simply through a linear extrapolation of existing processes, or whether new breakthroughs in diamond fabrication will be required.

Biomarking. Fluorescent biomarkers have opened a new window into the study of cellular processes. Fluorescent diamond nanocrystals offer new advantages over more conventional biomarkers (such as green fluorescent protein) owing to their brightness and photostability, and compare favourably with quantum dots because of their biocompatibility[24,62–67]. Nanoparticles can be functionalized with various biomolecules[68] such as peptides, proteins and DNA[69–71].

So far, the smallest nanodiamonds bright enough for biomarking applications are 19 nm in diameter[72]. These were mass-produced by the ion irradiation of non-luminescent nanodiamonds[64]. Figure 3c shows that the fluorescence from these nanodiamonds is an order of magnitude brighter than that of a traditional red chromophore. Figure 3d,e shows other examples of biomarking using nanodiamond crystals. However, recent studies have involved smaller nanodiamonds because these are more likely to pass freely through membranes. In particular, there is a push to develop stable, bright, single-digit nanodiamonds[25,32,36]. Recent studies have demonstrated the spin detection of a single NV embedded in a 7-nm-diameter nanodiamond crystal[22]. This is extremely significant for applications in bioimaging and nanosensing, as will be discussed in the following sections.

One of the key breakthroughs in understanding cell processes has been the real-time imaging of cell dynamics. Recent studies have involved the three-dimensional tracking of single fluorescent nanodiamonds inside cells[64], as well as preliminary experiments of imaging living cells[62], drug delivery and targeted bio-imaging[69]. The recent discovery of new ultrabright colour centres with narrow emission in the near-infrared is significant for such tracking techniques[15], as these wavelengths do not interfere with cell autofluorescence. However, no mass-production technique yet exists that can incorporate such bright emitters into nanodiamonds.

Improving the quality of nanodiamond colour centres towards those of emitters in bulk diamond requires a better understanding of the chemical and physical environments surrounding each colour centre. Fabricating subnanometre diamonds with stable luminescent defects remains a grand challenge in nanodiamond production.

The ability to functionalize nanodiamonds[68] is a key element to continued progress in bio-marking, particularly for the use of nanodiamonds in drug delivery and targeted bio-imaging. Acid treatment techniques can be used to prepare carboxylated surfaces for bioconjugation. Such nanodiamonds hosting optical defects can be used as delivery vehicles for transporting drugs, genes and vaccines into cells[69]. Functionalization with magneto markers[71] will provide new bio-imaging possibilities.

The agglomeration of nanodiamonds is a serious issue for many diamond-based quantum and biological applications. One of the main solutions at present is to use isolated nanodiamonds, although this is clearly not a satisfactory approach for scalable production processes. The de-agglomeration of nanodiamonds into their primary size is an important but complex alternative[32]. Researchers have so far investigated the mechanical de-agglomeration of nanodiamond suspensions using stirred media milling, bead-assisted sonic disintegration and the use of chemical modification and detergents[24,32,73,74], although the optimal technique is not yet known.

Nanoscopy. Nanoscopy is the emerging field of optical microscopy at the nanoscale, in particular using visible-wavelength photons and techniques to improve resolution far beyond the diffraction limit. All optical nanoscopic techniques employ some form of optical nonlinearity or spatially non-uniform field (as in the case of MRI scanning) to beat conventional imaging limitations. Diamond colour centres therefore have considerable potential in the field of nanoscopy.

The photostability of NVs enables them to be imaged using stimulated emission depletion (STED) microscopy[75,76]. This technique uses two beams: a Gaussian mode to excite the emitter and an STED beam to deplete the excited state through stimulated emission everywhere that the field is non-zero. The technique provides high spatial resolution because the only region that is undepleted — and hence fluoresces — is the central dot of the STED beam. The resolution is set by the intensity of the STED beam; the more intense the beam, the smaller the radius of the central undepleted region and hence the more well-defined the location of the emitter. The source under investigation must be photostable because the STED beam is extremely intense, thus making diamond-based SPSs the ultimate choice for this technique.

Figure 4a shows a confocal map recorded from an ensemble of conventionally unresolvable NVs. However, once the STED beam is applied, the emitters can be optically resolved and, with the assistance of fitting parameters, their location can be determined with subnanometre resolution[75]. Using STED, three-dimensional imaging from densely packed single emitters with extreme sub-diffraction resolution is feasible. Applying an axial beam pattern allows the super-resolution imaging of NV centres 6 μm deep inside the diamond crystal (Fig. 4b).

Magnetometry is another fascinating application for which diamond centres are considered to be prime candidates. Employing NV centres, revolutionary demonstrations of sensing magnetic fields at the nanoscale, at room temperature and under ambient conditions have been performed[77–79]. Such groundbreaking results may eventually allow single spins in biological molecules to be measured, which cannot be achieved through traditional electron spin resonance techniques. The use of diamond centres in magnetometry is based on the ability to measure the electron spin of an NV centre by exciting it with a green laser and switching the microwave frequency in close proximity to the centre. A simple NV magnetometer can be constructed by attaching an NV nanodiamond to a magnetic tip and scanning it over a specific substrate[78]. The NV centre can alternatively be embedded in ultrapure single-crystal diamond, where it is used to sense externally applied magnetic fields[77]. Researchers have proposed the sensing of fluctuating magnetic fields by probing the dephasing rate of the NV spin quantum bit placed in such an environment, which should ultimately allow the detection of ion channels[80].

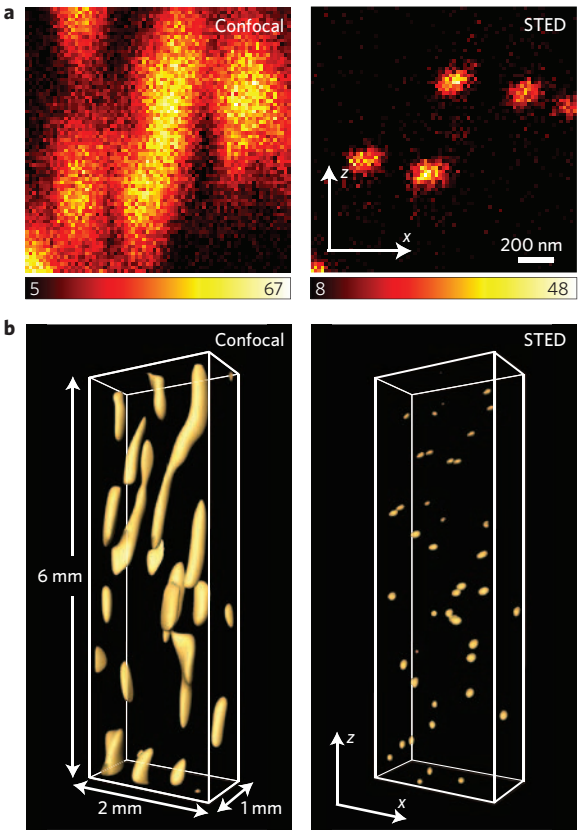

Figure 4 | 3D STED images of NV centres in bulk diamond. a, Individual colour centres are sharply resolved in the STED image (right) and blurred in the conventional confocal microscopy (left). **b**, 3D surface-rendered representation of NV centres employing confocal (left) and STED (right) microscopy. Figure reproduced with permission from ref. 76, © 2009 ACS.

Quantum applications. Quantum optical applications are the core of diamond photonics. Although remarkable progress has been achieved, there are still significant hurdles to overcome before diamond photonic devices can begin to constitute a practical quantum technology. The main issue is to optimize the emission from high-quality centres into defined radiative modes. This inexorably leads to the requirement that diamond emitters are coupled to optical structures, especially cavities[81,82] and waveguides, in a fashion that either preserves or improves their optical properties[83].

The Purcell factor, given as

$$F = \frac{3}{4\pi^2}\left(\frac{\lambda}{n}\right)^3\left(\frac{Q}{V}\right) \quad (1)$$

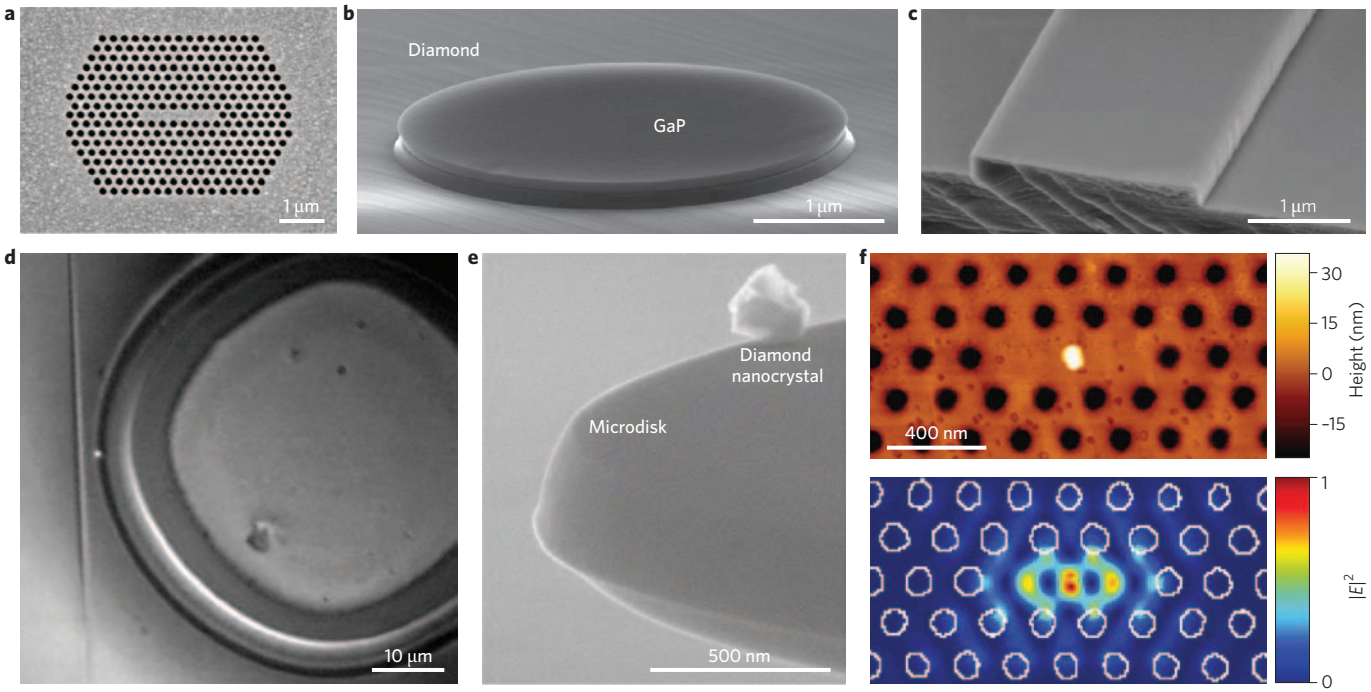

Figure 5 | Diamond nanophotonic components. a, 2D photonic crystal cavity made from a nanodiamond film. **b**,**c**, GaP microdisk (**b**) and GaP waveguide (**c**) deposited on single-crystal diamond. **d**, Manipulating nanodiamond hosting a single emitter onto a toroidal microresonator using a fibre taper. **e**, Nanodiamond hosting a single emitter on a silica microdisk. **f**, Atomic force microscopy image of a diamond nanocrystal placed in a GaP photonic crystal cavity (top) and the simulated electric field profile of the cavity's fundamental mode (bottom). Figure reproduced with permission from: **a**, ref. 85, © 2007 AIP; **b**, ref. 90, © 2009 AIP; **c**, ref. 89, © 2008 AIP; **d**, ref. 27, © 2009 AIP; **e**, ref. 31, © 2009 OSA; **f**, ref. 29, © 2010 AIP.

is used to describe quantitatively the enhancement to the spontaneous emission rate of a coupled emitter–cavity system, where λ is the emission wavelength, n is the refractive index, Q is the quality factor and V is the cavity mode volume. The expression Q/V is an important figure of merit for cavity design that clearly emphasizes the need for high Q and low V. Enhancing the spontaneous emission through the Purcell effect is the first step towards the realization of quantum systems because it can assist with directional emission and the generation of indistinguishable photons.

Many of the most powerful protocols for quantum information and communication technologies with diamond employ the strong coupling limit of cavity QED, which achieve a reversible interaction between the emitter and the cavity[81,82]. Such coherent control allows energy to be transferred between the cavity and the emitter for a finite period of time through optical Rabi oscillations. The Rabi frequency must be fast compared with the cavity loss rate and decoherence processes (including absorption). High Q can be understood as the minimization of decoherence, whereas decreased mode volume results in increased Rabi frequency. High Q and low V are therefore essential for the strong coupling regime, and researchers have already designed all-diamond cavities that operate in this regime[84]. Cavity QED has been demonstrated with atoms, single molecules and quantum dots[81,82], but not yet with colour centres in diamond.

Developing scalable technology to sculpt diamond waveguides/cavities and integrate them with current SPSs is a long-term goal in the field of diamond photonics. Early studies explored the fabrication of photonic crystal cavities in diamond films[85,86]. However, in these cases, scattering losses, surface roughness and high impurity concentration in the films limited Q values to around a few hundred[85]. Furthermore, optical properties such as the linewidth and spectral stability of active emitters in CVD films (or nanodiamonds) are poor compared with the same emitters in bulk diamond. Quantum computation and communication applications therefore have a strong incentive to exploit colour centres in monolithic single-crystal diamonds[16,40,43,87].

There has also been significant focus on hybrid systems, in addition to monolithic diamond approaches. Such hybrids combine mature photonic platforms with bright emitters in either nanodiamond or single-crystal diamond. GaP is a useful platform for such systems because of its transparency in the visible, the range of machining techniques available and its high refractive index (~3.25), which allows for the efficient coupling of light and the use of advanced high-Q, small-volume photonic crystal cavity designs. This approach has been used to deposit GaP waveguides[88,89] and microdisks[90] directly onto single-crystal diamond. In such cases, emission from NV centres close to the diamond interface is coupled to the waveguide or the cavity modes. This design is a prototype of a possible on-chip nanophotonic device that employs diamond single emitters as its fundamental constituents.

Although emitters in single-crystal diamond possess narrower and more stable spectral lines, nanodiamonds are very convenient for hybrid applications. Researchers have placed individual nanocrystals[38,91] on top of photonic crystal cavities[29] and coupled the resulting emission to optical fibres[92], waveguides[88,89], microspheres[93], microdisks[28,31] and toroidal resonators[27]. The deterministic placement of a GaP cavity slab onto a diamond nanocrystal has also been experimentally demonstrated[30].

Figure 5 shows examples of various diamond nanophotonic components, including photonic crystal cavities, microdisks and toroidal microresonators. A GaP microdisk placed on top of single-crystal diamond can achieve Q values of around 25,000 (ref. 90), whereas other systems exhibit lower values of around 1,000. The superiority of the GaP microdisk highlights the importance of material quality when designing an appropriate cavity QED system. Diamond nanoparticles scatter from both surface and bulk defects, which reduces Q despite high-quality cavity fabrication. It is therefore clear that cavities of single-crystal diamond must be developed

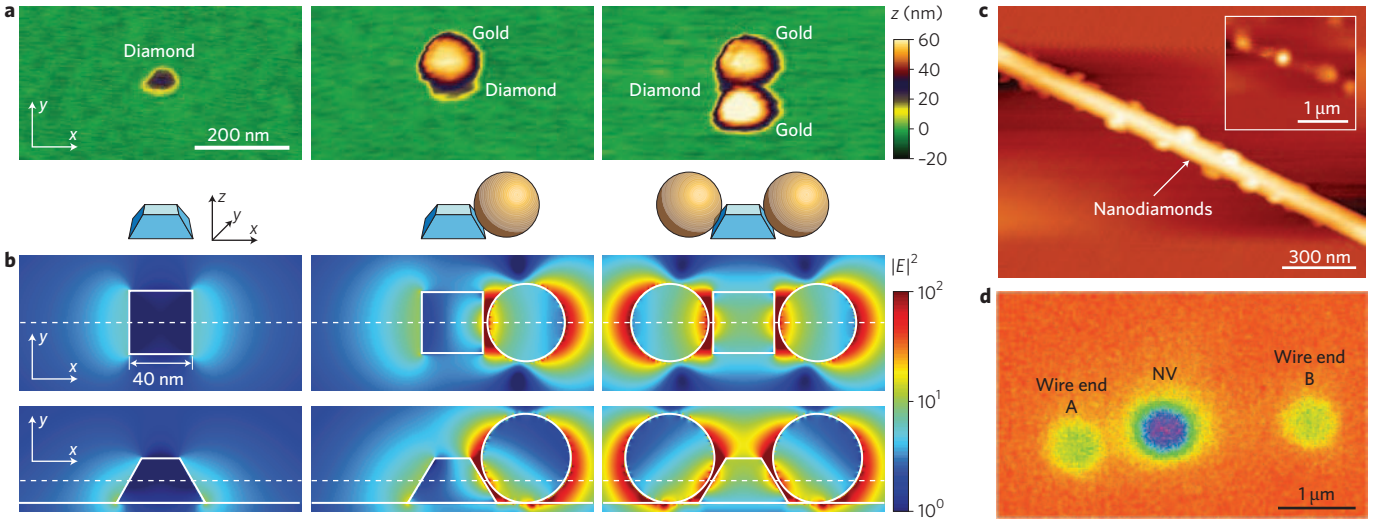

Figure 6 | Plasmonics with NV centres in diamond. a,**b**, Atomic force microscopy images (**a**) and simulated emission enhancement (**b**) of diamond nanocrystals (left), to which one (middle) or two (right) gold nanoparticles are coupled. **c**, Atomic force microscopy image of diamond nanocrystal hosting a single NV centre attached to a silver nanowire. **d**, Wide-field image of a single quantum emitter (NV centre) coupled to a silver nanowire. Both ends of the wire fluoresce, which indicates the propagation of plasmons along the wire. This set-up was used to demonstrate the wave–particle duality of plasmon polaritons. Figure reproduced with permission from: **a**,**b**, ref. 94, © 2009 ACS; **c**,**d**, ref. 95, © 2009 NPG.

before diamond can compete with current developments in other physical systems[81,82].

Researchers have also explored the resonant coupling of diamond SPSs to plasmonic waveguides. The understanding of a single emitting dipole in the proximity of a metal is important because such coupling modifies the dipole's emission lifetime and spectral distribution. For single emitters, this coupling is analogous to a cavity Purcell enhancement and can therefore not only significantly enhance the emitted count rate but also improve directionality, thus providing an enhanced SPS. Figure 6 shows two examples of how single NV centres can be coupled to plasmonic structures. In Fig. 6a,b, a single diamond nanocrystal is coupled to one or two gold nanoparticles[94]. Time-resolved measurements show a decrease in the excited state lifetime by factors of 7.5 and 9.5 for configurations i and ii, respectively[94]. The emission rate of the coupled nanodiamond was also enhanced. Figure 6c,d shows a diamond nanocrystal coupled to a silver nanowire[95], in which researchers demonstrated the wave–particle duality of surface plasmon polaritons.

The main challenge in plasmonic coupling is aligning the emitting dipole of the nanocrystal with the propagation mode of the plasmon. In the case of an NV centre, in which the emitting dipoles are perpendicular and the room-temperature emission is not polarized, this coupling results in high losses. Novel diamond emitters that exhibit linear dipole behaviour[15] are therefore more promising than NV centres for plasmonic coupling.

A key application of single photons is for quantum key distribution[96], particularly because long photon coherence and indistinguishability are not required. Quantum key distribution has been demonstrated using an NV centre as the SPS[9], but this situation is far from ideal. In free-space transmission, emission at 637 nm is highly absorbed by the atmosphere and the large emission width (~100 nm) causes significant dispersion in fibre-optic systems. Furthermore, the emission from an NV centre is not fully polarized, which further reduces efficiency. Narrowband polarized SPSs are therefore used to improve efficiency.

Challenges

One aspect of diamond fabrication that has not yet been satisfactorily addressed is the creation of an ensemble of high-spectral-quality single emitters in close proximity to the crystal surface. Such a system has its origins in the hybrid approach, in which SPS emission is coupled and transmitted through a conventional photonic medium[89,90]. Achieving this is a new optimization challenge that is different from the more conventional task of optimizing the yield in bulk diamond or nanodiamond. Additionally, the influence of the lattice environment on the optical properties of the centre and methods to controllably fabricate ensembles of centres with similar physical and optical properties are still not understood. Methods of controlling a centre's charge state are also unclear, but research in this direction is progressing[97].

Diamond machining is currently a major limitation in the development of all-diamond photonic platforms. Single-crystal diamond cannot be grown heteroepitaxially and therefore conventional sacrificial layer and etching techniques (such as those used to fabricate GaAs or Si) are not applicable. The chemical inertness of diamond is a significant hurdle for diamond nanofabrication and further research is necessary before all-diamond platforms can become viable alternatives to hybrid designs.

Fabrication yield — the number of optically active defects created for each implanted ion — is also an important figure of merit[98,99]. In the case of NV centres, excess nitrogen leads to increased decoherence through the presence of fluctuating charges as well as excess electronic and nuclear spins, so an accurate estimation of the conversion efficiency between residual/implanted nitrogen and optically active centres is required. Recent results seem to indicate that an increase in yield can be achieved by implanting nitrogen ions deep in a crystal using mega-electronvolt implantation energies[100], although the mechanisms for the increase are unclear and open to interpretation.

For hybrid schemes employing nanocrystals, the optical properties of the NV centre degrade as the crystal size is reduced. Proximity to the surface induces charge fluctuations and instabilities in the spectrum, which often causes severe blinking. The coherence time is also reduced and consequently Fourier-transform-limited emission has not yet been observed from a diamond nanocrystal. Surface chemistry seems to be a chief parameter for resolving this problem. Core–shell methods were developed to stop the blinking in quantum dots[101]. Inspired by this method, Bradac and co-workers demonstrated that the emission from NV centres is stable with no blinking when 5 nm nanodiamonds are embedded in a polyvinyl alcohol film[97]. Such approaches suggest new directions that may

further extend the use of nanodiamonds in both quantum and biological applications.

Summary and outlook

Research into diamond photonics is gaining momentum around the world. However, there are several key impediments to the technological applications of diamond colour centres, including improvements to the spectral properties of the available colour centres. In some cases, these improvements will be made by identifying and optimizing new and existing centres, and in other cases through the improved integration of colour centres with enhanced collection optics, such as solid immersion lenses[102,103] and cavities. This will open up applications for diamond in quantum information science and quantum metrology. Control of the base material and surface effects are crucial for achieving viable technological advances. The nanomachining of diamond is in its infancy, and although we have seen some dramatic demonstrations, a mature fabrication toolkit has yet to be developed. Nevertheless, the outlook is exciting. Diamond photonics is a thriving field, particularly given the recent demonstrations of ultrahigh-purity synthetic diamonds, new colour centres, new photonic designs and new applications. The advent of robust, room-temperature quantum platforms are sure to make diamond a breakthrough technology of the twenty-first century.

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