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**Review article** 

# Engineering colloidal semiconductor nanocrystals for quantum information processing

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Quantum information processing—which relies on spin defects or single-photon emission—has shown quantum advantage in proof-of-principle experiments including microscopic imaging of electromagnetic fields, strain and temperature in applications ranging from battery research to neuroscience. However, critical gaps remain on the path to wider applications, including a need for improved functionalization, deterministic placement, size homogeneity and greater programmability of multifunctional properties. Colloidal semiconductor nanocrystals can close these gaps in numerous application areas, following years of rapid advances in synthesis and functionalization. In this Review, we specifically focus on three key topics: optical interfaces to long-lived spin states, deterministic placement and delivery for sensing beyond the standard quantum limit, and extensions to multifunctional colloidal quantum circuits.

After decades of intensive theoretical and experimental efforts, the field of quantum information science and engineering is at a critical moment. Research areas within the field have expanded, including quantum information processors<sup>1,2</sup>, technologies to connect quantum processors by photons<sup>3</sup> and quantum sensing methods<sup>4</sup>. New possibilities are currently being unlocked in fields spanning defence and biomedical research. Agap remains, however, between proof-of-concept demonstrations and engineering commercially viable systems. To realize the next leap in performance, materials and device scaling challenges must be overcome<sup>5</sup>.

In this Review, we describe how leveraging the increasingly coherent and controllable atom-like properties of colloidal quantum dots (CQDs) can open disruptive opportunities in emerging quantum information processing (QIP) applications. In particular, we focus on three key topics that exploit the colloidal nature of CQDs: (1) optical interfaces to long-lived spin states, (2) deterministic placement and delivery for sensing, and (3) an emerging opportunity (from 1 and 2), multifunctional colloidal quantum circuits (MCQCs). We review advances that position CQDs closer to achieving the requisite properties for some initial QIP applications. Furthermore, we discuss design opportunities (such as bottom-up assembly methods and emerging synthesis techniques) for increasing coherence, as well as functionalities to address open technical challenges. Moreover, we envision that a process of iterative co-design of CQDs together with quantum control protocols and algorithms will enable new algorithmic quantum materials such as MCQCs that optimally combine form and function.

#### Single-photon emission in CQDs

A key component, crucial to the functionality of such technologies is the single-photon emitter, which serves as the quantum light source producing individual photons. Numerous materials have already been made to develop a practical single-photon source including colour centres in diamonds<sup>6,7</sup>, neutral atoms<sup>8,9</sup>, trapped ions<sup>10</sup>, two-dimensional materials<sup>11,12</sup>, organic molecules<sup>13,14</sup> and semiconducting quantum

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dots<sup>15,16</sup>. Key performance metrics of single-photon emitters are single-photon purity (that is, antibunching), brightness and indistinguishability<sup>17</sup>.

While each class of single-photon emitter has its unique strengths and limitations, semiconductor quantum dots have emerged as a leading solid-state single-photon emitter platform. Solid-state epitaxial quantum dots (EQDs) have demonstrated site-control growth<sup>18</sup>, integration of individual EQDs into microcavities<sup>19,20</sup>, electrical contacts to make devices and a versatile tunable emission from the visible to the infrared<sup>15</sup>. Although self-assembled EQDs have recently been integrated in photonic circuit platforms for QIP applications<sup>21-23</sup>, this hybrid integration comes at the cost of appreciably higher fabrication complexity as well as additional losses (for example, transferring quantum emission from the EQD host semiconductor membrane to the photonic integrated circuit waveguide layer).

These challenges motivate the continued development of CQDs, including perovskite quantum dots (PQDs). CQDs provide simpler and more scalable integration methods into QIP systems. As solution-processed semiconductor nanocrystals, their colloidal nature enables facile synthesis with capabilities of upscaling<sup>24,25</sup>. Through size control and surface engineering, their wavelength response and optical/magnetic properties can be engineered<sup>24</sup>. In addition, while retaining their coherence properties, they can be patterned with well-defined atomic spacing into one-dimensional and two-dimensional arrays and can be integrated easily into a wide range of nanophotonic structures to improve optical control<sup>26–28</sup> including assemblies of quantum emitters<sup>29,30</sup>, nanoparticle quantum memories, and auxiliary technologies such as laser amplifiers<sup>31</sup>, filters and detectors<sup>32,33</sup>.

In terms of single-photon emission, CQDs are aided by fast non-radiative biexciton recombination. These Auger-mediated processes mitigate emission from multiexciton states<sup>34</sup>, which would otherwise increase the probability of two-photon emission. Fast multiexciton Auger recombination (for example, 70 ps; ref. 35) thus ensures



spectra measured at zero and non-zero magnetic field where the Zeeman splitting is observed with the corresponding ODMR frequencies 0,  $v_1$  and  $v_2$ . **d**, Prospective material platforms that can be used for spin-qubit generation and control. Left: low-dimensional perovskites can be doped with magnetic cations and chiral ligands. *n*, the number of layers in the crystal structure of reduced-dimensional metal halide perovskite materials. Right: donor defects in ZnO crystals.

single-photon purity<sup>36,37</sup>, suppressing multi-photon events normalized to an equally bright coherent state by more than 10 times<sup>38</sup>. In 2019, shortly after the report of the bright triplet exciton in lead halide perovskites<sup>39</sup>, an optical coherence time ( $T_2$ ) of 80 ps was demonstrated in CsPbBr<sub>3</sub> PQDs<sup>40</sup>. Since then, there has been a growing interest to explore the potential of CQDs as sources of both pure and indistinguishable single photons. Recently, the Hong–Ou–Mandel effect was reported from the interference between indistinguishable single photons of CsPbBr<sub>3</sub> PQDs with visibilities of up to 0.56 (ref. 41). Similarly, heavy-metal-free InP-based CQDs have recently been shown to exhibit single-photon purities ( $g^{(2)}(0)$ ) of ~0.08 in the absence of spectral filtering and  $T_2 = 250$  ps (ref. 42). The introduction of a laser cavity, utilizing the Purcell effect, is expected to improve indistinguishability further.

#### **Optical interfaces to long-lived spin states**

Some single-photon emitters have associated solid-state electron and nuclear spins, unlocking many opportunities in the field of QIP. A leading example is the nitrogen-vacancy defect in diamond, exhibiting well-defined optical transitions and long-lived spin states<sup>43</sup>. Optically active spin-based solids enable the development of memories as well as localized nanoscale sensing and imaging. Furthermore, the resonantly driven magnetic interaction also enables coherent control of the system's fine and hyperfine states, enabling storage and processing of quantum information akin to methods demonstrated previously in semiconductor quantum dots<sup>44</sup>.

Towards this aim, semiconductor nanocrystals containing dopant atoms show great potential. The most common example is a dilute magnetic semiconductor, where a transition metal ion is doped into a semiconductor<sup>45</sup>. Upon the application of a magnetic field, the energy levels associated with spin-up and spin-down states undergo a splitting phenomenon (Zeeman splitting). This leads to a distinct separation in energy levels. Such a configuration forms a two-level system that can serve as a quantum bit (qubit), enabling spin-qubit control

### BOX 1

# Non-classical light emission

**Single-photon purity.** The probability of a light source to generate no more than one photon per excitation cycle, quantified by the second-order intensity correlation function  $g^{(2)}(\tau=0)$  in a Hanbury–Brown–Twiss interferometer<sup>103</sup>.  $g^{(2)}(0)$  values close to 0 are often considered a strong indicator of photon antibunching. Currently, the typical  $g^{(2)}(0)$  values of CQDs can reach ~0.05 (refs. 40,104).

**Determinicity.** Deterministic single-photon sources show non-classical behaviour by emitting precisely one photon at a specified time with a high level of certainty<sup>105</sup>. In practice, a single-photon source must have sufficient collection efficiency to be truly deterministic.

**Optical coherence time (T**<sub>2</sub>). The time duration over which the photon maintains its phase<sup>106</sup>.  $T_2$  values up to ~250 ps and 80 ps have been demonstrated for InP-based CQDs and CsPbBr<sub>3</sub> PQDs, respectively<sup>40,42</sup>.

**Purcell effect.** The modification of spontaneous emission in an engineered electromagnetic environment<sup>107</sup> (see figure).

**Photon indistinguishability.** The extent to which two optical pulses are identical in their quantum optical fields. This is represented by observing two identical photons coalesce in the Hong–Ou–Mandel interferometer. Measurements from CsPbBr<sub>3</sub> PQDs show a visibility of 0.56±0.12 (ref. 41). In general, photons lose their indistinguishability due to exciton–phonon coupling and interactions with a fluctuating environment over their dephasing time  $(T_2^*)$ . The

overall optical coherence time  $T_2$  is defined as  $\frac{1}{T_2} = \frac{1}{2T_1} + \frac{1}{T_2^*}$  (Fig. 1a), where  $T_1$  refers to radiative lifetime<sup>108</sup>.

Zeeman splitting. The phenomenon where the energy levels of elec-

trons in semiconductors are split in the presence of a magnetic field due to the interaction between the magnetic field and the spin<sup>109</sup> (Fig. 1b).

**Quantum sensor.** A system with four necessary attributes: (1) discrete, resolvable energy levels, (2) the possibility to be initialized into a well-known state capable of readout, (3) the ability to be coherently manipulated, and (4) the capability of interactions with a relevant physical quantity<sup>4</sup>.



**Non-classical light emission.** a, Controlled directionality and Purcell enhancement from coupling of CQDs to (1) plasmonic structures and (2) a quasi-two-dimensional cavity. DBR, distributed Bragg reflector; PhC, photonic crystal. **b**, Progress towards indistinguishability and entanglement by (1) post-growth tuning of the biexciton–exciton cascade to reduce the fine-structure splitting and (2) coupling the CQDs to a nanophotonic platform with a nanobeam and ring resonator to improve indistinguishability<sup>110</sup>. FSS, fine structure splitting;  $\sigma^+$  and  $\sigma^-$ , circularly polarized light; *J*, the coupling rate between the nanobeam cavity and ring resonator;  $\gamma$ , the decay rate.  $\gamma^*$ , the dephasing rate of the collidal quantum dot; *g*, the QD to nanobeam cavity coupling rate;  $\kappa_1$  and  $\kappa_2$  are the decay rates of the nanobeam cavity and ring resonator, respectively. Panel **b** adapted with permission from ref. 110, American Chemical Society.

and therefore QIP<sup>46-48</sup>. Owing to direct sp-d exchange between the dopant and semiconductor valence and conduction bands, giant Zeeman splitting occurs (Fig. 1b). Thus, the energy window is expanded (Fig. 1c) between spin states for energy-selective readout based on spin-to-charge conversion<sup>46,49</sup>.

Dilute magnetic semiconductor exploration has been carried out extensively with CdSe CQDs and Mn<sup>2+</sup>, where Zeeman shifts of greater than 50 meV have been shown<sup>50,51</sup>. Single Mn<sup>2+</sup> magnetic impurities in CdSe have also been explored, providing spin qubits with fewerspininteractions<sup>52</sup>. Incontrast, doping in PQDs (as well as quasi-twodimensional perovskites; Fig. 1d) has not resulted in giant Zeeman splitting<sup>53</sup>. However, instead of a magnetic field, photoexcitation has been used to demonstrate giant Zeeman splitting to the bright triplet state of  $Mn^{2+}$ -doped CsPbBr<sub>3</sub>, providing a potential pathway forwards<sup>54</sup>.

Nanoparticles doped instead with rare earth ions (REIs) show photoswitching behaviour that can be promising for QIP<sup>55</sup>. Timeresolved studies have revealed dynamic spin-state transitions where, by tuning the laser excitation from quasicontinuous to pulsed, individual or ensembles of spins can be photoswitched, respectively<sup>56</sup>. Reversible switching of strong light-matter coupling has been also demonstrated for bistable spin-crossover molecular materials<sup>57</sup>. Furthermore, REIs embedded in solid-state hosts and nanocrystals



a, Top: quantum sensing set-up with a laser beam focused through an objective lens, a microwave ring antenna and a permanent magnet. Bottom: a close-up image of the sensing components showing a receiver (RX), nanoparticle (NP) and transmitter (TX). The upconverted light at the receiver is transferred to the NP where the sensing takes place (or the sensing can be done at the receiver if it has good spin properties). The signal is enhanced at the transmitter before it is received at the detector. b-e, Based on the characteristics of the NP, different possible sensing modalities include: electrometry (E; b), magnetometry (M; c), thermometry (T; d)

and pH sensing (**e**).  $\lambda$ , the excitation/emission wavelength;  $I_{PL}$ , photoluminescence intensity;  $B_{NV}$ , the magnetic field along the nitrogen-vacancy centre;  $f_B$ , the frequency of the magnetic field;  $f_{PL}$ , the frequency of photoluminescence;  $\Delta f$ , the shift of the photoluminescence frequency;  $\gamma_{NV}$ , the gyromagnetic ratio of the nitrogen-vacancy centre;  $B_0$ , the applied magnetic field strength along a nitrogenvacancy axis. The inset in Fig. 2e shows how the zero-field splitting value varies with the measured temperature, which is modelled using a linear function. Panels adapted with permission from: **b**, **c**, ref. 102, under a Creative Commons licence CC BY 4.0; **d**, **e**, ref. 78, under a Creative Commons licence CC BY 4.0.

have highly coherent intra-4*f* optical and spin transitions suitable for a wide range of quantum applications, including quantum memories and transduction<sup>58-60</sup>. However, a notable challenge arises from their long lifetimes, which leads to low emission rates<sup>59</sup>. Recent advancements have focused on enhancing the low emission rates of REIs. This enhancement is achieved by integrating the ions with optical cavities and Fabry-Pérot resonators, which effectively shortens the lifetimes ( $T_1$ ) of the emission while simultaneously increasing the fluorescence yield. This has enabled single-shot readout and spin manipulation of optically addressable single REIs<sup>61-64</sup>.

In addition to metal doping, other routes are available to improve QIP applications via the addition of charge donors and acceptors. PQDs have been functionalized with anthraquinone molecules, which act as an electron acceptor<sup>65</sup>. After photoexcitation, anthraquinone extracts an electron from the PQD, which then exhibits a long-lived hole spin precession of hundreds of picoseconds. Neutral shallow

## BOX 2

# Semiconductor nanocrystal basics

**Semiconductor quantum dots.** Isolated nanometre-sized crystals that show a quantum confinement effect, leading to discrete energy levels and emission of quantized light<sup>111,112</sup> (panel **a**). Depending on their fabrication methods, these can be solid-state EQDs or CQDs.

**Colloidal quantum dots (CQDs).** Solution-processed semiconductor quantum dots that are stabilized and dispersed in non-polar solvents with the help of long-chain organic ligands<sup>25</sup>. Upon perturbation with bias or light, CQDs show a very bright luminescence on account of exciton confinement, with quantum efficiency that can reach near unity<sup>24</sup>.

**Types of CQD.** CQDs are commonly classified as II–VI (CdS, ZnS and so on) III–V (InP, InAs and so on) or IV–VI (PbS, PbSe and so on) CQDs based on the periodic table groups<sup>112,113</sup>. Another class of CQDs is based on metal halide perovskites (APbX<sub>3</sub>; A=Cs, CH<sub>3</sub>NH<sub>3</sub>; X=Cl, Br, I) known as PQDs<sup>113</sup>. Alloyed CQDs constitute a subclass that benefit from composition-dependent bandgap tuning in addition to quantum size effects on bandgap. CQDs can emit in the ultraviolet, visible, near-infrared or mid-infrared region of the electromagnetic spectrum depending on the class and particular type/composition of nanocrystal.

**Synthesis of CQDs.** The synthesis of CQDs is widely carried out using three popular methods: hot injection, thermal annealing and ligand-assisted re-precipitation (LARP) methods<sup>114</sup>. The hot injection method is very efficient in obtaining monodisperse particles by separating the nucleation and growth phases of CQDs, allowing more control over the size and shape of CQDs. This method works well for II–VI and IV–V CQDs, and PQDs. In the thermal annealing method, annealing at a high temperature promotes simultaneous nucleation and growth of CQDs<sup>115</sup>. This method relies on the use of highly reactive precursors and has been used to synthesize III–V CQDs, which are otherwise challenging to synthesize via the hot injection method. The LARP method works on a principle that involves the precipitation of reactant species from polar to non-polar media with the help of organic ligands<sup>113</sup>. Like the thermal annealing method, the LARP method provides less control over size and shape of particles.

**CQDs solids.** Assembly of CQDs into solids on a desired substrate can be achieved via well-established techniques such as spin-coating, dip-coating, blade-coating and so on<sup>26,116</sup>. CQDs have a tendency to self-assemble onto a substrate while also adapting ordered structures

(for example, upon mixing two different types of material)<sup>26,116</sup> (panel **b**). The 'pick and place' technique has been used as a proof-of-concept demonstration for deterministic placement<sup>117,118</sup>. Another way of assembly is via a template, and in this type of assembly, CQDs can be placed between two metallic contacts<sup>26,27</sup> (panel **c**).

**Blinking.** CQDs have a long-standing issue of exhibiting photoluminescence intermittency, vividly observed at a single-particle level, and is commonly termed as blinking<sup>119</sup>. This blinking is detrimental for quantum technologies that demand continuous supply of single photons. Poor surface passivation and the presence of surface dangling bonds contribute substantially to photoluminescence blinking<sup>40</sup>.

Surface passivation. The surface passivation of CQDs is crucial to enhance photoluminescence quantum efficiency and to avoid blinking problems<sup>40,119</sup>. In conventional approaches, growing a thick shell on CQDs can effectively suppress the blinking and improve the emission yield of CQDs<sup>120</sup>. A potential outlook would be developing an asymmetric shell around faceted CQDs, which was previously used to demonstrate continuous lasing in CdSe CQDs<sup>121</sup>. In addition to the shelling methods, investigations have been conducted on resurfacing with inorganic polar ligands in III-V<sup>122</sup> and II-VI<sup>123</sup> CQDs. This process facilitates the efficient elimination of trap states by passivating surface defects. Growing a shell onto PQDs remains a challenge. In this context, zwitterionic ligands are commonly used to reduce photoluminescence blinking, enhance stability and demonstrate single-photon emission in PQDs<sup>40,124,125</sup>. Recent studies have shown the growth of an ionic matrix, including perovskite<sup>126</sup> and phenethylammonium bromide<sup>127</sup>, onto PQDs. The latter exhibits nearly blinking-free characteristics.

**Janus nanoparticles.** The integration of two or more components with distinct physical or chemical properties into a single particle. Janus particles have been used as biometric logic gates for drug delivery where one side detects the disease and outputs the signal to the other side that releases the drug after an internal processing<sup>128</sup>. They have also been demonstrated for directional heating, sensing and imaging<sup>129</sup>. The main methods of synthesizing Janus particles include masking (protecting one side while modifying the other), self-assembly (block copolymers in solvents) and phase separation (mixing incompatible phases to form one single nanoparticle)<sup>130</sup>.



**Non-classical light emission and bottom-up assembly and integration. a**, Transmission electron microscopy image of monodisperse CdSe/CdS core/shell CQDs with a core (total) size of 3.5 nm (8 nm) and a photoluminescence quantum yield of 80%. Inset: colloidal CdSe/CdS QDs in toluene under UV illumination (ref. 131). b, Self-assembly of two different types of CQD adapting a perovskite lattice structure. Inset: a close-up of the superlattice domain<sup>116</sup>. **c**, Template assisted self-assembly of CQDs in between two metallic contacts. Inset: high-resolution scanning electron microscope image of the nanohole arrays<sup>27</sup>. Panels reproduced with permission from: **a**, ref. 131, IEEE, **b**, ref. 116, Springer Nature Ltd. Panel **c** adapted with permission from ref. 27, American Chemical Society.



**Fig. 3** | **Multifunctional colloidal quantum circuits. a**, A dual-compartment Janus particle forming a p-n junction. Upon illumination, a photovoltage (*V*) is created at the interface, which can be modulated through controlling the light intensity  $I(\omega, t)$ , linking the time and frequency domains. Finally, an electric field is formed at the free space between two Janus particles owing to the opposite doping charges at the sphere ends. The junction forms the building block for **b-d**.  $V_a$  and  $V_b$ , the electrostatic potentials of QD1 and QD2; *d*, the distance between QD1 and QD2; *E*, the resulting electric field. **b**, a.c. control of spin

states in nanoparticles. **c**, A nanoinductor through the intensity modulated photocurrent  $J(\omega, t)$  and coiled carbon nanotubes (CNTs) to control the spin states in nanoparticles. **d**, Simultaneous sensing and actuation in cell membranes. **e**, A single-photon detector where the signal that is received at the QD antenna is transferred to the biased semiconducting carbon nanotubes and the excitons are channelled through exciton diffusion. SPE, single photon emitter;  $\sigma_{QDs}$ , the absorption cross section of QDs; CNTs, carbon nanotubes.

donors, while not yet tested with PQDs or other CQDs, show promise as well. Donor-bound excitons have been formed in silicon, generating qubits with coherence times in minutes, and have more recently been shown in ZnO with a  $T_1$  and  $T_2$  of ~500 ms and 0.05 ms, respectively<sup>66,67</sup>. Depending on the doping density, the application can be tailored from quantum memories to individually selected qubits for quantum computation. We expect that this type of doping will help unlock new opportunities for CQDs.

#### Deterministic placement and delivery for sensing

Within quantum technology, quantum sensing has emerged as a rapidly advancing application. Unlike conventional sensing methods, which face challenges in further optimizing localization and sensitivity, high-precision quantum sensing highlighted here is based on the utilization of quantum mechanics and spin states to detect the properties of individual particles at the nanoscale (Box 1). Solid-state spin-based sensors have achieved subpicotesla sensitivity<sup>68</sup> and subnanometre resolution in three-dimensional magnetometry<sup>69</sup>.

When compared with solid-state quantum sensors, CQDs offer entirely new approaches to address unsolved challenges in sensing. For example, they can serve as receivers in a two-photon absorption set-up with excitation wavelengths in the infrared and upconverted emission in the visible<sup>70</sup> (Fig. 2a). Longer wavelengths are desirable in biosensing as the detection can take place in a deeper area with less damage to the tissue. The upconverted photons can then be used to perform the sensing in the same CQD or transferred to another nanoparticle with good spin properties (in this example, say, a nanodiamond with nitrogen-vacancy centres)<sup>71</sup>. The sensing output signal-to-noise ratio can be further enhanced, if the nitrogen-vacancy centres have low quantum efficiency, by a Förster resonance energy transfer-assisted method to an adjacent CQD-based antenna (for example, CQDs sandwiched between two metal nanoparticles), generating an amplified emission with engineered wavelengths and narrower bandwidth that can be easily read out. Optically active spin-based nanoparticles allow for different sensing modalities. For example, each of the nanoparticles or spin-doped CQD emitters can individually serve as an optically interfaced electrometer<sup>72</sup> (Fig. 2b). Together, this creates a distributed quantum sensor surface with numerous potential applications, including electric-field noise spectroscopy with optimized spin readout sensitivity and performance beyond the standard quantum limit<sup>73</sup>.

Spin-dependent fluorescence can be extended to detect other external stimuli such as temperature and magnetic fields, enabling selectivity to individual sensing quantities<sup>74</sup>. One example is optically detected magnetic resonance (ODMR). In the presence of a magnetic field, the degeneracy of the  $|\pm 1\rangle$  state is removed and Zeeman splitting is observed (Fig. 2c). Thermometry is also possible as, in the absence of a magnetic field, the ODMR resonance at the energy of zero-field splitting is temperature-dependent<sup>75-77</sup> (Fig. 2d). Likewise, pH sensing can be obtained via ODMR by observing changes in longitudinal relaxation time (Fig. 2e)<sup>78</sup>. A key requirement, however, is having sufficient spin coherence time (ensemble  $T_2^*$  or individual-spin  $T_2$ ), which can be considered the immunity window of spin coherence against local environmental fluctuations. In general, the sensitivities of these techniques scale with  $\frac{1}{\sqrt{T_2^*}}$  (ref. 79). The additional enhancement of

sensitivity in CQDs comes from the close proximity of the CQD-based sensor to the surface, compared with other quantum sensing platforms such as solid-state colour centres in diamond and silicon<sup>80</sup>. Nanocrystals can be localized on different target samples through ligand engineering, enabling simultaneous sensing and quantification with subatomic resolution.

#### Multifunctional colloidal quantum circuits

Beyond the single-particle methods, MCQCs combine the advantages of subcomponents. In MCQCs, the CQDs are arranged in a specific pattern on a substrate and connected by thin metallic wires (for example, carbon nanotubes to create a circuit)<sup>81</sup>. The circuit can be designed to exhibit specific electronic properties, such as the ability to switch on and off or to amplify electric signals, all with an open geometry and at a scale inaccessible to current lithography tools. We propose using a Janus particle (Box 2) with a dual compartment to form a p-n junction converting the electromagnetic field (that is, laser) into an electric field<sup>82</sup> (Fig. 3a).

Recently, Janus particles with dual Au–TiO<sub>2</sub> compartments have exhibited a light-responsive, self-induced electric field. When illuminated, electron–hole pairs form in the TiO<sub>2</sub> semiconductor hemisphere and separate at the metal–semiconductor boundary. Electrons transfer to the Au metal side, serving as an electron sink, and leave the TiO<sub>2</sub> side positively charged. The resulting photovoltage depends on the potential difference between the compartments (that is, the Schottky barrier height), the light's excitation wavelength and the photon flux<sup>83,84</sup>. Sub-100 nm light-powered Janus nanoparticle motors have also been demonstrated<sup>85</sup>. Critical next steps include investigating how the physical properties scale down to the 10 nm regime.

Another promising component for MCQCs is photoswitching nanoparticles. Recently, photoswitching with high volume of photodarkening-photobrightening cycles and negligible photobleaching has been demonstrated for subångström super-resolution imaging in packed nanoclusters<sup>55</sup>. Such nanoparticles can find applications in conjunction with quantum sensing for dual imaging and enhanced sensing accuracy. Furthermore, they can be used as logic gates in MCQCs analogous to the logic gates in molecular digital information processing<sup>86,87</sup>.

Potential MCQCs devices include: localized a.c. field for spin control in qubits (Fig. 3b); nanoinductors (Fig. 3c); simultaneous quantum sensing and actuation to apply action potentials in cell membranes<sup>88</sup> (that is, to trigger a cell to fire) and to provide actuation<sup>89</sup> and feedback (for example, for epilepsy: we could detect an epileptic episode and immediately provide feedback to quench it) (Fig. 3d); and single-photon detectors with CQD antenna and semiconducting carbon nanotubes<sup>90</sup> (Fig. 3e). Finally, while using quantized states in individual quantum sensors can provide high precision and accuracy, and operate with a high spatial resolution inaccessible to classical sensors, sensitivity can be improved past the standard quantum limit, by non-classical, entangled states. The minimum detectable signals can scale as low as 1/N for an N-particle quantum state, instead of  $1/\sqrt{N}$  for an N-size ensemble measurement. Ouantum coherence (from the superposition of multiple energy states) and the more advanced entanglement-enhanced quantum sensing (between indistinguishable and distant quantum systems) would bring the sensitivity closer to the quantum metrology limit (the Heisenberg limit)<sup>91,92</sup>. Creating these entangled states could be done by, perhaps, many-body Rydberg coupling in solids, or by the maximally entangled Greenberger-Horne-Zeilinger states<sup>93,94</sup>. Further opportunities in the field of quantum sensing with CQDs will require continued advances in their synthesis, reproducibility and stability.

#### Outlook

CQDs have unique challenges as their mesoscopic environment has more fluctuations (that is, is noisier) than in conventional quantum emitter systems that are buried hundreds of nanometres or more within a homogeneous host material. This is currently a major issue for CQDs and leads to decoherence. As the quantum system interacts with the surrounding environment, phonons and other CQDs, the system loses the integrity of quantum information, and the superposed states gradually dephase and collapse into classical states. Additional sources of decoherence include phonon–exciton coupling from high temperature and excitation power<sup>95</sup>, as well as exciton–exciton interparticle interactions<sup>42</sup>.

Potential solutions include surface passivation to suppress interaction with the environment such as improved isolation of the CQDs through shell and ligand engineering, exploitation of material symmetries to reduce electric-field-induced energy level perturbations (as seen in 'symmetry-protected' group-IV colour centres), and other methods to minimize coupling to phonons and stray electric fields<sup>96</sup>. For coherent spin ground states, magnetically 'quiet' material systems such as ones with spin-neutral isotopic compositions should help.

Another promising direction includes on-chip integration with CQDs. With the rise of photonic integrated circuits for QIP applications<sup>23,97</sup>, there is a growing interest in integrating CQDs as single-photon sources. However, unlike epitaxial quantum dots where their integration into photonic structures can be complex and requires high-temperatures, CQDs can be spin-coated onto a passivated silicon substrate for large-area integration with minimal defect density<sup>98</sup> and implemented on complementary metal–oxide–semiconductor circuits enabling scaling and miniaturization<sup>99,100</sup>. Further device integration strategies are already underway.

It is therefore the right time to take advantage of the rapid progress and precise engineering of CQDs to push further the exploration for quantum optics and improve the key metrics of their single-photon emission for QIP applications and devices.

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### **Competing interests**

The authors declare no competing interests.

### **Additional information**

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