Room-temperature single-photon sources with definite circular and linear polarizations based on single-emitter fluorescence in liquid crystal hosts

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Abstract. Definite circular and linear polarizations of room-temperature single-photon sources, which can serve as polarization bases for quantum key distribution, are produced by doping planar-aligned liquid crystal hosts with single fluorescence emitters. Chiral 1-D photonic bandgap microcavities for a single handedness of circularly polarized light were prepared from both monomeric and oligomeric cholesteric liquid crystals. Fluorescent emitters, such as nanocrystal quantum dots, nitrogen vacancy color centers in nanodiamonds, and rare-earth ions in nanocrystals, were doped into these microcavity structures and used to produce circularly polarized fluorescence of definite handedness. Additionally, we observed circularly polarized resonances in the spectrum of nanocrystal quantum dot fluorescence at the edge of the cholesteric microcavity’s photonic stopband. For this polarization we obtained a ~4.9 enhancement of intensity compared to the polarization of the opposite handedness that propagates without photonic bandgap microcavity effects. Such a resonance is indicative of coupling of quantum dot fluorescence to the cholesteric microcavity mode. We have also used planar-aligned nematic liquid crystal hosts to align DiI dye molecules doped into the host, thereby providing a single-photon source of linear polarization of definite direction. Antibunching is demonstrated for fluorescence of nanocrystal quantum dots, nitrogen vacancy color centers, and dye molecules in these liquid crystal structures.

1. Introduction

Single-photon sources are distinct from faint light sources in that all emitted photons are separated in time (photon antibunching). In the case of photon antibunching, the second-order correlation function \( g^{(2)}(0) < 1 \), where \( g^{(2)}(\tau) \) is defined at time \( \tau \) as [1]

\[
g^{(2)}(\tau) = \frac{\langle I(t)I(t+\tau) \rangle}{\langle I(t) \rangle^2}.
\]  

(1)

In equation (1), \( I(t) \) is the light intensity and, for the practical case of stationary fields, averaging is performed over time. In the ideal case for antibunching, \( g^{(2)}(0) = 0 \). For laser light, \( g^{(2)}(0) = 1 \), and for thermal light, \( g^{(2)}(0) = 2 \).
Single-photon sources are of interest for applications in the field of quantum communication and quantum metrology. A particularly intriguing usage for single photons is as vectors of quantum information in quantum key distribution (QKD) schemes [2-6]. QKD allows for the secure distribution of randomly generated cryptographic keys between two parties, whereby any passive eavesdropper performing measurements on quantum states transmitted via a QKD scheme will alter the state in a manner detectable by legitimate parties [7-10]. Many QKD schemes specifically require usage of single photons to transmit information, as multi-photon packets can be split, potentially allowing eavesdroppers to siphon off information about the key without risking detection [11,12].

Approaches to generating on-demand single photons usually rely on individual fluorescent emitters, which will emit a single photon at a time when excited. Major challenges to this approach include practicality of implementation, as many single photon emitters require cryogenic temperatures. Another issue of note is that many QKD approaches encode information via photon polarization to generate a key. A quantum cryptography system based on an unpolarized single-photon source, however, uses polarizers to encode information and therefore has a maximum efficiency of 50%. Thus, an ideal single-photon source would emit photons on-demand, with high efficiency and definite polarization, and would operate at room temperature.

This paper concerns our efforts in producing room-temperature single-photon sources that emit photons of definite polarization [13-20]. We have achieved this by fabricating liquid-crystal planar-aligned structures doped with single emitters, including nanocrystal quantum dots, fluorescent molecules, and color centers in nanodiamonds. Cholesteric liquid crystal chiral 1-D photonic bandgap structures were used to create circularly polarized single photons. Nematic liquid crystals were used to align the dipole moment of emitters, allowing for definite linear polarization of emitted photons. It should be noted that single-photon sources with definite linear polarization have also been obtained using heterostructured quantum dots at cryogenic temperatures in elliptical-shape microcavities by the authors of Refs [21-25].

We have also observed coupling of nanocrystal quantum dot fluorescence to the modes of a chiral photonic bandgap microcavity made from an aligned cholesteric liquid crystal structure. Due to the Purcell effect [26], coupling of emitted light to the microcavity causes enhanced rates of spontaneous emission at the edge of the photonic stopband [18]. Planar-aligned cholesteric liquid crystal microcavities have previously been doped with dye molecules (1-3% concentration by weight) for obtaining lasing in cholesterics at the bandedge of a photonic stopband [27-30]. Indeed, a great deal of research has been devoted to the control of spontaneous emission using a variety of photonic crystal architectures, see, e.g., Refs [24,31-35]. Spontaneous emission enhancement is relevant to QKD as enhancing the rate of single photon emission might potentially be used to increase the bit rate of a practical QKD implementation.

2. Experimental Setup

Fluorescence antibunching proves the single-photon nature of the source. Measuring the time interval between two consecutive photons and building the histogram of coincidence counts \( c(\tau) \) allows us to measure \( g^{(2)}(\tau) \), which is proportional to \( c(\tau) \) for sufficiently small count rates and short interphoton times [36,37]. Such measurements are carried out using a Hanbury Brown and Twiss (HBT) intensity interferometer, which consists of a beamsplitter and two single-photon detectors.

For examining our samples, we used two confocal fluorescence microscopes (see Figure 1) equipped with HBT interferometers: a homemade microscope and a Witec alpha-SNOM in a confocal mode. The homemade confocal microscope had an oil-immersion, 1.35 NA objective to focus laser light into the sample, with more detailed schematics given in Figure 2. Information on the Witec alpha-SNOM can be found at Witec’s website (see Ref [38]).
Figure 1. Experimental setups. Left – homemade confocal fluorescence microscope. Right – Witec alpha-SNOM microscope.

Figure 2. Schematics of experimental setup with a homemade confocal microscope. Top – single-emitter confocal fluorescence imaging and photon antibunching measurements. Bottom – spectral and polarization measurements. L1-L6, lenses; BS, beamsplitter; SPAD, single-photon counting avalanche photodiode modules; $\lambda/4$, achromatic quarter wave plate; LP, linear polarizer, $M_2$, mirror on a translation stage; ND filters, neutral density filters.
Single emitters were excited using either a cw, 633 nm HeNe laser, a pulsed, 532 nm Nd:Vanadate laser (76 MHz repetition rate, 6 ps pulse duration), or a cw, 976 nm diode laser depending on the emitter being examined. Sample fluorescence was collected by the same microscope objective, with any remaining excitation light filtered out using a dichroic mirror and interference filters. The fluorescence light could then be directed to one of two microscope ports, which were connected to diagnostics such as:

- An HBT interferometer, utilizing two SPCM-AQR-14 single-photon counting avalanche photodiode modules (SPADs), from Perkin Elmer. SPADs were connected to a TimeHarp 200 time-correlated single-photon counting PCI board (PicoQuant). We used this setup to image the fluorescence of several single emitters, then focused the laser light on one of these single emitters and characterized the fluorescence (photon antibunching or fluorescence lifetime measurements). See Figure 2, top.

- A Princeton Instruments Acton SP2150i spectrometer, used in conjunction with an iXon DV 887 electron multiplying (EM) CCD camera from Andor Technology. Collected fluorescence could be sent through an achromatic quarter waveplate and a linear polarizer, allowing for distinction between right-handed circularly polarized (RHCP) and left-handed circularly polarized (LHCP) fluorescence. In some measurements, we used an Ocean Optics spectrometer with a long acquisition time. See Figure 2, bottom.

3. Single-Photon Sources with Circular Polarization

3.1. Monomeric Cholesteric Liquid Crystals

We used planar-aligned cholesteric liquid crystal (CLC) to fabricate 1-D chiral photonic bandgap microcavities. CLC consists of rod-shaped chiral anisotropic molecules that form periodic helical structures of pitch \( p \) when planar-aligned [39]. In CLC chiral photonic bandgap structures, circularly polarized light of handedness with electric-field vector following the rotation of the liquid crystal molecules is reflected within a spectral band centered at \( \lambda_c = n_{av}p \), with bandwidth of roughly \( \Delta \lambda = \lambda_c \Delta n/n_{av} \). The variables \( n_{av} \) and \( \Delta n \) are defined from the extraordinary index of refraction, \( n_e \), and the ordinary index of refraction, \( n_o \), where \( n_{av} = (n_o + n_e)/2 \) and \( \Delta n = n_e - n_o \). Light with polarization of opposite handedness is unaffected by the structure and propagates without any changes. When doped with single emitters, such chiral photonic bandgap structures allow for the creation of single-photon sources with a high purity of circular polarization, with the potential for resonance effects of the microcavity being used to enhance the rate of single photon emission.

To create CLC microcavities, we used a mixture of E7 nematic liquid crystal and chiral additive CB15 [16]. When using this liquid crystal mixture to produce a photonic bandgap structure, the stopband center wavelength was tuned by the ratio of CB15 to E7, with a CB15 concentration by weight \( c_m \) being related to the stopband position by \( c_m = n_{av}/(\lambda_c \times HTP) \), where the chiral twisting power of CB15 HTP is equal to \( \sim 7.3 \mu m^{-1} \). Thus it was possible to tune the resulting bandgap based on the relative concentrations of these compounds, allowing us to create photonic bandgap structures with predetermined stopband features. Figure 3 provides examples of the selective transmission curves of prepared CLC hosts, with quantum dot fluorescence spectral curves superimposed, demonstrating the viability of this approach in creating chiral microcavity structures tuned to the fluorescence maxima of chosen emitters.

Once a CLC mixture was created, the liquid crystal was mixed with a solution containing \( \sim nM \) concentrations of quantum dots, and the solvent was evaporated. The resulting monomeric CLC doped with quantum dots was placed between two glass coverslips and planar-aligned via a mechanical shearing motion of the coverslips (for further details see Ref [14]). This relatively simple process created a chiral microcavity doped with single emitters.
Selective transmission curves of right-handed circularly polarized light through chiral photonic bandgap structures formed from planar-aligned monomeric CLC hosts (dashed curves), with different concentrations $c_m$ (by weight) of CB15 in E7 adjusted to give different stopband positions. Superimposed on these curves are the fluorescence spectra (solid lines) of the nanocrystal quantum dots that these transmission curves were tuned to. Left – $c_m = 36.6\%$, CdSe quantum dots, maximum fluorescence wavelength $\lambda_0 = 580$ nm; Right – curve 1 corresponds to $c_m = 36.0\%$ and curve 2 corresponds to $c_m = 29\%$, CdSeTe quantum dots, maximum fluorescence wavelength $\lambda_0 = 700$ nm.

Figure 4, left, shows the emission spectra of CdSe quantum dots in one of these chiral microcavities, with curve 1 (black) showing RHCP fluorescence and curve 2 (red) showing LHCP fluorescence. The degree of circular polarization is measured by the circular polarization dissymmetry factor [40]

$$g_e = \frac{2(I_L - I_R)}{(I_L + I_R)},$$

where $I_L$ and $I_R$ are the intensities of left and right-handed circularly polarized light respectively. The observed dissymmetry factor was $g_e = -1.6$ at 580 nm. For unpolarized light, $g_e = 0$.

Our HBT interferometer was used for examining the photon statistics of these samples. Detector coincidence counts were histogrammed based on the time delay between photon detection events.
(interphoton times), with \( g^{(2)}(\tau) \) found by normalizing these coincidence histograms [36,37]. Figure 5 presents the \( g^{(2)}(\tau) \) histograms we obtained from our microcavities under pulsed, 532 nm excitation.

**Figure 5.** Histograms showing the second-order correlation function \( g^{(2)}(\tau) \) (obtained by normalization of coincidence counts) versus interphoton times found from the fluorescence of quantum dots in a CLC host under pulsed, 532 nm excitation. The dip at the zero interphoton time indicates photon antibunching. Left – Antibunching from CdSe quantum dots from Figure 3, left. Right – Antibunching from CdSeTe quantum dots from Figure 3 (right, curve 1) with superimposed red dashed line showing fit of form \( g^{(2)}(\tau) = 1 - \frac{1}{N} e^{(\tau/\tau_0)} \) (see explanation in the text).

Figure 5, left, shows the \( g^{(2)}(\tau) \) histogram from the fluorescence of the CdSe quantum dots from Figure 3, left. The peaked structure occurs due to the pulsed excitation. A distinctive dip in the height of the peak at the zero interphoton time is clearly present, indicating photon antibunching and corresponding to a value of \( g^{(2)}(0) = 0.76 \pm 0.04 \). Such a large value of \( g^{(2)}(0) \) can be explained by the background from the liquid crystal host, located at the same spectral region as the quantum dot fluorescence, with the spectrum of the raw liquid crystal host shown in Figure 4, right.

Figure 5, right, in turn, presents the \( g^{(2)}(\tau) \) histogram from the fluorescence of the CdSeTe quantum dots from Figure 3, right, with the sample’s selective transmission presented by curve 1, where the quantum dot fluorescence spectrum is outside the range of liquid crystal background. While a pulsed, 532 nm laser is still used to excite this sample, the peaked structure seen in Figure 5, left, is not present in Figure 5, right, due to these CdSeTe quantum dots having fluorescence lifetimes larger than the time between two laser pulses (a 76 MHz pulse repetition rate corresponds to \( \sim 13 \) ns between pulses). A clear dip at the zero interphoton time is present, regardless, indicating antibunching. A fit of form [41] \( g^{(2)}(\tau) = 1 - \frac{1}{N} e^{(\tau/\tau_0)} \) was found for this histogram, where \( \tau_0 = 1 / (k_{12} + k_{21}) \), \( k_{12} \) is the excitation pump rate, \( k_{21} \) is the spontaneous decay rate, and \( N \) is the number of emitters producing the fluorescence spot under examination (at \( N = 1, g^{(2)}(0) = 0 \), at \( N = 2, g^{(2)}(0) = 0.5 \)). This fit is shown by the red dashed curve in Figure 5, right, and yielded a \( g^{(2)}(0) \) value of 0.001 ± 0.034 and a decay time constant of \( \tau_0 \sim 15 \) ns.

### 3.2. Glassy Cholesteric Liquid Crystal Oligomers: Microcavity Resonances

We used left-handed cyclosiloxane oligomeric CLC powder from Wacker Chemie [42,43] and produced a planar-aligned glassy CLC structure doped with CdSeTe quantum dots (Qdot 800 ITK organic, Invitrogen, fluorescence maximum at 790 nm) [18]. Doping was accomplished by heating the CLC to \( \sim 135^\circ \) C (the oligomer’s melting temperature) and then mixing the melted CLC with quantum dots dispersed in toluene at a concentration of \( \sim 1 \) µM, with heating allowed to continue until the toluene evaporated. Subsequently, cells were prepared using two polyimide buffed glass coverslips.
The CLC doped with quantum dots was placed on a buffed coverslip and heated beyond the oligomer clearing temperature of 180°C. After the sample was cooled to ~135°C, the second buffed coverslip was placed on the first and sheared along the direction of polyimide buffing. A slow cooling process back to a glassy (solid) state preserved the CLC order. This resulted in a photonic bandgap microcavity with a center wavelength of 910 nm.

Prepared samples were analyzed using our homemade confocal microscope setup. We excited the sample with cw, 633 nm laser light from a HeNe laser and observed the spectrum of the sample’s fluorescence. By placing an achromatic quarter waveplate and linear polarizer in front of the spectrometer, we were able to filter for different handedness of circularly polarized fluorescence.

The resulting fluorescence spectra can be seen in Figure 6. LHCP light experienced the photonic bandgap and therefore the black LHCP curve in Figure 6 (curve 1) shows microcavity resonance, indicating that the LHCP light coupled to the cavity mode. The LHCP fluorescence had a center wavelength of 833 nm and a FWHM of 16 nm ($Q \sim 50$), as compared to a FWHM of 76 nm for RHCP fluorescence. The center wavelength of this resonance roughly matches the edge of the photonic stopband, centered at 910 nm, and shown in Figure 6 by the blue curve 3. The observed RHCP fluorescence is shown in red in Figure 6 (curve 2) and was less intense due to not experiencing the CLC microcavity, showing no sign of line narrowing. The maximum intensity ratio between LHCP and RHCP was a factor of 4.9, and the circular polarization dissymmetry factor, from equation (2), was $g_c = 1.3$.

**Figure 6.** Circularly polarized fluorescence spectrum from CdSeTe quantum dots doped in a planar-aligned glassy cholesteric liquid crystal microcavity. Curve 1 shows LHCP fluorescence and has a resonance at 833 nm. Curve 2 shows RHCP fluorescence from the same source. Curve 3 shows the selective transmission of LHCP light through the aligned liquid crystal host, showing a stopband centered at 910 nm. Inset: Dependence of resonance peak intensity on the rotation of a linear polarizer (LP) after a fixed quarter-wave plate.

Figure 7, left, shows a confocal fluorescence raster scan taken of a sample prepared using a relatively low concentration of quantum dots dispersed in toluene (~10 nM), with the higher intensity spots indicating the location of fluorescing quantum dots in the glassy CLC microcavity. Focusing on a spot (circled in white on Figure 7, right), we checked for photon antibunching and obtained the coincidence histogram shown in Figure 7, right.

Figure 7, right, displays the histogram of coincidence counts $c(t)$ in blue, with $g^{(2)}(r)$ derived by normalizing $c(t)$. The measured $g^{(2)}(0)$ value from the fit shown in Figure 7, right (green, solid curve) is $g^{(2)}(0) = 0.382 \pm 0.037$. As $g^{(2)}(0) < 0.5$, this indicated that we have managed to isolate the
fluorescence of a single quantum dot, serving as a source of antibunched light in a glassy CLC microcavity.

**Figure 7.** Left – Confocal fluorescence microscopy scan of CdSeTe quantum dots doped in planar-aligned glassy oligomeric cholesteric liquid crystal. Right – Raw coincidence counts $c(\tau)$ (right axis) of fluorescence from the circled quantum dot in the confocal scan on left, as measured by an HBT interferometer. The second-order correlation function, $g^{(2)}(\tau)$ (left axis) is found by normalizing $c(\tau)$ and indicates antibunched fluorescence.

3.3. Rare-Earth-Doped Nanocrystals and Color Centers in Nanodiamonds

Additional work with monomeric CLC based microcavities included doping with emitters such as nitrogen vacancy color centers in nanodiamonds and rare-earth ions in nanocrystals. These alternative emitters can provide a source of single photons that is less likely to bleach.

Figure 8, left, shows antibunching of nitrogen vacancy color centers in nanodiamonds doped in a CLC (E7 and CB15) microcavity with a stopband centered at 725 nm, under cw, 514 nm excitation. The $g^{(2)}(0)$ value determined via fit (shown in Figure 8, left, as the dashed red line) was $g^{(2)}(0) = 0.74 \pm 0.08$. The fluorescence spectrum of nitrogen vacancy centers in nanodiamonds doped in this monomeric CLC when excited using cw, 514 nm excitation from an argon ion laser is shown in Figure 8, right.

**Figure 8.** Left – Photon antibunching observed in fluorescence from nitrogen vacancy centers in a CLC host. Black curve shows the raw data. Red dashed curve shows the fit. Right – Fluorescence spectrum of nitrogen vacancy centers in nanodiamonds doped in CLC host when excited by cw, 514 nm laser light.

A monomeric CLC microcavity made from a mixture of E7 and CB 15 was also doped with rare-earth Er$^{3+}$ and Yb$^{3+}$ ions in 20 nm to 30 nm sized NaYF$_4$ nanocrystals with 20% Yb and 2% Er. When
these ions were excited using a cw, 976 nm diode laser at incident powers of ~500 µW, we were able to observe upconverted fluorescence of Er³⁺, as shown in Figure 9. The emission lines observed were attributed to the transitions ⁴H₁₁/₂, ⁴S₅/₂ → ⁴I₁₅/₂ (green) and ⁴F₉/₂ → ⁴I₁₅/₂ (red) of the Er³⁺ ions [44]. The populations of upper levels in Er³⁺ occur due to an efficient energy transfer from the Yb³⁺ to the Er³⁺. This fluorescence was measured to have a circular polarization dissymmetry factor of gₑ = -0.77 at 680 nm. These nanocrystals doped with rare-earth ions were prepared at the University at Buffalo (The Institute for Lasers, Photonics and Biophotonics).

Figure 9. Red solid lines show the fluorescence spectrum of Er³⁺ ions doped in NaYF₄ nanocrystals dispersed in a chiral microcavity formed from planar-aligned monomeric CLC (E7 and CB15). The blue dashed curve shows the spectral transmission of the doped microcavity, measured using unpolarized light.

4. Single-Photon Sources with Linear Polarization

Another intriguing application of liquid crystal as a host for single emitters is using the planar-alignment of nematic liquid crystals to provide doped molecular dipoles definite alignment along a preferred direction for efficient excitation [15]. This allows for the creation of a single-photon source with fluorescence of definite linear polarization.

Samples were prepared using DiIC₁₈(3) dye (DiI) molecules from Molecular Probes doped in a glassy nematic liquid crystal oligomer synthesized by S. H. Chen’s group of University of Rochester [15]. Planar-aligned films of this doped liquid crystal host of ~100 nm thickness were prepared using photoalignment.

Photoalignment was performed by first spin-coating a Staralign-2100 linearly photopolymerizable polymer (Rolic Technologies Ltd.) to a cleaned cover glass slip, which was then cured at ~135° C. This film was then irradiated by a polarized UV light for 10 to 15 minutes, with further irradiation used to bleach the polymer impurity fluorescence.

Figure 10. Confocal fluorescence microscopy scans of DiI dye molecules in a planar-aligned nematic liquid crystal host. Left – Fluorescence with polarization perpendicular to the alignment direction of the host. Right – Fluorescence with polarization parallel to the alignment direction of the host.
An oligomer solution doped with dye and diluted in chloroform was subsequently spin-coated onto these Staralign coated glass slips. After the chloroform evaporated, we heated the sample to ~80°C, slightly above where the oligomer transition to a nematic state occurs, after which the sample was slowly cooled to a glassy state, preserving the planar-aligned nematic order.

To characterize prepared samples, we used the Witec alpha-SNOM (see Figure 1, right) in a confocal mode, exciting the sample using a Nd:YAG laser for cw, 532 nm irradiation. Single-photon counting avalanche photodiodes were used as photodetectors for confocal fluorescence scans, with a polarizing beamsplitter used so that each photodetector collected light of orthogonal polarization. Figure 10 shows confocal fluorescence microscopy images taken of a planar-aligned sample of glassy nematic liquid crystal doped with DiI dye, with one photodetector used to collect fluorescence polarized parallel to the sample alignment direction, while the other photodetector collected fluorescence polarized perpendicular to the sample alignment direction.

Molecules were identified by the peak pixel intensity values from Figure 10 and a linear polarization measure $\rho$ was determined for each molecule by comparison of the perpendicular and parallel polarization images. Our measure of linear polarization was

$$\rho = \frac{I_{\text{par}} - I_{\text{perp}}}{I_{\text{par}} + I_{\text{perp}}},$$

where $I_{\text{par}}$ and $I_{\text{perp}}$ are, respectively, the fluorescence intensities parallel and perpendicular to the direction of alignment. The only difference of $\rho$ with the degree of polarization [45] is that it allows inclusion of the direction of polarization as part of the measure, depending on whether $\rho$ is positive or negative.

The different $\rho$ values that we found are histogrammed in Figure 11. A clear asymmetry in Figure 11 demonstrates a preference towards fluorescence polarized perpendicular to sample alignment. This is in contrast to the expected fluorescence from an unoriented sample, which would yield a symmetric $\rho$ histogram [46].

![Figure 11](image)

**Figure 11.** Histogram of polarization measure $\rho$ from the fluorescence of 38 different DiI dye molecules in a planar-aligned nematic liquid crystal host.

That DiI molecules doped in this planar-aligned liquid crystal host tend to fluoresce with polarization perpendicular to the alignment of the liquid crystal can be explained by the molecular structure of these molecules. As illustrated in Figure 12, it is likely that two alkyl chains orient themselves parallel to the rod-like molecules of the nematic liquid crystal host. The absorbing and emitting dipoles, however, are parallel to the bridge between these alkyl chains. Therefore, these dipoles end up oriented perpendicular to the direction of liquid crystal alignment, hence the fluorescence having a polarization orthogonal to the alignment of the liquid crystal.
Figure 12. Schematic view of a DiI dye molecule in a nematic liquid crystal host. The long axes (alkyl chains) of the DiI molecules tend to orient themselves along the rod-like nematic liquid crystal molecules, while the dipole, which is parallel to the bridge between the alkyl chains, orients perpendicular to the direction of liquid crystal alignment.

Figure 13. Polarized fluorescence of DiI dye in planar-aligned nematic liquid crystal hosts. Samples were excited using a 532 nm laser. Left – Intensity of DiI fluorescence in E7 as linear polarization of exciting light was rotated over 360°, with red dots showing experimental measurements and the blue solid curve providing a sinusoidal fit. The zero angle of polarization corresponds to the direction perpendicular to liquid crystal alignment. Center – Red (curve 1) and black (curve 2) show fluorescence spectrum with polarization perpendicular and parallel to the host alignment direction respectively. Right – Antibunching histogram taken from the fluorescence of a single molecule of DiI dye in an E7 nematic host under pulsed, 532 nm excitation.

To confirm these results, additional experiments were carried out both with monomeric nematic liquid crystal and nematic glassy liquid crystal oligomers. Figure 13, left, shows intensity changes of fluorescence of DiI molecules in a planar-aligned monomeric nematic liquid crystal (E7), as the linear polarization of the exciting 532 nm beam was rotated over 360° degrees. Figure 13, left, shows a clear dependence of the intensity on the exciting angle of polarization, where the maximum fluorescence occurred when the DiI molecules were excited by light with linear polarization perpendicular to the alignment of the liquid crystal.
Figure 13, center, shows spectrofluorimeter measurements made for polarization perpendicular and parallel to the sample alignment, this time having used a sample of planar-aligned glassy nematic liquid crystal oligomer doped with DiI molecules of more than 1% concentration by weight. The value of $\rho$ was measured from Figure 13, center, by comparing the peak intensities of the curves, yielding $\rho = -0.5$. The results of Figure 13, left and center, indicated a clear preference for fluorescence with definite linear polarization perpendicular to the sample alignment.

Figure 13, right, shows an antibunching histogram for a single molecule of DiI dye doped in an E7 nematic host, taken using pulsed, 532 nm excitation [47]. The value for $g^{(2)}(0)$ is $g^{(2)}(0) = 0.77 \pm 0.10$. This relatively high value of $g^{(2)}(0)$ may be due to the fact that the Raman spectrum of the E7 overlaps with the fluorescence spectrum of DiI dye.

5. Conclusion
We have employed liquid crystals as the hosts for single emitters to create room-temperature single-photon sources that emit photons of definite circular and linear polarization. Circularly polarized light of definite handedness was observed from the fluorescence of nanocrystal quantum dots, nitrogen vacancy centers in nanodiamonds, and rare-earth ions in nanocrystals doped in a photonic bandgap cholesteric liquid crystal microcavity. We also observed circularly polarized resonance from quantum dot fluorescence in a cholesteric microcavity. The resonance was located at 833 nm, at the edge of a stopband centered at 910 nm, and had a FWHM of 16 nm as compared to a FWHM of 76 nm for fluorescence with polarization of opposite handedness, demonstrating coupling of left-handed circularly polarized light to a microcavity mode. Circularly polarized fluorescence from the microcavity had a circular polarization dissymmetry factor of 1.3 and an intensity ratio between left and right hand circularly polarized light of 4.9 at the resonant wavelength. Antibunching was demonstrated in this architecture, showing the feasibility of this approach for creating circularly polarized single-photon sources. Additional work was done on the usage of nematic liquid crystal for aligning the orientation of DiI dye molecules, providing a source of photons of definite linear polarization.

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