JOURNAL OF MODERN OPTICS, 15 JUNE–10 JULY 2004
VOL. 51, NO. 9–10, 1535–1547

Taylor & Francis
Taylor & Francis Group

Dye-doped cholesteric-liquid-crystal room-temperature single-photon source*

SVETLANA G. LUKISHOVA†, ANSGAR W. SCHMID‡, CHRISTOPHER M. SUPRANOWITZ†, NADINE LIPPA‡, ANDREW J. McNAMARA†, ROBERT W. BOYD† and C. R. STROUD, JR†

†The Institute of Optics, University of Rochester, Rochester, New York 14627-0186, USA; email: lukishov@optics.rochester.edu
‡Laboratory for Laser Energetics, University of Rochester, 250 East River Road, Rochester, New York 14623-1299, USA

(*Received 30 June 2003*)

Abstract. Fluorescence antibunching from single terrylene molecules embedded in a cholesteric-liquid-crystal host is used to demonstrate operation of a room-temperature single-photon source. One-dimensional (1-D) photonic-band-gap microcavities in planar-aligned cholesteric liquid crystals with band gaps from visible to near-infrared spectral regions are fabricated. Liquid-crystal hosts (including liquid crystal oligomers and polymers) increase the source efficiency, firstly, by aligning the dye molecules along the direction preferable for maximum excitation efficiency (deterministic molecular alignment provides deterministically polarized output photons), secondly, by tuning the 1-D photonic-band-gap microcavity to the dye fluorescence band and thirdly, by protecting the dye molecules from quenchers, such as oxygen. In our present experiments, using oxygen-depleted liquid-crystal hosts, dye bleaching is avoided for periods exceeding one hour of continuous 532 nm excitation.

1. Introduction

A *single-photon source* (SPS) [1, 2] that efficiently produces photons with *antibunching* characteristics [3] is a pivotal hardware element for quantum communication technology [4–6]. Using a SPS, secure quantum communication will prevent any potential eavesdropper from intercepting a message with a secure key without the receiver noticing [4]. Quantum communication has a potential large market [7, 8], but its practical realization is held back in part because of the difficulties in developing robust sources of antibunched photons on demand. In another implementation, a SPS becomes the key hardware element for quantum computers with linear optical elements and photodetectors [9–12].

In spite of several solutions for SPSs presented in the literature, significant drawbacks remain. They are the reason why current quantum communication systems are baud-rate bottlenecked, causing photon numbers from ordinary photon sources to attenuate to the single-photon level (approximately 0.1 photon per pulse

* The referee process for this paper was handled by the Special Issue Editors.

Journal of Modern Optics ISSN 0950–0340 print/ISSN 1362–3044 online
http://www.tandf.co.uk/journals
DOI: 10.1080/09500340410001674439

on average). In addition to the low efficiency, the drawback of such faint-pulse quantum cryptography is pollution by multiple photons. The pollution restriction does not vanish in quantum cryptography based on parametric-downconversion entangled-photon pairs. A parametric-downconversion photon source may contain a coherent superposition of multiple pairs.

An efficient (with a photon number per pulse an order of magnitude higher) and reliable light source that delivers a train of pulses containing one, and only one, photon is a very timely challenge. To meet this challenge, several issues need to be addressed, from achieving full control of the quantum properties of the source to easy handling and integrability of these properties into a practical quantum computer and/or communication set-up. In addition, in quantum information systems it is desirable to deal with single photons synchronized to an external clock, namely triggerable single photons [13]. *Polarization states* of single photons are also important since they enable polarization-qubit encoding of information.

The critical issue in producing single photons in another way than by trivial attenuation of a beam is the *very low concentration of photon emitter*s dispersed in a host such that, within a laser focal spot, only one emitter becomes excited, emitting only one photon at a time. In initial demonstrations of resonance-fluorescence photon antibunching [14], SPSs possessed a random photon emission time [14–22]. Single photons 'on demand', that is, triggerable single photons, were obtained only recently [13, 23–34]. De Martini *et al*. [23] used an active microcavity excited by a mode-locked laser. In experiments by Kim *et al*. [24], Imamoglu and Yamamoto [25] and Moreau *et al*. [26], a cryogenic-temperature single-photon turnstile device utilized Coulomb blockade of tunnelling for electrons and holes in a mesoscopic double-barrier p–n junction. Single photons were generated at the modulation periodicity of the junction voltage. Michler *et al*. [27], Santori *et al*. [28] and Zwiller *et al*. [29] demonstrated cryogenic-temperature single-photon devices using pulsed-laser excitation of a single semiconductor heterostructure quantum dot. Electrically driven single photons (also at cryogenic temperatures) were obtained by Yuan *et al*. [30]. Experiments by Brunel *et al*. [31], Lounis and Moerner [32] and Treussart *et al*. [33] were based on an entirely different system, namely single dye molecules embedded at a low concentration within organic single-crystal platelets or covered by a polymer layer. Single photons were triggered either by a combination of continuous-wave laser excitation and an electronic signal [31] or by short-pulse laser irradiation [32, 33]. A triggered on demand SPS was also obtained recently with colour centres in diamond [34]. Most of these sources (see for example [24–31]) operate reliably only at liquid-helium temperature—a major impediment to widespread use.

To date, three approaches are known to operate at room temperature: two inorganic and one organic. The first inorganic room-temperature approach involves a single-crystal or polycrystal diamond and one of its colour centres [19, 20, 34]. The second inorganic approach uses single colloidal CdSe/ZnS quantum dots [35–37]. The alternative organic approach, based on the first experiment of Ambrose *et al*. [18] at room temperature and numerous previous experiments around liquid-helium temperature (see for example [38]), uses single dye molecules (e.g. rhodamine and terrylene) as emitters [18, 21, 22, 32, 33].

As acceptable as these approaches may be strictly on quantum-optics grounds, all suffer from shortcomings. Their specific shortcomings include the following:

(i) Polarization of the single photons varies from one emitter to another (nondeterministic).
(ii) Single photons are produced with very low efficiency and polluted by additional photons at about the same frequency from the host material [13, 32].
(iii) Alternatives such as colour centres in diamond and colloidal CdSe/ZnS quantum dots possess long fluorescence lifetimes; for instance, the diamond colour centre has 11.6 ns and 22.7 ns fluorescence lifetimes in a single crystal and polycrystal respectively, and CdSe/ZnS quantum dots have a fluorescence lifetime of approximately 22 ns.

The key advantage of dye molecules is that their excited-state lifetime of only a few nanoseconds permits excitation repetition rates above approximately 100 MHz. Another advantage is the variety of dyes with fluorescence in the visible and near-infrared spectral regions. In amorphous media, dye molecules tend to be unstable; they blink at various characteristic time intervals, change their spectral behaviour and are easily bleached. Recently, however, single terrylene molecules have been doped into *p*-terphenyl molecular crystals prepared by a sublimation procedure that produced tiny platelets [21, 32, 39, 40]. In this host, the chromophore is protected from exposure to diffusing quenchers (such as oxygen) and benefits from strong phonon emission into the host, preventing rapid thermal decomposition of the chromophore under intense irradiation. It was found that for 'thick' *p*-terphenyl crystals (about 10 μm), this system becomes extremely photostable [21, 32, 39, 40], allowing hours of continuous illumination of individual molecules without photobleaching [32]. It assures long-term spectral stability and reproducibility from one terrylene absorber to the next [40]. Pumped by periodic short-pulse laser radiation [32], single photons were generated at predetermined times at pump-pulse repetition rates within the accuracy of the emission lifetime (about 3.8 ns).

Technical implementation of a *robust SPS* based on this system is difficult because these monoclinic sublimation-produced microcrystals are stress sensitive and fragile. In addition, terrylene's molecular dipole moment in the *p*-terphenyl host crystal takes on an orientation perpendicular to the platelet's surface (i.e. perpendicular to the incident light's *E* field) [32]. This, in turn, leads to poor coupling with the polarized excitation light, prompting poor fluorescence emission even at high excitation intensities (the saturation intensity is about 1 MW cm^{-2} at room temperature). In spite of the elegance of the terrylene–*p*-terphenyl experiments, this technology must be considered unrealistic for practical application. Its weak point is also a background from 'ordinary photons' from out-of-focus molecules or Raman scattering because of the very high pumping intensities required. Emitted photons are not polarized deterministically (there is no known efficient method for aligning rapidly a multitude of micrometre-sized monoclinic crystallites relative to one another).

Note that non-crystalline amorphous hosts, for example polymers, do not offer the same spectral stability in single-molecule emission even in the case of terrylene [40], nor do they provide long-time protection against bleaching as the *p*-terphenyl host does. To date, no crystal hosts other than the fragile sublimated *p*-terphenyl flakes have been proposed in single-molecule room-temperature experiments.

This article describes some new approaches toward implementing an efficient, deterministically polarized SPS on demand:

(i) Using liquid-crystal hosts (including liquid-crystal oligomers and polymers) to align the emitter molecules preferentially for maximum excitation efficiency (deterministic molecular alignment will also provide deterministically polarized output photons);
(ii) Using planar-aligned cholesteric-liquid-crystal hosts [41] as one dimensional (1-D) photonic-band-gap microcavities tunable to the dye fluorescence band [42–44];
(iii) Using liquid-crystal technology to prevent dye bleaching.

2. Preparation of one-dimensional photonic-band-gap cholesteric-liquid crystal samples

In planar cholesterics (chiral nematics), rod-like anisotropic molecules with small chiral 'tails' are self-assembled in the space. The axes of the molecular director rotate monotonically to form a periodic helical structure with pitch P_0 [41] and helix axis perpendicular to the molecular director. For liquid crystal thicknesses of 10 μm or greater, the reflectance of normally incident, circularly polarized light with electric-field vector rotation opposite to the rotation of molecules in the helical structure (Bragg condition) approaches 100% within a band centered at $\lambda_0 = n_{av}P_0$, where $n_{av} = (n_e + n_o)/2$ is the average of the ordinary and extraordinary refractive indices of the medium. This is the so-called selective reflection of cholesteric liquid crystals. The bandwidth is approximately $\Delta\lambda = \lambda_0 \Delta n/n_{av}$, where $\Delta n = n_e - n_o$. Such a periodic structure can also be viewed as a 1-D photonic crystal, with a band gap within which propagation of light is forbidden. For emitters located within such a structure, the rate of spontaneous emission is suppressed within the spectral stop band and enhanced near the band edge [42]. Several groups have reported lasing in photonic band-gap material hosts, including cholesteric liquid crystals [42–44]. Generation of strongly circularly polarized photoluminescence from dye-doped planar layers of glass-forming chiral-nematic liquid crystals was also reported [45, 46].

To prepare the 1-D photonic-band-gap structures three different planar alignment procedures for liquid crystals were used: substrate shearing, buffing and photoalignment. For sheared samples, no additional substrate coatings were needed. For buffing, substrates were spin coated with either of two polymers: Nylon-6 or Polyimid. For buffing, we used a standard velvet-surface buffing machine.

Single-molecule fluorescence microscopy imposes a requirement on the sample thickness: the working distance of about 180 and 300 μm for high-numerical-aperture objectives permits use of samples only with a thickness not exceeding this value. For this reason, glass microscopic cover slip substrates (Corning) of about 170 μm thickness were used that both are fragile and need special care in handling. To prevent damage to the fragile substrates during the buffing procedure, cover slips were 'blocked to' microscope slides 1 mm thick with water-soluble acetate, using heating at 80°C for 40 min to achieve better results. After buffing, the cover slips were unblocked by standing in deionized water overnight. This was followed by a rinse in flowing deionized water to rid the samples of acetate traces. For photoalignment, substrates were spin coated with Staralign. Photoalignment of coated polymer was achieved using six ultraviolet (UV) discharge lamps RPR 3000 (Southern New England Ultraviolet Co.) with a maximum wavelength of about 302 nm (40 nm bandwidth) and UV linear dichroic

polarizer (Oriel), 1.74 in × 1.74 in size, placed in a hermetic box. The photoalignment procedure at about 5 mW cm^{-2} power density at 302 nm lasted 10 min.

We used two types of liquid crystals: Wacker oligomer cholesteric-liquid-crystal (OCLC) powders [47], and mixtures of low-molecular-weight E7 nematic-liquid-crystal blend with a chiral additive CB15. Both E7 and CB15 are fluids at room temperature. Both materials were supplied by EM Industries.

For the Wacker oligomer liquid-crystal powders, the samples were prepared by mixing different concentration of two powders with individually known selective-reflection wavelengths provided by the vendor. In order to obtain a desired selective-reflection wavelength mixture, mixing rules were found empirically from a set of multiple different mixtures. To change the pitch of each mixture, powders were dissolved in methylene chloride, mixed for 2 h under agitation and, at an elevated temperature, purified through a 0.45 μm particle filter and dried from solvent under vacuum. For planar alignment an uncoated cleaned cover slip with a Wacker powder was placed on a hot plate and melted at 118°C. A second cover slip was used to shear the melted oligomer at temperature (and to also form the second window of the liquid-crystal cell). Slowly cooling the cell to room temperature froze in the planar alignment. For some Wacker powders, we used spin coating with Polyimid and buffing of substrates. Cells with known and uniform thickness (10–15 μm) were created by using four drops of a UV-cured epoxy mixed with calibrated glass-bead spacers at the substrate corners. After that, cells containing Wacker powder were heated into the isotropic phase and slowly cooled.

For low-molecular-weight liquid crystals, the coated substrate surfaces were either buffed or photoaligned. Cell thickness was again set by UV epoxy mixed with glass-bead spacers. To find the weight concentration C of the components in a mixture of chiral additive and nematic liquid crystal with desired selective reflection wavelength λ_0, we used a well-known relationship $C = n_{av}/(\lambda_0 \times \mathrm{HTP})$, where HTP is the helical twisting power of the chiral additive in a nematic liquid crystal. For CB15 in E7, HTP $\approx 7.3\,\mu m^{-1}$. A E7 + CB15 liquid-crystal mixture with selected concentration was fed through a 0.45 μm particle filter and a stainless-steel syringe into the assembled cell parallel to the polymer alignment direction inscribed in the cell walls.

We prepared several tens of planar-aligned cholesteric-liquid-crystal samples with band gaps in different spectral regions (λ_0 varied from 430 to 2200 nm). We used a Perkin–Elmer Lambda 900 spectrophotometer to measure the wavelength response of each prepared sample, thereby determining the specific selective reflection (photonic band gap) for each sample. A zero-order quarter wave plate and a thin-film linear polarizer were used in both spectrophotometer channels to create circularly polarized incident light of desired handedness. Samples were tested in unpolarized, as well as in left-handed and right-handed circularly polarized incident light. Figure 1 shows transmittance of Wacker OCLC samples versus wavelength in left-handed circularly polarized light. In these experiments, we used both pure Wacker OCLCs with selective reflection bands centred around 450 nm, 535 nm and 760 nm, and mixtures of two OCLCs possessing selective reflection closest to the desired wavelength.

Similar results were achieved with the E7+CB15 mixture, both with buffed Polyimid/Nylon-6 (figure 2) and with photoalignment (figure 3), in right-handed circularly polarized light (handedness strictly determined by the CB15 structure).

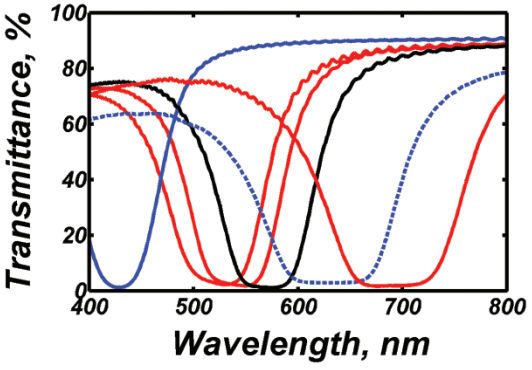


Figure 1. Selective reflection of left-handed circularly polarized light from photonic-band-gap Wacker OCLCs.

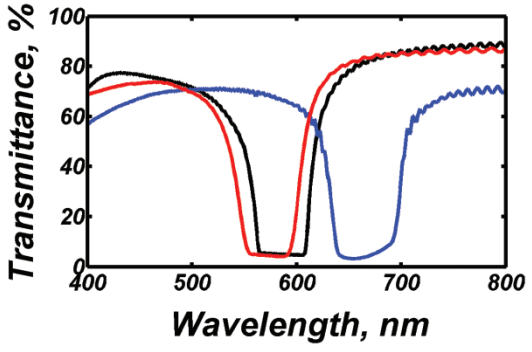


Figure 2. Selective reflection of right-handed circularly polarized light from a photonic-band-gap E7+CB15 mixture. Planar alignment was made with the buffing method.

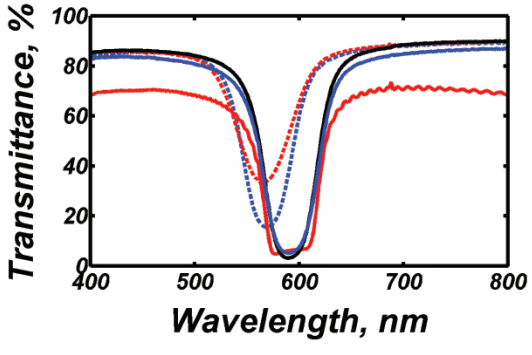


Figure 3. Selective reflection of right-handed circularly polarized light from a photonic-band-gap E7+CB15 mixture. Planar alignment was made with the photoalignment method.

3. Preparation of liquid crystals doped with single-molecules of terrylene dye

To minimize false fluorescence contributions by contaminants during single-molecule fluorescence microscopy, rigorous cleaning of glass substrates is

mandatory. Liquid-crystal cells were fabricated in a class 10 000 liquid-crystal clean-room facility. Ultrasonic cleaning for 60 min freed the 1 in × 1 in substrates from any dirt particles. Substrates were then rinsed in flowing deionized water, dried in a stream of compressed nitrogen and washed in toluene from organic components. To remove the toluene, substrates were washed again with pure ethanol and dried. After that, they were etched in piranha solution ($H_2SO_4 + H_2O_2$ in equal volume concentration) for about 20 min, rinsed in flowing deionized water and dried in a stream of oil-free nitrogen.

The appropriate terrylene concentration for single-molecule fluorescence microscopy was established by iterative trial and error. In sequential dilution steps of terrylene in chlorobenzene solvent, solutions were spun on to glass slips and, for each concentration, confocal fluorescence microscopy determined the final emitter concentration per irradiation volume. Once single molecules were predominantly observed, the dilution end point was reached. This final terrylene solution was mixed with Wacker OCLC starting material (8 wt% concentration of oligomer), E7+CB15 or 5CB+CB15 mixtures.

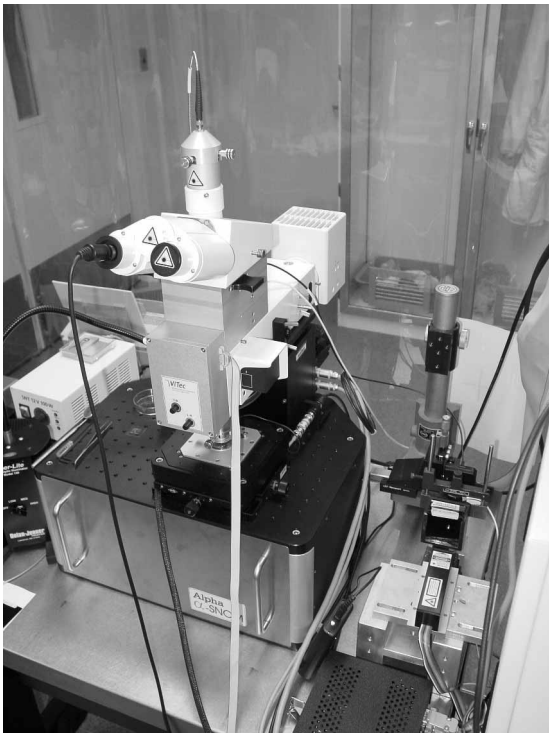
Terrylene's fluorescence maximum lies near 579 nm with a bandwidth of about 30 nm. Figure 1 (red curves) shows matching the cholesteric liquid crystal's λ_0 to the dye-fluorescence band, but in our current experiments we used Wacker OCLC with $\lambda_0 = 2.2\,\mu m$, that is outside the terylene dye fluorescence band. The liquid crystal was doped with terrylene at an extremely low concentration, such that the final sample contained only a few molecules per square micrometre of irradiation area.

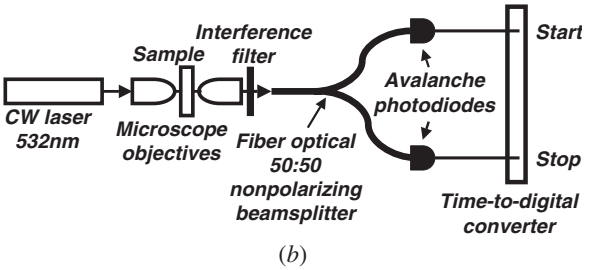
4. Experimental set-up for single-dye-molecule fluorescence microscopy and antibunching correlation measurements

Photon-antibunching correlation measurements are carried out using the set-up shown in figure 4. The terrylene-doped liquid-crystal sample is placed in the focal plane of a microscope objective of 0.8 numerical aperture. (Witec Alpha-SNOM platform (figure 4(*a*)). The sample is attached to a piezoelectric XYZ translation stage. Light emitted by the sample is collected by a confocal set-up using an oil immersion objective of 1.25 numerical aperture together with an aperture in form of optical fibre of 50 μm core. The continuous wave, spatially filtered (through a single-mode fibre), linearly polarized (contrast, $10^5 : 1$), 532 nm diode-pumped neodymium-doped yttrium aluminium garnet laser output excites single molecules. In focus, the intensities used are of the order of several kilowatts per square centimetre. The collection fibre is part of a non-polarization-sensitive 50 : 50 fibre splitter that forms the two arms of a Hanbury Brown–Twiss [48] correlation set-up (figure 4(*b*)). Residual transmitted excitation light is removed by two consecutive dielectric interference filters yielding a combined rejection of better than six orders of magnitude at 532 nm.

Photons in the two Hanbury Brown–Twiss arms are detected by identical cooled avalanche photodiodes in single-photon-counting Geiger mode (Perkin–Elmer SPCM AQR-14). The time interval between two consecutively detected photons in separate arms is measured by a Phillips Scientific 7186 time-to-digital converter with a full scale of 68 ns using a conventional start–stop protocol. Within this converter's linear range, the time uncertainty in each channel corresponds to 25 ps.

(*a*)





(*b*)

Figure 4. (*a*) Witec Alpha-SNOM microscope with a laser; (*b*) experimental set-up for photon antibunching correlation measurements: CW, continuous wave.

It has been proved experimentally (see for example [21, 22]) that a very good approximation of the autocorrelation function $g^{(2)}(\tau)$ comes directly from the coincidence counts (event distribution) $n(\tau)$, for relatively low detection efficiency and therefore low counting rate. That is why we consider that $n(\tau)$ is proportional to the autocorrelation function $g^{(2)}(\tau)$. For single photons, $g^{(2)}(0) = 0$, indicating the absence of pairs, that is, antibunching.

5. Experimental results: single-dye-molecule fluorescence in a cholesteric-liquid-crystal host; photon antibunching correlation measurements

Figure 5 shows single-terrylene-dye-molecule fluorescence images obtained by confocal fluorescence microscopy. Single terrylene molecules are embedded in a Wacker OCLC host. For this image, the scan direction is from left to right

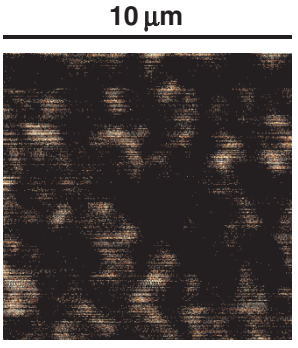


Figure 5. Single-terrylene-molecule fluorescence.

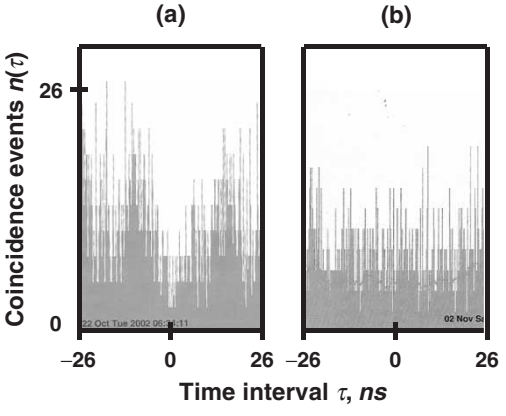


Figure 6. The histogram of coincidence events of (*a*) single-terrylene-molecule fluorescence in a Wacker OCLC host and (*b*) an assembly of several uncorrelated molecules.

and line by line from top to bottom. Most single molecules in this sample exhibited fluorescence blinking in time. In figure 5, this 'blinking' behaviour by single molecules manifests itself as bright and dark horizontal stripes in the image.

Blinking is a common phenomenon and convincing evidence of the single-photon nature of the source. The explanation of the nature of this long-time blinking from milliseconds to several seconds remains a subject of debates in the literature (see for example [49]).

Figure 6 (*a*) shows a coincidence-count histogram $n(\tau)$ from a single terrylene molecule in a Wacker OCLC host. The scan speed is about 3 s per line (512 pixels). This histogram exhibits a dip at $\tau = 0$. The measured signal-to-background ratio of our experiments ranges from 2 to 8; so the probability that a photon from the background triggers a coincidence with a photon from the molecule is very low. Because $n(\tau)$ is proportional to the autocorrelation function $g^{(2)}(\tau)$, $n(0) \approx 0$ means that $g^{(2)}(\tau) \approx 0$ in our experiments. Two fluorescence photons are not observed within an arbitrarily short time interval. This fluorescence antibunching is due to the finite radiative lifetime of the molecular dipole and is therefore clear proof that we observed the emission of one, and only one, molecule. The histogram in figure 6 (*b*) from multiple uncorrelated molecules in the same host but a different sample shows no such dip at $\tau = 0$, that is, no antibunching.

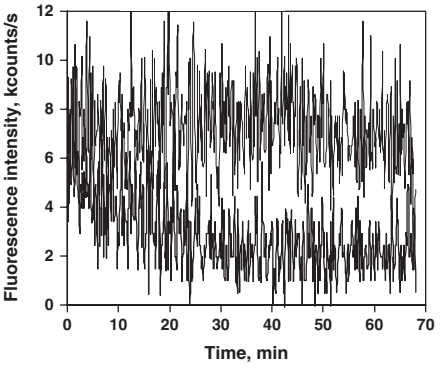


Figure 7. Fluorescence bleaching behaviour of an assembly of terrylene molecules as a function of time and in two different liquid-crystal hosts.

6. Experimental results: preventing dye bleaching in liquid crystal hosts

Practical device implementation also depends on the photochemical stabilities of both emitters and hosts. We increased terrylene fluorescence stability in monomeric liquid-crystal hosts by saturating the liquid crystals with helium in a sealed glove-box for 1 h. Oxygen, which is mostly responsible for dye bleaching, is displaced by helium during this procedure. Ground-state oxygen can form highly reactive singlet oxygen by quenching a triplet state of the dye. The singlet oxygen can then react with its surroundings, including dye molecules. Figure 7 shows fluor-escence-bleaching results of terrylene molecules at a concentration two orders of magnitude higher than in single-molecule experiments in different liquid-crystal hosts: either immobilized in an OCLC or dissolved in monomeric cyanobiphenyl 5CB saturated with helium (both at identical excitation intensities and identical terrylene volume concentrations). Over the course of more than 1 h, *no* dye bleaching was observed in the *oxygen-depleted* liquid-crystal host (upper curve).

Dye bleaching is not a critical impairment for an efficient SPS, but it is an important factor for device simplicity and cost. When one molecule is bleached, the system can be rapidly realigned to utilize another isolated dye molecule, allowing almost continuous source action.

7. Conclusion

A robust room-temperature SPS based on fluorescence from a single dye molecule (fluorescence antibunching) was demonstrated for the first time for liquid-crystal hosts. Planar-aligned 1-D photonic-band-gap structures for visible and near-infrared in dye-doped cholesteric oligomer and low-molecular-weight liquid-crystal mixtures were prepared. Avoiding bleaching of the terrylene dye molecules for excitation times longer than 1 h was achieved by innovative preparation procedures.

The estimated efficiency p_α of our current SPS is about 4% [50], but it can be increased to one order of magnitude, firstly, by aligning the dye molecules along a direction preferable for maximum excitation efficiency and, secondly by tuning a 1-D photonic-band-gap microcavity of a planar-aligned cholesteric liquid crystal to the dye fluorescence band.

Detailed source-efficiency issues will be addressed in a subsequent article. Here we evaluate the probability P_2 of two-photon emission. If P_2 is much smaller than unity, $p_2 = C_N(0)P_1^2/2$ [20, 34]. P_1 is the probability of single photon emission, $C_N(0)$ is the zero time *normalized* coincidence rate that can be taken directly as the correlation function $g^{(2)}(0)$. For Poissonian light, $C_N(0) = 1$. In our case, for single-terrylene-molecule fluorescence in a Wacker oligomer liquid-crystal host (figure 6), $C_N(0) = 0.25$–0.33. It means that the rate of two-photon pulses is three to four times lower than for Poissonian light. It should be noted that an efficiency p_α introduced earlier, $p_\alpha = \alpha P_1$. Here α is a collection efficiency including losses in filters.

Our future work is directed towards increasing the efficiency, life and polarization purity of the SPS by improved selection of dye, liquid crystal, and the photonic-band-gap structure matching with the dye fluorescence band. A pulsed laser source will be used to create a real quantum cryptography system with a cholesteric-liquid-crystal SPS on demand. We are also planning to develop a 1-D photonic-band-gap liquid-crystal SPS at the communication wavelengths of 1300 and 1500 nm. Current colloidal quantum-dot technology, for instance, using PbSe quantum dots of specific size [51] provides single emitters with a fluorescence in a spectral region between 1000 and 2200 nm. These quantum dots can be easy dissolved in liquid crystals.

Acknowledgments

The authors acknowledge the support by the US Army Research Office under award DAAD19-02-1-0285. The work was also supported by the US Department of Energy (DOE) Office of Inertial confinement fusion under cooperative agreement DE-FC03-92SF19460, the University of Rochester, and the New York State Energy Research and Development Authority. The support of DOE does not constitute an endorsement by DOE of the views expressed in this article.

Receipt of OCLC starting material from Dr F. Kreuzer of Wacker, Munich, is gratefully acknowledged. The authors thank L. Novotny, K. Marshall, J. Shojaie, O. Hollricher and T. Kosc for advice and help, J. Howell and D. A. Voloshchenko for fruitful discussions, and J. Starowitz for help in the Optical Materials Laboratory.

References

[1] Solid state sources for single photons, European program S4P, http://www.iota.u-psud.fr/~S4P/.
[2] Jonsson, P., 2002, Doctoral Dissertation, Royal Institute of Technology, Stockholm, Sweden, 101 pp.
[3] Walls, D. F., and Milburn, G. J., 1995, *Quantum Optics* (Berlin: Springer Verlag), p. 42.
[4] Gisin, N., Ribordy, G., Tittel, W., and Zbinden, H., 2002, *Rev. Mod. Phys.*, **74,** 145.
[5] Kwiat, P. G. (editor), 2002, *New J. Phys.*, **4,** July.
[6] Klarreich, E., 2002, *Nature*, **418,** 270.
[7] MagiQ Technologies, http://www.magiqtech.com, accessed 18 March 2004.
[8] id Quantique, http://www.idquantique.com, accessed 18 March 2004.
[9] Knill, E., Laflamme, R., and Milburn, G. J., 2001, *Nature*, **409,** 46.
[10] Calsamiglia, J., 2001, Academic Dissertation, University of Helsinki, Helsinki, Finland.

[11] Pittman, T. B., Jacobs, B. C., and Franson, J. D., 2002, *Phys. Rev. Lett.*, **88,** 257 902.
[12] Shields, A., 2002, *Science*, **297,** 1821.
[13] Benjamin, S., 2000, *Science*, **290,** 2273.
[14] Kimble, H. J., Dagenais, M., and Mandel, L., 1977, *Phys. Rev. Lett.*, **39,** 691.
[15] Hong, C. K., and Mandel, L., 1986, *Phys. Rev. Lett.*, **56,** 58.
[16] Dietrich, F., and Walther, H., 1987, *Phys. Rev. Lett.*, **58,** 203.
[17] Grangier, P., Roger, G., and Aspect, A., 1986, *Europhys. Lett.*, **1,** 173.
[18] Ambrose, W. P., Goodwin, P. M., Enderlein, J., Semin, D. J., Martin, J. C., and Keller, R. A., 1997, *Chem. Phys. Lett.*, **269,** 365.
[19] Kurtsiefer, C., Mayer, S., Zarda, P., and Weinfurter, H., 2000, *Phys. Rev. Lett.*, **85,** 290.
[20] Brouri, R., Beveratos, A., Poizat, J.-P., and Grangier, P., 2000, *Optics Lett.*, **25,** 1294; Beveratos, A., Brouri, R., Gacoin, T., Poizat, J.-P., and Grangier, P., 2001, *Phys. Rev.* A, **64,** 061 802R/1–4.
[21] Fleury, L., Segura, J.-M., Zumofen, G., Hecht, B., and Wild, U. P., 2000, *Phys. Rev. Lett.*, **84,** 1148.
[22] Treussart, F., Clouqueur, A., Grossman, C., and Roch, J.-F., 2001, *Optics Lett.*, **26,** 1504.
[23] De Martini, F., Di Giuseppe, G., and Marrocco, M., 1996, *Phys. Rev. Lett.*, **76,** 900.
[24] Kim, J., Benson, O., Kan, H., and Yamamoto, Y. A., 1999, *Nature*, **397,** 500.
[25] Imamoglu, A., and Yamamoto, Y., 1994, *Phys. Rev. Lett.* **72,** 210.
[26] Moreau, E., Robert, I., Gerard, J. M., Abram, I., Manin, L., and Thiery-Mieg, V., 2001, *Appl. Phys. Lett.*, **79,** 2865.
[27] Michler, P., Kiraz, A., Becher, C., Schoenfeld, W. V., Petroff, P. M., Zhang, L., Hu, E., and Imamoglu, A., 2000, *Science*, **290,** 2282.
[28] Santori, C., Pelton, M., Solomon, G., Dale, Y., and Yamamoto, Y., 2001, *Phys. Rev. Lett.*, **86,** 1502.
[29] Zwiller, V., Blom, H., Jonsson, P., Panev, N., Jeppesen, S., Tsegaye, T., Goobar, E., Pistol, M.-E., Samuelson, L., and Björk, G., 2001, *Appl. Phys. Lett.*, **78,** 2476.
[30] Yuan, Z. L., Kardynal, B. E., Stevenson, R. M., Shields, A. J., Lobo, C. J., Cooper, K., Beattie, N. S., Ritchie, D. A., and Pepper, M., 2002, *Science*, **295,** 102.
[31] Brunel, C., Lounis, B., Tamarat, P., and Orrit, M., 1999, *Phys. Rev. Lett.,* **83,** 2722.
[32] Lounis, B., and Moerner, W. E., 2000, *Nature*, **407,** 491.
[33] Treussart, F., Alleaume, R., Le Floch, V., Xiao, L. T., Courty, J. M., and Roch, J. F., 2002, *Phys. Rev. Lett.*, **89,** 093 601.
[34] Beveratos, A., Kuhn, S., Brouri, R., Gacoin, T., Poizat, J. P., and Grangier, P., 2002, *Eur. Phys. J.* D, **18,** 191; Beveratos, A., Brouri, R., Gacoin, T., Villing, A., Poizat, J. P., and Grangier, P., 2002, *Phys. Rev. Lett.*, **89,** 187 901.
[35] Michler, P., Imamoglu, A., Mason, M. D., Carson, P. J., Strouse, G. F., and Buratto, S. K., 2000, *Nature*, **406,** 968.
[36] Lounis, B., Bechtel, H. A., Gerion, D., Alivisatos, P., and Moerner, W. E., 2000, *Chem. Phys. Lett.*, **329,** 399.
[37] Messin, G., Hermier, J. P., Giacobino, E., Desbiolles, P., and Dahan, M., 2001, *Opt. Lett.,* **26,** 1891.
[38] Basché, Th., Moerner, W. E., Orrit, M., and Talon, H., 1992, *Phys. Rev. Lett.*, **69,** 1516.
[39] Fleury, L., Sick, B., Zumofen, G., Hecht, B., and Wild, U. P., 1998, *Molec. Phys.*, **95,** 1333.
[40] Kulzer, F., Koberling, F., Christ, Th., Mews, A., and Basché, Th., 1999, *Chem. Phys.*, **247,** 23.
[41] Chandrasekhar, S., 1977, *Liquid Crystals* (Cambridge University Press).
[42] Kopp, V. P., Fan, B., Vithana, H. K. M., and Genack, A. Z., 1998, *Opt. Lett.*, **23,** 1707.

[43] IL'CHISHIN, I. P., TIKHONOV, E. A., TISHCHENKO, V. G., and SHPAK, M. T., 1980, *JETP Lett*., **32,** 24.
[44] FINKELMAN, H., KIM, S. T., MUNOZ, A., PALFFY-MUHORAY, P., and TAHERI, B., 2001, *Adv. Mater*., **13,** 1069.
[45] CHEN, S. H., KATSIS, D., SCHMID, A. W., MASTRANGELO, J. C., TSUTSUI, T., and BLANTON, T. N., 1999, *Nature*, **397,** 506.
[46] KATSIS, D., SCHMID, A. W., and CHEN, S. H., 1999, *Liq. Cryst*., **26,** 181.
[47] BUNNING, T. J., and KREUZER, F.-H., 1995, *Trends in Polym. Sci.* **3,** 318.
[48] HANBURY BROWN, R., and TWISS, R. Q., 1956, *Nature*, **177,** 27.
[49] VARGAS, F., HOLLRICHER, O., MARTI, O., DE SCHAETZEN, G., and TARRACH, G., 2002, *J. Chem. Phys*., **117,** 866.
[50] LUKISHOVA, S. G., SCHMID, A. W., MCNAMARA, A. J., BOYD, R. W., and STROUD, C. R., JR, 2003, *IEEE J. Selected Topics Quant. Electron*., **9,** 1512.
[51] WEHRENBERG, B. L., WANG, C., and GUYOT-SIONNEST, P., 2002, *J. Phys. Chem*. B, **106,** 10 634.