

Chiral Single-Photon Generators

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Cite This: *ACS Nano* 2021, 15, 1912–1916

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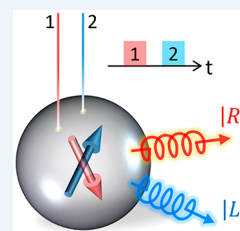


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ABSTRACT: Chiral photons have the potential to advance information technologies due to their robustness in carrying binary data against noisy backgrounds as well as their capacity for constructing single-photon isolators and circulators through nonreciprocal photon propagation. In this Perspective, we highlight recent efforts to generate chiral single photons using circularly polarized light sources. We delve into possible future technologies that integrate these light sources with other active optical elements as a versatile platform for information processing.



Photons are ideal information carriers due to their ultrafast transmission speed and minimum interactions among themselves. The information carried by photons is encoded into waves by frequency, amplitude, and phase modulation. Optical chirality, classified by right- and left-circular polarization, provides additional encoding capacity.¹ In particular, combining the chirality of light with single-photon emission has created a new forefront of research in quantum optics. A single-photon source opens the gateway for manipulating light–matter interactions at the single-quanta level. Investigations of the superposition and entanglement properties of quantum states enhance secure communication and quantum computing. In this Perspective, we highlight recent advances in chiral light generators and discuss the possibility of harnessing chiral single photons for future applications.

Thanks to continuing improvements in nanofabrication techniques, quantum emitters that combine high brightness, high single-photon purity, and high indistinguishability have been routinely demonstrated in various materials systems. Nanostructures can boost the quantum emitters' emission rates *via* the Purcell effect and increase collection efficiencies *via* mode modification. One key feature of these nanostructures, especially nanofibers and photonic waveguides, is their naturally chirality-dependent propagation, known as optical spin–orbit coupling.^{2,3} Interactions between this momentum-locked light and a polarization-dependent quantum emitter lead to propagation-direction-dependent absorption and scattering, enabling the assembly of nonreciprocal single-photon devices and deterministic spin–photon interfaces. These efforts have opened up a new frontier of research known as chiral quantum optics,⁴ which takes advantage of unidirectional coupling to control the flow of light and to tailor the phase of photons. Efficient chiral light–matter interactions have empowered on-chip universal quantum computing based on high-fidelity intranode two-qubit parity measurement and

internode entanglement,⁵ providing an example of practical application.

Combining the chirality of light with single-photon emission has created a new forefront of research in quantum optics.

One way to generate circularly polarized single photons is to enable the transfer of angular momentum from excitons to photons upon recombination of charge carriers. For quantum emitters such as InGaAs quantum dots, the chirality of scattered photons is not immediately distinguishable due to the degeneracy in trion transitions. A strong magnetic field is applied to lift this degeneracy to determine the definite helicity of emissions. Integration of quantum dots in microring resonators⁶ or nanobeam waveguides⁷ offers spin-controlled photonic flow, underpinning the essential technique to build nonreciprocal single-photon devices, such as on-chip single-photon isolators and circulators.⁸ Alternatively, chiral single photons can be obtained by passing the linearly or elliptically polarized light emitted from a single-photon source through a chiral antenna⁹ or metastructural filters (Figure 1a).¹⁰ These structural filters acquire chiral selectivities by actuating its three-dimensional (3D) shape to the rotation of the electric field in circularly polarized light, which results in an enhanced response to one specific chirality as compared to the other. In

Published: February 5, 2021



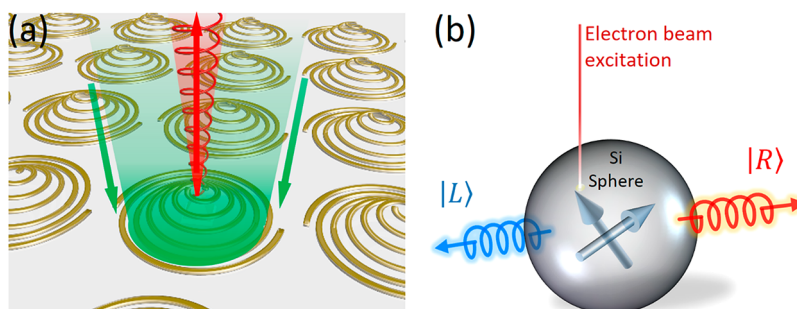


Figure 1. (a) Schematic for generation of chiral single photons with metastructure filters. The single photons (red arrow) emitted by a quantum emitter under nonresonant excitation (green arrow) acquire a specific chirality after passing through a three-dimensional spiral structure that is engineered to possess an imbalanced response to different chiral light. (b) Generation of chiral photons by using electron beam excitation. Depending on the excitation beam's energy and location, two orthogonal electric dipoles can be generated on the nanosphere with a specific phase difference to support circularly polarized light emission, as demonstrated by Matsukata *et al.* in ref 16.

particular, 3D pinwheels¹¹ can produce a circular dichroism of 0.6 in transmitted infrared light, and 3D spirals¹² are able to polarize the THz beam up to an ellipticity of 28°. Recently, Cai *et al.* reported a fabrication method based on simultaneous modulation of resist stencils and base molds that were applied to fabricate 3D spirals and were able to reduce its dimension to the micron regime with sub-100 nm spatial resolution.¹⁰ This advance extends the working frequency of these patterned chiral filters to the near-infrared and visible regimes where an abundance of quantum emitters exists and can be incorporated for technological development.

Breakthroughs at room temperature have been made by utilizing the strong chiral coupling induced by the evanescent field of plasmonic nanostructures to modify the emission properties of achiral emitters. In principle, magnetoelectric coupling in plasmonic nanoparticles enables the transfer of chiral optical information from near-field to far-field.⁹ As demonstrated with achiral colloidal quantum dots placed next to chiral gold nanoslits, these nanostructures modified the achiral absorption and emission properties of bare quantum dots, yielding a 17% degree of circular polarization (DCP).¹³ Kan *et al.* achieved higher DCP using sculptured plasmonic nanostructures with diamond nitrogen vacancy (NV) color centers.¹⁴ The nonradiative coupling between the NVs and SiO₂–Ag substrate generated surface plasmon polaritons that could interact with structured optical metasurfaces atop the substrate. The authors carefully designed the metasurface to possess a specific phase gradient, enabling collimated emissions of single photons with a chirality up to 80%. In contrast, the maximum DCP for bare NVs is 10% at a magnetic field of 10 T.¹⁵ Thus, spin-coded single-photon states can be tailored with far-field distribution at room temperature, facilitating applications in laser suppression through resonant photoluminescence excitation, optical circular dichroism engineering, and quantum key distributions. As pointed out by the authors, with some modifications to the shape and size of circular nanoridges, it may even be possible to produce vector beams out of the emission possessing nontrivial orbital angular momentum.

In this issue of *ACS Nano*, Matsukata *et al.* report an approach for exciting single silicon nanospheres with an electron beam.¹⁶ The axial excitation beam breaks the spherical symmetry of the silicon nanosphere and generates the needed electric and magnetic multipoles to control the emission polarization (Figure 1b). To harness circularly polarized light from excited multipoles, Matsukata *et al.* judiciously adjusted the electron beam energy and the excitation location on the

sphere to ensure that only two orthogonal degenerate electric dipoles or one electric plus one magnetic dipoles were excited over the sphere. The interference between these dipoles with a well-controlled phase produced high-quality circularly polarized light along specific emission directions. One key feature of this approach is the controllability of the excited multipoles and the relative phases *via* excitation parameters, presenting potential advantages for on-demand generation of circularly polarized light. This method is extremely useful for binary data encoding because chiral photons are generally robust against accidental errors. In addition, the energy of scattered photons can be tuned by changing the size of the nanosphere. In combination with the techniques of field electron emission and Yagi-Uda antenna for directing emissions,¹⁷ photonic integration and scale-up of this electron-beam-based circularly polarized light scheme is promising.

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FUTURE DIRECTIONS

Chiral Molecule Detection Using a Single Chiral Photon. Plasmonic chiral metasurfaces are known for the chiral-dependent enhancement of electromagnetic fields that underpin ultrasensitive probing of chiral molecules. In practice, the difference in a sample's response to left- and right-handed fields can be enhanced by factors of 10⁶ compared to those observed in optical polarimetry measurements, thus enabling picogram quantities of adsorbed molecules to be characterized.¹⁸ To push this probing sensitivity to the ultimate quantum limit, researchers have proposed schemes based on large photonic Fock states or single chiral photons.¹⁹ However, because large Fock states with photon-number-resolving detection capabilities are not yet available with current technologies, chiral single photons with single-photon detection are a promising route to pursue the limit. By spreading a thin layer of dilute quantum emitters (e.g., perovskite quantum dots) over nanosphere surfaces, the emitted ensemble of chiral photons from the nanosphere can be transformed into a stream of single chiral photons with the same chirality after going through the epilayer (Figure 2a).

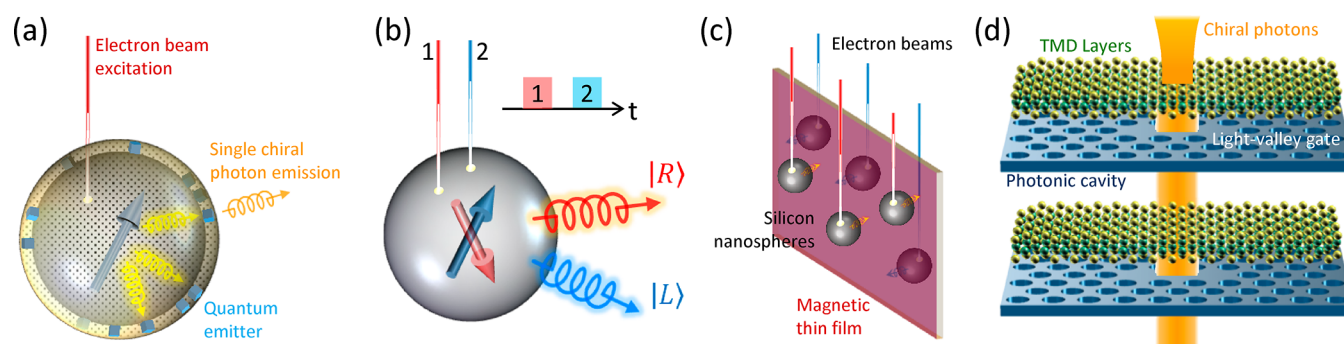


Figure 2. (a) Schematic for *in situ* chiral photon generations using the core–shell structure. A thin layer of diluted perovskite quantum dots spread over the surface of Si nanospheres can absorb the photons emitted by the nanosphere under electron beam excitation and scatter single chiral photons from trion states for quantum information applications. (b) Schematic for ultrafast chirality modulation of generated photons. By quickly adjusting the excitation beam positions on the nanosphere (e.g., from location 1 to location 2), the emitted photons can possess opposite chiralities as a result of electric dipole tailoring. (c) Schematic of all-optical control of magnetism in a magnetic thin film using Si nanospheres. The circularly polarized light generated by the nearby nanospheres can realign local magnetic moments *via* optothermal and optomagnetic coupling in the system. (d) Light–valley gate based on layered transition metal dichalcogenides (TMDs) placed on a photonic crystal cavity. The strong coupling between the valley-tagged transition and the cavity mode enables polarization modulation of the incoming photons conditional on the valley polarization in TMD layers. This sequential photonic coupling promises establishing valley entanglement among multiple TMD layers by switching photons.

Using the same scheme, the information carried by the classical chiral light generated by the electron beam excitation of the Si nanosphere can be transferred to the single photons emitted by the quantum dots atop the sphere.

Chiral Light Modulation. With electron beam excitation, the chirality of light can be adjusted *via* steering the electron beam on the nanosphere. By judiciously choosing two excitation spots on the nanosphere that correspond to emitting photons in the same direction but with opposite chirality, fast modulation of an electron beam's location enables a rapid switch of luminescence chirality (Figure 2b). The technology of steering electron beams is relatively mature as it has been massively deployed in commercial streak cameras,²⁰ where sub-picosecond time resolution is routinely obtained *via* electric-field sweeping.²¹ Successful modulation of chiral polarization not only enables direct information encoding for communication but also underpins the applications relying on different chiral light as described below.

Optomagnetism. Recent studies on ultrafast switching of magnetization in magnetic materials have unveiled all-optical approaches to manipulate the local magnetic moment using circularly polarized pulses, with switching times typically 3 orders of magnitude shorter than those achieved with conventional methods (e.g., using magnetic-field pulses or spin-polarized current).^{22,23} Physically, this process involves fast demagnetization of local electrons *via* the thermal effect on picosecond time scales followed by realignment of magnetic moments along the local magnetic field induced by optothermal and optomagnetic coupling on femto- and picosecond time scales.²⁴ By spreading a layer of nanospheres over the two surfaces of ferromagnetic thin films, one can control its magnetization using the electron beam excitation (Figure 2c). Specifically, depending on which side of the thin film is exposed to the electron beam, the nanospheres can either emit left-handed light (from the front surface) or right-handed light (from the opposite surface) toward the magnetic thin films and induce all-optical ultrafast switching of the magnetic moments within the thin film.

Valleytronics and Spintronics. Since the first demonstration of the use of circularly polarized femtosecond light

pulses to control the valley degree of freedom selectively in monolayer WSe₂,^{25–27} these momentum- and spin-locked valleys of 2D semiconductors have opened up new frontiers for quantum information processing based on the momentum state of electrons, holes, or excitons. Similar to spintronics, where information is encoded in quantum spin number, valleytronics encodes information on quantum valley number, which is intrinsically insensitive to the magnetic field fluctuations in the environment. Chiral light can be used for the initialization, control, and detection of the valley degree of freedom. Currently, the light–valley interaction works in the weak coupling regime; with the assistance of optical cavity, it is possible to achieve strong coupling between a chiral single photon and the valley degree of freedom. Similar to the photonic gates demonstrated on single atoms²⁸ and semiconductor quantum dots,²⁹ the valley–light gate makes it possible to achieve the entanglement of the valley degree of freedom using a chiral photon as a coupling bus (Figure 2d).

Optical spin polarization enabled by the chirality transfer from the organic chiral ligands to the perovskite layers³⁰ paints an alternative picture of using Si nanospheres to inject spins indirectly into semiconductors for spintronics applications. The scheme is similar to Figure 2c but with the magnetic thin film replaced by reduced-dimensional chiral perovskite. The unpolarized electron beams excite the nanospheres to release chiral photons whose angular momentum can be transferred to the electron spins in the perovskite upon the absorption *via* chiral ligands.

In closing, chiral interfaces provide a powerful platform to achieve full control over light–matter interactions. Non-reciprocal propagation light paths resulting from chiral interactions add interesting dimensions to explore in research ranging from quantum sensing to quantum communication, quantum optics, and condensed matter physics.

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<https://pubs.acs.org/10.1021/acsnano.0c10420>

Notes

The authors declare no competing financial interest.

ACKNOWLEDGMENTS

We acknowledge the financial support from the Singapore National Research Foundation through its Competitive Research Program (CRP Award Nos. NRF-CRP21-2018-0007, NRF-CRP22-2019-0004, and NRF-CRP23-2019-0002), Singapore Ministry of Education (MOE2016-T2-2-077, MOE2016-T2-1-163, and MOE2016-T3-1-006 (S)), and the A*Star QTE programme.

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