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TOPICAL REVIEW

Multi-emitter solid state quantum optics

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Abstract

Photonic quantum science—which harnesses light-matter interactions down to the single photon level—provides advanced light sources for quantum sensing, communication, and computing. Integrating solid-state light sources on a photonic chip provides a means of creating scalable on-chip quantum networks with sophisticated optoelectronic circuitry. Extending beyond single emitters to multi-emitter systems is a frontier that offers substantial opportunities for fundamental scientific advances as well as applications that require multi-emitter quantum states. A key challenge in this endeavor is the spectral inhomogeneity of solid-state emitters, which results in non-identical emitters that cannot be interfaced with one another. This review describes recent advances in tailoring the emission properties of individual emitters and the multi-emitter demonstrations that are enabled by this control for a variety of prominent materials, as well as prospects for further development.

1. Introduction

Combining the components for the generation, manipulation, storage, routing, and detection of quantum light on a photonic chip provides a compact and scalable quantum science platform. Proof-of-concept demonstrations of various combinations of these components in photonic integrated circuits (PICs) [1] have enabled fundamental tests of quantum mechanics [2], quantum simulation [2], and quantum machine learning [3, 4].

Artificial atoms are an essential building block of quantum PICs, providing a light-matter interface for optical nonlinearities down to the single-photon level [5–8], deterministic generation of single photons [9], and entangled photon pairs [10–12]. However, these quantum emitters (QEs) are among the most challenging PIC component to integrate [1], notably due to the inhomogeneous distribution of emission wavelengths and random positions within a sample. Over the last two decades, numerous advances have been achieved in a variety of material systems. For example, QEs have been integrated in solid-state diodes for charge injection [13–15], to create spin memories [16], and photonic architectures with high-Q cavities [17–19], and waveguides [7, 15, 20–25], and detectors [26]. These advances have resulted in the on-chip generation of pure, indistinguishable single photons [9, 27], entangled photons [10, 11], and spin-photon entanglement [28–30].

The extension to multiple interacting QEs provides numerous possibilities that are not possible with single QEs [31], including a path to deterministic quantum gates between emitters [32–34], and collective quantum states [35–37]. Embedding QEs in photonic crystal waveguides (PhC WGs) is a particularly versatile multi-emitter platform that provides dispersion and modal engineering regimes that are not possible in traditional cavity quantum electrodynamics [38]. In contrast to atomic emitters, solid-state emitters face challenges from variation in composition and/or differences in the local strain and electromagnetic environment. The resulting spectral inhomogeneity of the center emission wavelengths has been a major barrier for multi-emitter quantum optics. Similarly, spectral diffusion, which occurs on a smaller spectral scale, also reduces the indistinguishability of the emitted photons. In recent years, significant progress has been made in developing techniques for the precise control of the emission wavelength of individual QEs. Progress has also been made to mitigate spectral diffusion with sample growth, dynamic control, and optical techniques to achieve radiatively-limited linewidths. These advances have helped initiate exploration in the field of multi-emitter solid state quantum optics, enabling demonstrations of collective phenomena such as superradiance and subradiance.

Color center defects in crystals [1, 19, 23, 39, 40], self-assembled semiconductor quantum dots (QDs) [14, 20, 25, 41–43], and organic molecules [44–49] depicted in figure 1 represent the current state of the art for multi-emitter integrated quantum optics. This review highlights recent progress and ongoing challenges in creating multi-emitter solid state quantum optics platforms in these materials. Additionally, the development of heterogeneous integration techniques [40, 50, 51], has expanded the possibilities for integrating QEs in other material systems. Ultimately, considering low-loss and large-scale fabrication requirements, heterogeneous integration with SiN or Si PICs may be required. Although the particular challenges in achieving a multi-emitter PICs differ between materials, the progress made in color centers, QDs, and organic molecules should be readily extended to other promising material systems. Prominent multiqubit systems such as cold atoms, ions, superconducting qubits, and electronically-gated QDs are beyond the scope of this review, but are covered in [52].

2. Materials for multi-emitter quantum optics

2.1. Color center defects in crystals

The field of defects in materials for quantum optics is broad and rapidly expanding. This review is focused primarily on defects for which multiemitter demonstrations have been demonstrated.

2.1.1. Diamond

The discovery of the nitrogen-vacancy (NV) color center in diamond nearly three decades ago, and the ability to initialize, manipulate, and optically read out its spin state, has given rise to the field of diamond photonics [54]. Since then, substantial scientific and engineering effort has been devoted to producing high-quality NV centers in nanostructured devices, resulting in milestone achievements of on-demand entanglement [55], long-lived quantum memories [56], and nuclear magnetic resonance [57, 58], and multiemitter effects [59–63]. However, many applications require indistinguishable, fast, and high-efficiency single photon emission. These properties are challenging to achieve with NV centers, considering the long spontaneous emission lifetime and low emission rate into the zero-phonon line (ZPL) [64]. This has motivated the exploration of other color centers in diamond, notably the group-IV color centers (Si–, Ge–, Sn–, and Pb-vacancies) [64]. The inversion symmetry of these defects renders their optical transitions insensitive to electric field fluctuations due to the absence of the first-order Stark shift [64, 65], enabling nearly lifetime-limited single photon emission. This property is particularly desirable when integrating these defects in nanostructured devices with charge fluctuations occurring on nearby surfaces.

2.1.1.1. Silicon-vacancy (SiV) in diamond

The inversion symmetry of the group-IV color centers has been most prominently leveraged with the negatively-charged SiV center [19, 23, 53, 66, 67]. Essential components of a diamond integrated quantum network have been realized with this system, which combines a long-lived quantum memory at sub-Kelvin temperatures [68] with efficient photon generation and collection [69]. However, the inhomogeneous distribution of SiV emission frequencies and spectral diffusion have remained persistent challenges to produce multiqubit networks. The insensitivity of the SiV center to electric fields has required the use of Raman [23], magnetic field [19], and strain [66, 70, 71], tuning to bring SiV centers into mutual resonance.

Considering the essential role quantum entanglement plays in quantum networks [23, 72], optically-mediated entanglement of solid-state emitters marks an important milestone for multi-emitter quantum optics. By leveraging the Raman transitions to tune SiV centers into mutual resonance, entanglement was first demonstrated by Sipahigil *et al* in 2016 via the superradiant emission of two SiV centers in a single waveguide [23], with each defect excited by a different laser as depicted in figure 2(a). The quantum interference arising from the superradiant emission from two emitters manifests as a bunching around zero delay in a second-order correlation function, $g^{(2)}(\tau)$. In contrast, a single SiV center and two SiV centers emitting distinguishable photons have antibunched statistics with $g^{(2)}(0) \rightarrow 0$ and $g^{(2)}(0) \rightarrow 0.5$, respectively, as shown in figure 2(a). Further spectroscopic evidence of super- and subradiance was obtained by coupling two SiV centers to the same high-cooperativity cavity mode and tuning optical transitions into resonance with a magnetic field (figure 2(b)) [19].

Extending beyond two emitters is very challenging with Raman or magnetic field tuning. Furthermore, the spectral diffusion of the optical transition energy is uncontrolled with either approach. A more scalable

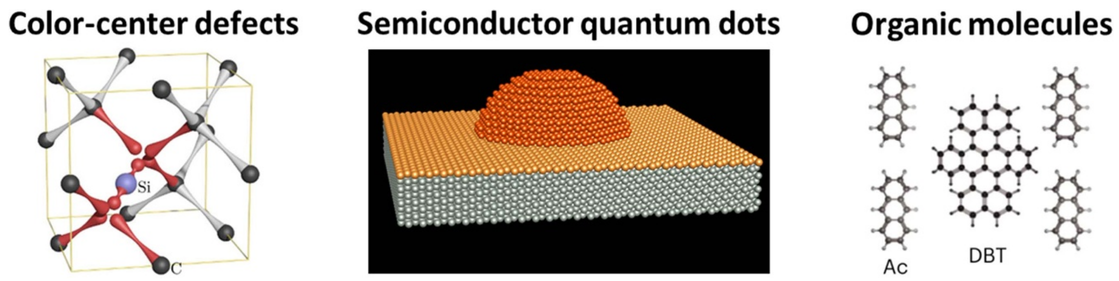

Figure 1. Cartoons depicting the three quantum emitters discussed in this review: color-center defects in crystals (Reprinted figure with permission from [53], Copyright (2014) by the American Physical Society), semiconductor quantum dots, organic molecules (Reproduced from [44], with permission from Springer Nature).

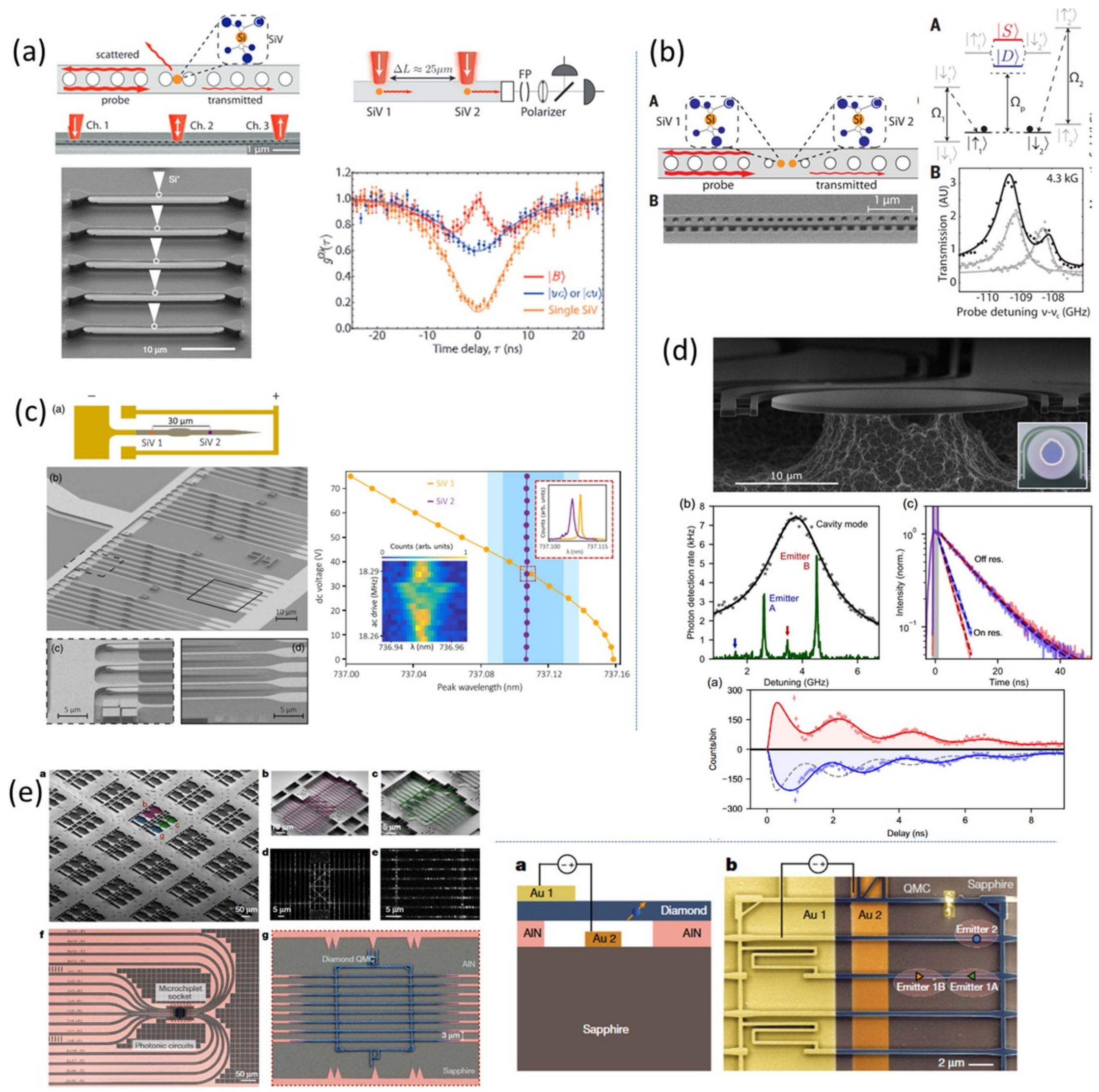

Figure 2. Defects in crystals (a) two SiV color-center superradiance in a diamond WG with Raman tuning. From [23]. Reprinted with permission from AAAS. (b) Photon-mediated interactions of SiV color-centers in a diamond nanocavity enabled by magnetic field tuning. From [19]. Reprinted with permission from AAAS. (c) Quantum interference of the emission from SiV centers with dynamic electromechanical stabilization. Reproduced from [66]. CC BY 4.0. (d) Superradiance from V_{Si} centers in a SiC microdisk resonator. Reproduced from [39]. CC BY 4.0. (e) Large scale integration of defects in silicon. Reproduced from [1], with permission from Springer Nature.

method has been demonstrated using strain to control and stabilize the emission energies of SiV defects embedded in the same electromagnetically deflectable WG, as shown in figure 2(c) [66]. In addition to allowing SiV centers to be tuned into resonance due to varying strain responses at different positions in the WG, spectral diffusion was also substantially reduced with a feedback scheme using the SiV optical transition energy and the high-bandwidth electromagnetic actuation of the WG. A similar strain-tuning scheme was used in a hybrid diamond-PIC platform for large-scale integration of SiV and GeV centers, as shown in figure 2(e) [1].

2.1.2. Silicon carbide

Color centers in silicon carbide (SiC) presents a promising, cost-effective alternative to diamond magnetometry [73] and integrated quantum optics [74, 75]. In addition, leveraging compatibility with existing silicon fabrication processes raises the possibility of repurposing existing silicon foundries for manufacturing SiC nanophotonic structures. Silicon-vacancy (V_{Si}) centers in 4H-SiC [73, 76–79], can have optical coherences that are preserved up to 20 K [80, 81], and millisecond spin coherence times [82]. Furthermore, despite the lack of inversion symmetry, several studies have demonstrated excellent optical coherence of the V_{Si} center embedded in nanophotonic structures [39, 76]. The lack of inversion symmetry also allows spectral electric field tuning over a wide range [83, 84].

The prospects for multiemitter quantum optics with SiC was highlighted by recent work in which V_{Si} defects embedded in a microdisk resonator were found to have near-transform limited linewidths. (see figure 2(d)) [39]. In addition to achieving near-unity cavity coupling for a single V_{Si} center, two multiemitter effects were reported. First, evidence of optical superradiance was observed by non-resonant c.w. laser excitation of two V_{Si} centers that were coupled to the same cavity mode (see figure 2(d)). Performing a $g^{(2)}(\tau)$ measurement on the resulting emission collected from a waveguide that was evanescently coupled to the microcavity produced the characteristic bunched photon statistics as discussed above for diamond SiV defects. Second, single-photon interference between the emitters was generated by resonantly exciting the V_{Si} centers (to minimize spectral diffusion). This manifested as temporal oscillations of the single-photon wave packet due to the relative emitter phase, as shown in the bottom panel of figure 2(d) for clockwise and counterclockwise propagation in the microdisk.

In contrast to diamond, SiC has a strong second-order nonlinearity [85], which can be used for frequency conversion, including into the telecommunication bands [86]. Along with recent advances in integrating defects into photonic nanostructures and the single dipole orientation of defects such as V_{Si}, SiC is positioned as a highly promising platform for multiemitter quantum optics.

Prominent challenges faced by diamond and SiC material platforms in developing scalable quantum optics systems include inefficient emission into telecommunication bands and the lack of large-scale manufacturing processes. One solution to the former challenge is nonlinear frequency conversion into the telecommunication bands [86], although loss in the conversion process may exclude some applications. There has also been recent discovery of defects that emit in the telecom O-band silicon-doped diamond [87] and vanadium-doped SiC [88]. Significant progress has been made on the latter challenge with heterogeneous integration techniques [1, 50, 89], which interface color centers with low-loss PICs.

2.1.3. Silicon

A promising alternative for large-scale integrated quantum optics has recently emerged involving the direct incorporation of optically-active defects into silicon that emit directly into the telecommunication bands, including the G-center [90–92], W-center [93–95], and T-center [96]. Several defects have been embedded in nanophotonic cavities and waveguides, including the relatively bright G-center, and T-center that offers a long-lived ground state electron spin [97]. Although the fastest radiative emission rates have been modest, sub-nanosecond Purcell-enhanced emission rates have been achieved by coupling the G-center to a nanobeam photonic crystal cavity [98]. Initial steps have also been take to control the optical emission wavelength G centers in Si using 532 nm laser irradiation [90] and strain [99], but, as in diamond and SiC, emission broadening due to spectral diffusion remains a challenge.

2.2. Semiconductor QDs

2.2.1. QD introduction

Self-assembled InGaAs semiconductor QDs grown by molecular beam epitaxy are the state-of-the-art solid-state source of bright, high-purity, and indistinguishable single photons [12]. They have also been incorporated into sophisticated photonic circuitry combining waveguides, cavities, and on-chip superconducting detectors [25, 100, 101]. This progress has enabled demonstrations of the entanglement of spin memories with single photons [28–30], photon-mediated entanglement of distant spin qubits [102, 103], and the generation of cluster states [11]. However, the short spin coherence times in QDs has limited their use as spin qubits. This has motivated recent work to extend the coherence time by coupling electron spins to collectively-excited nuclear spin ensembles [104, 105].

2.2.2. Spectral inhomogeneity and tuning techniques

A prominent challenge for QDs is the scale of the inhomogeneous distribution, which is typically tens of meV for InGaAs QDs and a few meV for both droplet epitaxy GaAs QDs [106, 107], and mesa-top InGaAs QDs [108]. This is orders of magnitude greater than the few-μeV individual QD homogeneous linewidths, which determines the resolution that spectral matching must be done. This presents the demanding requirement for a tuning mechanism that can shift QD emission wavelengths on an meV scale with μeV resolution. As a consequence, substantial attention has been devoted to developing techniques to tune the emission wavelengths of QDs. The Stark [109] and ac Stark effects [110], temperature [24, 111], and Raman transitions [14, 112], have all been used for dynamic, precise, short-range tuning. However, the limited tuning range combined with the significant fabrication overhead and/or operational complexity make it challenging to independently tune more than two QDs in within the same cavity or waveguide with these techniques. Longer range techniques that can tune QDs across the entire inhomogeneous distribution with high resolution has been accomplished via strain using piezoelectric actuators [113–115], or strain caused by a laser-induced phase change of a thin film deposited on the sample surface [7, 15, 116].

These advances in controlling the emission properties of QDs, in combination with their fast, bright, and indistinguishable photon generation, has established them as an ideal quantum optics testbed. This has led to numerous recent multi-emitter quantum optics demonstrations. Embedding QDs in PhC WGs creates an advantageous multi-QD environment by providing strong light confinement to enhance the light-matter interaction, and extending the spatial range of dipole-dipole interactions far beyond the sub-wavelength range in free space. This system has been used to generate two-QD superradiance, which was achieved by bringing the QDs into mutual resonance using temperature (figure 3(a)) [24], Zeeman (figure 3(b)) [117], and electric field (figure 3(c)) [118] tuning. In [117]and [118] the dynamics of both super- and subradiance were probed directly by observing enhanced and suppressed emission rates, respectively, as shown in the right panels of figures 3(b) and (c). In the same work, it was also shown that the QD detuning and excitation conditions could be used to control those dynamics [117]. Extending beyond two QDs within the same PhC WG is challenging with the tuning approaches used in these two-QD demonstrations. Extending beyond two QDs was made possible using a strain tuning technique involving local laser-induced crystallization of a thin phase change material [15, 119], deposited on the surface of the PhC WG [15]. As shown in figure 3(d), controllable quantum interference between two and three QDs was enabled using this technique, resulting in bunched photon statistics with $g^{(2)}_{N=2}(0) \rightarrow 1$ for two QDs and $g^{(2)}_{N=3}(0) \rightarrow 4/3$ for three QDs, in good agreement with theory.

Ultimately, the optical propagation loss III–V waveguides—often exceeding 15 dB cm^{-1} [120]—is a limitation that must be addressed to advance integrated quantum optics with QDs toward large-scale quantum information technologies. Hybrid integration of QDs with Si and SiN PICs has emerged as a promising solution to this challenge [50, 51, 89, 115, 121–123]. Figure 3(e) shows a recent advance in this area was accomplished with the large-scale integration of telecom Stark-tunable InAs/InP QDs on a silicon-on-insulator PIC platform that was fabricated in a 300 mm foundry [40]. The spectral instability of the InAs/InP QDs remains a challenge, but new techniques for QD selection and integration [124], advances in QD growth [125, 126], and charge stabilization [107] provide a path to achieving the interactions between multiple emitters.

2.3. Organic molecules

Single molecules, particularly polycyclic aromatic hydrocarbons (PAHs), have emerged as coherent QEs [44–49]. They exhibit ultranarrow lifetime-limited emission linewidths down to 10 MHz at cryogenic temperatures [127, 128], optically-addressable spin memories [129], and facile integration with nanophotonic structures [45, 130–133]. Furthermore, they offer inexpensive mass production of nearly identical QEs [134].

PAH molecules are often introduced as low-concentration impurities into a host crystalline lattice, such as anthracene [47]. Similar to color center defects and QDs discussed above, individual molecules can be optically excited and measured. Although molecules can be identical to one another, local strain [135] and charge variations introduce spectral inhomogeneities that can range from a few GHz [136] to a few thousand GHz [132] depending on the preparation of the host material. The Stark effect has become a well-established approach to compensate for this inhomogeneity [137, 138]. It was used to tune the emission wavelengths of dibenzoterrylene (DBT) molecules doped into anthracene nanocrystals across the entire inhomogeneous distribution, exceeding 400 GHz [139]. However, implementing the electrically-induced Stark effect faces similar scalability challenges as those discussed above for QDs. To enable scalable and independent tuning of multiple molecules on the same chip, a fabrication-free optically-induced Stark tuning approach was recently introduced [134, 140]. In this approach, laser irradiation was used to tune the DBT ZPL by inducing long-lived charge-separated states that can persist for several hours at cryogenic temperatures and allow the

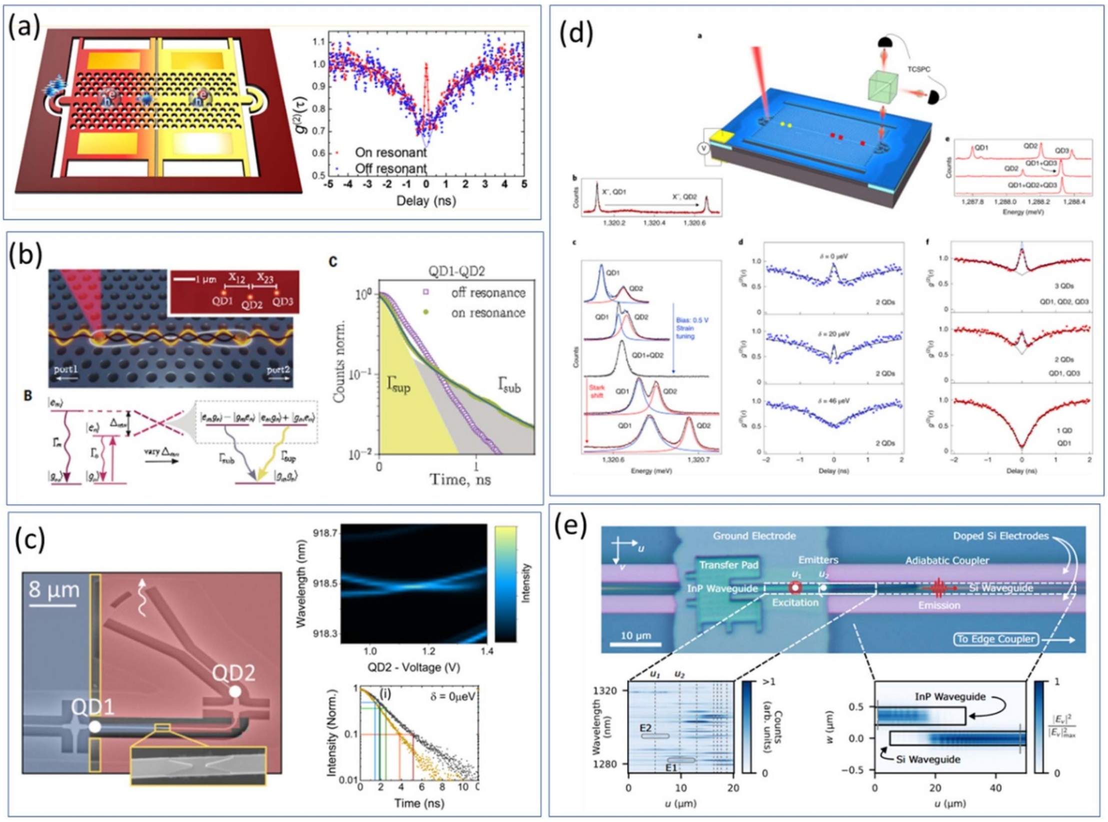

Figure 3. Semiconductor quantum dots (a) temperature tuning QDs in a PhC WG to produce two QD superradiance. Reprinted with permission from [24]. Copyright (2018) American Chemical Society. (b) Two-QD super- and subradiant dynamics achieved by magnetic field tuning QDs into resonance. From [117]. Reprinted with permission from AAAS. (c) Two-QD superradiant dynamics achieved by tuning QD emission with an electric field. Reproduced from [118]. CC BY 4.0. (d) Scalable strain tuning approach used three-QD quantum interference. Reproduced from [15], with permission from Springer Nature. (e) Heterogeneous integration and tuning of QDs in foundry silicon photons. Reproduced from [40]. CC BY 4.0.

emission wavelength to be continuously tuned [134]. Figure 4(a) shows this technique being applied to tune five DBT molecules into near resonance on the same chip.

These wavelength tuning methods, combined with the relatively small spectral inhomogeneity and radiatively-limited linewidths below 3 K, have enabled the study of controlled multi-emitter quantum optics in molecular systems (figure 4) [44–46, 48, 141, 142]. In [44], the optically-induced Stark effect first demonstrated in [134]. was used to tune radiatively-limited DBT molecules embedded in an anthracene host crystal with close spatial proximity (∼10 nm) into resonance, as shown in figure 4(c). Optical pumping of that system resulted in the formation of sub- and superradiant states, supported by spectroscopic, lifetime, and second-order photon correlation measurements [44]. Similarly, evidence for both sub- and superradiance was shown with dibenzanthanthrene (figure 4(c)) [48] and terrylene (figure 4(d)) [46] molecules. As in the case of QDs [48, 117]. direct evidence for sub- and superradiance is provide with longer and shorter emission lifetimes, respectively. Although the nanoelectrodes used to implement Stark tuning of the molecular optical resonances in [48]. provides desirable dynamic control of the emission wavelength, it remains challenging to scale this approach.

Manipulating the classical coherent state of laser light with a two-level QE in a waveguide is an effective means of producing a nonclassical output [5–7, 143], including energy-time entangled bound states of light [144]. In [45], this concept is realized for two molecules in a hybrid TiO_2 photonic circuit. Figure 4(e) shows a micrograph and schematic of two DBT molecules embedded on a microdisk resonator that is evanescently coupled to excitation and collection waveguides [45]. Two molecules located on opposite sides of the resonator were Stark-tuned with integrated microelectrodes, resulting in collectively-enhanced extinction of coherent laser light in both the CW and CCW propagating modes of the resonator.

The absence of an optically-addressable spin due to the singlet ground state of PAH molecules limits their use in some quantum information protocols [47]. However, the discovery and development of optically-addressable spins in organometallic, chromium (IV) molecules [129] opens new avenues for pursuing spin-photon interfaces [145]. Similar to color center defects and QDs, spectral wandering due to charge noise is also a persistent challenge with organic molecules. Recent work has made progress in

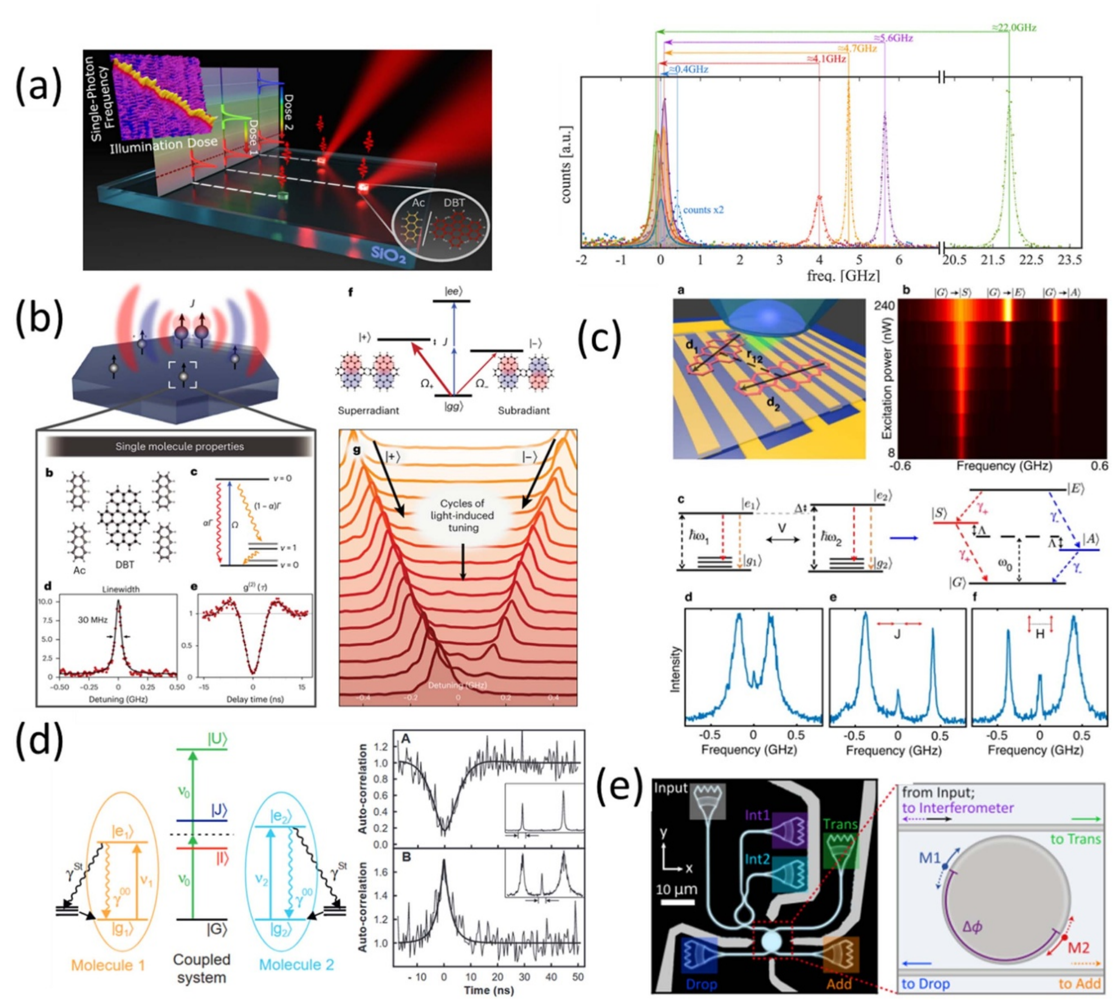

Figure 4. Organic molecules (a) fabrication-free tuning of five DBT molecules with a Stark effect induced by local laser irradiation. Reprinted with permission from [134]. Copyright (2020) American Chemical Society. (b) Super- and subradiance from two DBT molecules achieved by wavelength tuning with a laser-induced Stark effect [44]. Reproduced from [44], with permission from Springer Nature. (c) tailoring the sub- and superradiant nature of two dibenzanthanthrene molecules through Stark shifts to control the degree of superposition in their electronic states. Reproduced from [48]. CC BY 4.0. (d) Coherent optical dipole coupling of two terrylene molecules. From [46]. Reprinted with permission from AAAS. (e) Collective scattering of laser light on two DBT molecules embedded on a mircrodisk resonator. Reproduced from [45]. CC BY 4.0.

suppressing spectral wandering by combining an electric field generated by electrodes with the local, optically induced Stark effect [140]. More details on the use of organic molecules for quantum science and technology and the photophysics can be found in recent reviews (see [47, 145]).

3. Outlook

We have presented a brief overview of recent progress in multi-emitter quantum optics for three prominent material systems: color center defects, semiconductor QDs, and organic molecules. The spectral inhomogeneity of these emitters—resulting in distinguishable, non-interacting emitters—has been a prominent challenge. Substantial research has been devoted to the development of wavelength tuning techniques that has enabled the different emitters to be brought into mutual resonance. This effort has resulted in techniques that can be applied to emitters embedded PICs that are coupled to the same photonic mode, and enabled recent demonstrations of controllable collective QE states including sub- and superradiant states. These quantum phenomena serve both as important fundamental tests, as well as steps toward applications of technological importance, such as long-lived quantum memories [146, 147], and quantum logic gates [34].

Despite the progress in tuning QEs into mutual resonance, variation in other properties such as the homogeneous linewidth (which depends on the radiative lifetime). One solution to the variation in homogeneous linewidths is to tailor the temporal profile using coherent Raman photons in a Λ energy-level system, which can be used to produce tunable single photon pulses far below the emitter radiative lifetime

[148]. However, scaling this approach is challenging because of the substantial experimental overhead to excite and tailor Raman photons for each emitter. A more scalable approach involves characterizing and integrating selected emitters onto PICs using correlative microscopy [124], which combines microscopy of QEs in a cryogenic environment with the subsequent lithographic placement in photonic structures.

Near-unity photon indistinguishability from multiple emitters integrated on the same chip remains an ongoing challenge, yet is essential for many quantum photonic applications [106, 149, 150]. Addressing this challenge will likely involve wavelength tuning, suppression of spectral diffusion (e.g. via resonant excitation or dynamic stabilization), and heterogeneous integration techniques in combination with pre-characterization and selection of QEs.

Data availability statement

No new data were created or analyzed in this study.

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