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Room temperature source of single photons of definite polarization

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A definite polarization in fluorescence from single emitters (dye molecules) at room temperature is demonstrated. A planar-aligned, nematic liquid-crystal host provides definite alignment of single dye molecules in a preferred direction. Well-defined polarized fluorescence from single emitters (single photon source) is important for applications in photonic quantum information. Polarized single-photon sources based on single emitters, for example, are key hardware elements both for absolutely secure quantum communication and quantum computation systems.

1. Introduction

The fields of quantum communication and quantum computing need a simple reliable source of single photons propagating in a given direction with a definite polarization in order to function efficiently. It is important that within a specified time interval there be one and only one photon in the given direction and polarization. Weak attenuated sources are not satisfactory because in order to attenuate the source sufficiently so that two simultaneous photons are very unlikely the probability of no photons at all becomes large. Similarly, if the single photon is radiated into a large solid angle it is quite difficult to collect it into the optical system with good efficiency. If the photon has unknown polarization, then filtering it through a polarizer to produce the desired polarization will cause a loss of a large percentage of the photons so that again we have zero photons much of the time. It is a difficult engineering problem to produce the ideal source.

We demonstrate in this paper an important step along the way to developing such a source. We place isolated dye molecules into a liquid crystal matrix that has several desirable properties as an environment for the molecules. The molecules

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themselves have a very large absorption cross-section for laser radiation and then once excited they will emit a photon, Stoke-shifted in frequency by tens of nanometres from the exciting pulse. The lifetime of the photon emission is a few nanoseconds. By exciting the molecule with a sub-nanosecond pulse of sufficient energy we can guarantee that, with good probability, the single dye molecule will emit a single photon in a few-nanosecond time window after the excitation. The liquid crystals serve two purposes. Nematic liquid crystals align the dye molecules along a definite axis. Such aligned molecules would normally emit in a dipole pattern which is a big improvement over the normal situation in which the molecular axes and thus the dipole patterns are randomly oriented in space. The chiral nematic (cholesteric) liquid crystals further improve the situation by providing a photonic crystal environment into which the molecules will emit. This environment is provided by supplying a torque to the medium as it is being laid down. The liquid crystals are dichroic, and this torque causes the principal axis of the index of refraction to rotate periodically about an axis perpendicular to the thin layer sample. By making the pitch angle of the rotation of the order of a wavelength of the emitted photon we can form a photonic crystal that will modify the radiation pattern of the molecule. The photon will be preferentially emitted along the rotation axis. These 1-D photonic band-gap structures in cholesteric liquid crystals (figure 1) possess two advantages over conventional 1-D photonic crystals:

(1) Because the refractive index n varies gradually rather than abruptly in cholesterics, there are no losses into the waveguide modes, which in the case of conventional 1-D photonic crystals, arise from total internal



Figure 1. Perspective view of the AFM topographical image of a 1-D photonic band-gap planar-aligned, glassy, cholesteric liquid crystal. The full vertical scale corresponds to 30 nm.

reflection at the border between two consecutive layers with a different *n*. These waveguide losses can reach $\sim 20\%$.

(2) High-polarization purity for circular polarization of definite handedness can be reached for single emitters with both polarized and unpolarized emission.

There are various other known methods for the production of single photons at definite time intervals [1–4], which are based on a single atom [5, 6], a single trapped ion [7], a single molecule [8–10], a single colour centre in diamond [11], or the Coulomb-blockade effect in a micropin junction with a quantum well as the active layer [12–14]. Tremendous progress has been made in the realization of single photon sources (SPSs) based on excitonic emission from single heterostructured semiconductor quantum dots excited by pulsed laser light (see reviews in [1] and [3]). In heterostructured-quantum-dot SPSs [15–28], microcavities have been used for spontaneous emission enhancement in the form of a whispering-gallery-mode resonator (turnstile device), 1-D photonic band-gap, three-dimensional pillar microcavity, and 2-D photonic crystals. Definite polarization of single photons from heterostructured quantum dots both in elliptical pillar microcavities [22, 24, 25] and in 2-D photonic crystal [23, 26] was reported for the resonance wavelength. A weakness of heterostructured-quantum-dot SPSs is that they operate only at liquid-helium temperatures. In addition, they are not readily tunable.

To date, three approaches have been suggested for room-temperature SPSs: single molecules [8–10, 29–37], colloidal semiconductor quantum dots (nanocrystals) [38, 39], and colour centres in diamond [11, 40–43]. The colour-centre source suffers from the challenge that it is not easy to couple out the photons and that the spectral bandwidth of the light is typically quite large (\sim 120 nm).

Both single molecules and colloidal semiconductor nanocrystals dissolved in a proper solvent can be embedded in photonic crystals to circumvent the deficiencies that plague the other system. Colloidal semiconductor nanocrystals dissolved in polymethylmethacrylate were placed inside a 2-D photonic crystal cavity [44]. The nanocrystal emission at room temperature mapped out the cavity resonances and was enhanced relative to the bulk emission. A planar cavity was recently used to control the single-dye molecule fluorescence spectra and decay rate [45]. The primary problems with using fluorescent dyes and colloidal semiconductor nanocrystals in cavities are the emitters' bleaching and blinking, nontunability of the source, and random polarization of photons.

We are working on a room-temperature alternative to cryogenic SPSs based on semiconductor heterostructures using single emitters in specially prepared liquid crystal hosts, which can exist both as monomers (fluid media) and oligomers or polymers. In addition to emitter alignment and self-assembled structures with photonic band-gap properties, such a host with special treatment (oxygen depletion) can protect the emitters from bleaching. In [35–37], we reported on a significant diminishing of dye bleaching by saturation of liquid crystals with helium. In that work molecules did not bleach for periods of more than one hour under continuous wave (cw) excitation [35–37]. (The first impressive experiments on avoiding dye bleaching in oxygen-depleted hosts were reported in [9] and [30].



Figure 2. Near-field optical image of a 2-D hexagonal self-assembly of glassy cholesteric liquid crystal ($5 \mu m \times 5 \mu m$ scan).

In [9], single terrylene-dye molecules in a p-terphenyl molecular crystal host did not bleach during several hours of pulsed, several-megahertz, pulse-repetition-rate excitation.)

Recently we reported the first demonstration of dye-fluorescence antibunching in liquid crystal hosts [35–37] that is evidence of the single-photon nature of the source. We prepared 1-D photonic crystals (figure 1) and 2-D hexagonal structures (figure 2) in cholesteric liquid crystals as well [35–37, 46].

Recent advances in liquid crystal technology, especially in the fabrication of electric-field/temperature-controlled 1-D, 2-D, 3-D photonic crystal structures and the infiltration of photonic crystals with liquid crystals, can be used in SPS preparation with properties that other SPS methods failed to provide, for instance tunability of the source.

This paper highlights another advantage of liquid crystal hosts – definite polarization of fluorescence from single dye molecules [47–49] at room temperature. Single molecules of $\text{DiIC}_{18}(3)$ dye were embedded in a planar-aligned, glassy, nematic liquid crystal host and were aligned by liquid crystal molecules. It should be noted that each molecule of $\text{DiIC}_{18}(3)$ dye without liquid crystal host emits polarized light, the polarization direction of which varies from molecule to molecule. But for practically efficient SPS devices it is important to have a well-defined polarization for all molecules of the sample.

In addition to single-molecule experiments, our results with high concentration of emitters reported in this paper also confirm well-defined linear or circular polarization in photoluminescence of the same dye from planar-aligned nematic and cholesteric liquid crystals.



Figure 3. Experimental setup.

2. Experimental setup

Single-molecule fluorescence microscopy was carried out on a Witec alpha-SNOM device in confocal transmission mode (see schematics of this instrument in [50]). Figure 3 shows the abbreviated schematic of our experiment. The dye-doped liquid crystal sample was placed in the focal plane of a 1.4-numerical aperture, oil-immersion microscope objective used in confocal transmission mode (instead of SNOM objective with cantilever). The sample was attached to a piezoelectric, XYZ translation stage. Light emitted by the sample was collected by a confocal setup using a 1.25-numerical aperture, oil-immersion objective, an imaging lens with a focal length 12.5 cm and an aperture in the form of an optical fibre. The cw, spatially filtered (through a single-mode fibre connected directly to the Witec microscope input port), 532 nm, diode-pumped, Nd:YAG laser output excited single molecules. To obtain a diffraction limited spot on the sample, the excitation beam was expanded and collimated by the optical system of the Witec microscope [50]. In-focus, the intensities used were of the order of several kW/cm².

For polarized fluorescence measurements, we placed inside the Witec microscope both a 50/50 polarizing beamsplitter cube (as opposed to our antibunching correlation measurements [35–37], in which a nonpolarizing beamsplitter was used) and the second arm of the confocal detection. The confocal microscope apertures were 100 µm core optical fibres placed in each arm of the beamsplitter's output (figure 3). Both beamsplitter cube and the second arm's fibre were placed on adjustable mounts. Beam alignment into the first arm's fibre was carried out by alignment of the Witec microscope objective using a piezo-drive.

Residual transmitted excitation light was removed by either two consecutive dielectric interference filters or by a combination of a 532 nm notch filter with bandwidth 20 nm and a Schott orange-glass filter, yielding a combined rejection of better than seven orders of magnitude at 532 nm.

Photons in the two arms were detected by identical cooled avalanche photodiode modules (APD) in single-photon counting Geiger mode (Perkin Elmer SPCM AQR-14).



Figure 4. Molecular structure of $DiIC_{18}(3)$ dye.

3. Sample preparation

For these experiments, we used DiIC₁₈(3) dye from Molecular Probes in planaraligned, glassy nematic liquid crystal hosts. The dye molecular structure is presented in figure 4. An oligomer host material was synthesized by Prof. S.H. Chen's group (University of Rochester) [51]. The nematic liquid crystal state of this material, which exists at elevated temperatures, was preserved at room temperature by slowly cooling the liquid crystal to the glassy state with frozen nematic order. We prepared ~100 nm thick films of this glassy, nematic liquid crystal guest–host system by uniaxial photoalignment. We did not use standard rubbing (buffing) alignment of liquid crystals with the polyimide or nylon 6/6 thin films because of the finding that they possess a higher fluorescence count rate than single molecules of fluorescence dyes.

For photoalignment, a Staralign-2100 linearly photopolymerizable polymer (Rolic Technologies Ltd.) was spin coated on a pirhana-solution cleaned cover glass slip, cured at 130°C and irradiated for \sim 10–15 minutes by linearly polarized UV light in the spectral region 300–365 nm (65 W Xenon lamp with filters and UV quartz multistec linear polarizer) at an intensity of \sim 10 mW/cm². This new alignment technique prevents material contamination by particulates, but to bleach its own single-molecule fluorescence, the UV-irradiation time may be increased.

An oligomer solution with a 1% concentration by weight in chloroform with 10^{-8} M concentration of the dye in chloroform was prepared. After spin-coating the dye-doped oligomer solution onto the aligned Staralign-coated slip, the sample was heated on a hotplate above transition to nematic state (~80°C) for 30 minutes and slowly cooled to the glassy state with preserved nematic order. Polarizing microscope images showed that we succeeded in the preparation of planar alignment over areas exceeding 10 mm × 10 mm [47–49]. To remove the oxygen from the



Figure 5. Confocal fluorescence microscopy images of $\text{DiIC}_{18}(3)$ single-molecule fluorescence in a planar-aligned, glassy, nematic liquid crystal host (10 μ m × 10 μ m scan): left – polarization perpendicular to the alignment direction; right – parallel polarization. Scale shows counts/pixel.

nematic layer, which causes bleaching of single-molecule fluorescence, heating and annealing of the sample can be carried out under vacuum or under argon.

4. Experimental results: well-defined single-molecule fluorescence polarization

Figure 5 shows images of single-molecule fluorescence for polarization components perpendicular (left) and parallel (right) to the alignment direction under 532 nm cw excitation. These two polarization components in the sample plane have been separated with a polarizing beamsplitter cube (figure 3). In this host, the fluorescence maximum of $DiIC_{18}(3)$ dye occurs at ~580 nm and its bandwidth is ~50 nm. Note that the images in figure 5 are taken by raster scanning the sample relative to the stationary, focused laser beam. The scan direction is from left to right and, line by line, from top to bottom. The size of the bright features is defined by the point-spread function of the focused laser beam. These images not only contain information about the spatial position of the fluorescent molecules, but also about the changes of their fluorescence in time. Dark horizontal stripes and bright semicircles instead of circles represent the blinking and bleaching of the molecules over time. Blinking and bleaching are a common single-molecule phenomenon and convincing evidence of the single-photon nature of the source. The explanation of the nature of the long-time blinking from milliseconds to several seconds remains a subject of debate in the literature [52]. To reduce bleaching, oxygen needs to be removed from the liquid crystal host which was reported in previous papers [35–37], but was not done in the present experiments.

Figure 5 clearly shows that for this sample, the polarization direction of the fluorescence of single molecules is predominantly in the direction perpendicular to the alignment of liquid crystal molecules. It is important that the background levels of figure 5 (left) and figure 5 (right) are the same (~ 10 counts/pixel

or ~640 counts/s). The maximum pixel intensity (counts/pixel) of single-molecule fluorescence exceeds this background by up to 15 times. The maximum count rate of single-molecule images is approximately 10 kcounts/s (~160 counts/pixel with ~4 s per line scan, 256 pixels per line). Note that the detector dark counts are fewer than 100 counts/s.

The difference in single-molecule fluorescence signal for different molecules can be explained by the difference in their long-lived, triplet-state lifetimes ($\sim 10^{-4}$ s) which depend on environmental conditions, e.g. surrounding oxygen molecules. Fluorescence lifetime of a single dye molecule is $\sim 10^{-9}$ s. Being in the triplet state, molecules do not emit any light.

The polarization anisotropy is defined here [53-55] as $\rho = (I_{par} - I_{perp})/(I_{par} + I_{perp})$, where I_{par} and I_{perp} are fluorescence intensities for polarization components parallel and perpendicular to the alignment direction. Processing the images in figure 5 with background subtraction shows that from a total of 38 molecules, 31 molecules have a negative ρ value (figure 6), three molecules a zero value, and only four molecules have a positive ρ value. Molecules are identified by their maximum count rate per pixel. In figure 6, a highly asymmetrical histogram for ρ is depicted, that differs markedly from both theoretical and experimental histograms of ρ for the same DiIC₁₈(3) dye when randomly oriented (see [53], figure 3(*a*) and (*c*)). For random orientation [53] this histogram is symmetrical: the number of molecules with positive ρ is the same as that with negative ρ in contrast with the histogram in figure 6.

This predominance of 'perpendicular' polarization in figures 5 and 6 can be explained by the $\text{DiIC}_{18}(3)$'s molecular structure (figure 4). The two alkyl chains likely orient themselves parallel to the rod-like liquid crystal molecules, but the emitting/absorbing dipoles that are nearly parallel to the bridge (perpendicular to



Figure 6. The histogram of polarization anisotropy of 38 molecules of $DiIC_{18}(3)$ dye in a planar-aligned, glassy, nematic liquid crystal host.

alkyl chains) will be directed perpendicular to the liquid crystal alignment. $DiIC_{18}(3)$ molecules orient in the same manner in cell membranes [56, 57]. It should be noted that in [58] single uniaxially oriented terrylene dye molecules in tensile deformed polyethylene were imaged at cryogenic temperatures, however, this paper did not provide the results on definitely polarized fluorescence of single molecules.

Seven molecules in figure 5 and figure 6 have either positive or zero anisotropy. These molecules can be either a small amount of impurities in the Staralign photoalignment agent, which have not been bleached even after the UV irradiation of Staralign-coated slips, or impurities of the glassy oligomer host itself. The single-molecule fluorescence microscopy method is very sensitive to such material impurities. We sometimes observed single-molecule fluorescence from the impurities in undoped glassy liquid crystal oligomers even when a chromatographic analysis did not show any [59]. Imperfect alignment of some dye molecules on the defects in glassy liquid crystal can be another reason for the deviation of ρ from a negative value close to -1. Our next step will be the improvement of the proportion of molecules with desired ρ with identification of impurities using a spectrometer.

It should be mentioned that if one calculates the value of ρ from polarized fluorescence data of heterostructured semiconductor quantum dots in elliptical micropillars and 2-D photonic crystals reported in the literature [22–26], this value will be between 0.2 and 0.95.

5. Polarized photoluminescence from glassy liquid crystal with high dye concentration

The same negative sign of the polarization anisotropy was obtained in spectrofluorimeter measurements for a sample with a high (~0.5% by weight) concentration of the same dye in a planar-aligned, glassy, nematic liquid crystal layer that is ~4.1 µm thick (figure 7, left). Planar-aligned orientation of liquid crystal was made by the rubbing technique using a polyimide orientational layer on both substrates.



Figure 7. Spectrofluorimeter measurements of a polarized fluorescence of $\text{DiIC}_{18}(3)$ dye doped in planar-aligned, glassy, liquid crystal hosts under excitation with a nonpolarized, 532 nm light: left – fluorescence spectra in a nematic host for different linear polarizations; right – fluorescence spectra in a cholesteric host for circular polarization of different handedness.

The value of ρ in this case is -0.5 for fluorescence maximum. This is a typical value for polarized fluorescence from planar-aligned liquid crystals; see [54], where a value of ρ equal to 0.6 was reported for polarized electroluminescence from liquid crystal polymers.

Circularly polarized single photons with well-defined handedness can be produced with high efficiency using 1-D photonic band-gaps (figure 1) in cholesteric liquid crystals matching the dye-fluorescence band. Figure 7, right, shows predominance of left-handed circularly polarized fluorescence over right-handed fluorescence from a planar-aligned, glassy cholesteric liquid crystal layer that is \sim 4.1 µm thick and has a 0.5% weight concentration of DiIC₁₈(3) dye. Our next step will be to make a single-photon source with predominant circular polarization of definite handedness in this material.

6. Conclusions and future directions

This paper shows the advantages of using liquid crystals as hosts for single-photon sources; definite polarization in fluorescence from single emitters embedded in liquid-crystal hosts was demonstrated for the first time at room temperature. Single-dye molecules were uniaxially aligned by liquid crystal molecules in one direction and produced linearly polarized single photons with definite polarization. Current technology needs to be improved in the direction of further purification of the host material and better alignment.

We estimated the efficiency P of polarized single-photon emission into the collecting objective by comparing the number of exciting 532 nm photons per unit per time N_{inemol} on the absorption cross-sectional area σ of DiIC₁₈(3) dye with the measured polarized photon count rate $N_{\text{outpol}} = 3.65 \text{ kc/s}$ from a single molecule with $\rho = 1$. For the laser power incident on the sample (input power on the first objective) ~8 μ W, a beam radius ~0.2 μ m, and using value of σ ~1.3 × 10⁻¹⁶ cm² [60] for DiIC₁₈(3) molecules, we arrive at photons/s per molecule $N_{\text{incmol}} = 2.21 \times$ 10^6 photons/(s mol). We can evaluate an efficiency *P* from the following expression: $N_{\text{outpol}} = 0.96 N_{\text{incmol}} \alpha \beta PDQ$. Here $\alpha = 0.525$ is the measured transmission of rejection filters, $\beta = 0.45$ is the measured total transmission and collection coefficient of two objectives and microscope optics after the second objective but before the beamsplitter cube (without rejection filters), D = 0.2 is the measured coupling efficiency of the fibre optics used in this setup, Q = 0.64 is the photon detection efficiency of the avalanche photodiode at 580 nm, 0.96 is the coupling efficiency from the fibre to the APD-FC-connector. In this calculation we considered a fluorescence quantum yield of the dye molecule close to 1 [60]. From these data we deduced $P \approx 6\%$.

The value of *P* can be increased several times using the Purcell factor in a 1-D photonic band-gap microcavity made of cholesteric liquid crystal. Proper selection of incident polarization can increase the source efficiency in a nematic sample as well. Using a photonic band-gap will decrease fluorescence bandwidth and fluorescent lifetime. 1-D photonic band-gap structures in cholesteric liquid crystals, 2-D/3-D photonic crystals in holographic polymer-dispersed liquid crystals [61], a 3-D

photonic crystal structure of a liquid crystal blue phase [62], and photonic crystals/microstructured fibres infiltrated with liquid crystals [63, 64] may be prepared for this purpose. By using various fluorescence emitters which can be either dissolved or dispersed in a liquid crystal host [dye molecules, colloidal semiconductor nanocrystals (dots and rods), carbon nanotubes or rare-earth ions], it will be possible to extend the working region of this source from the visible to communication wavelengths (1.3 and $1.55 \,\mu$ m).

Our main directions of improvement of current technology will be a combination of liquid crystal alignment technique with oxygen depletion to reduce dye bleaching. Efficiency increase of the source will also be made by using cholesteric 1-D photonic band-gap structures.

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