Deterministically polarized, room temperature source of single photons


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The purpose of our project is to make a robust, deterministically polarized and efficient room-temperature source of single photons on demand by special preparation of host with single-emitters [1-4];

The source based on a single-emitter fluorescence in a nanostructured liquid-crystal host [3,4] is a room temperature alternative to cryogenic single-photon sources based on semiconductor heterostructures.

Content:

• Introduction: single emitters in nanostructured liquid crystal hosts;
• Glassy nematic and cholesteric liquid crystals;
• Deterministically polarized fluorescence from single dye molecules in aligned nematic host;
• Antibunching in cholesteric liquid crystal host;
• Current source efficiency;
• Reducing dye bleaching by special host treatment;
• Summary and future plans
Different types of fluorescence emitters for room-temperature SPSs can be dissolved and/or dispersed in liquid crystal hosts

- *single-dye-molecules*;
- *colloidal semiconductor quantum dots and rods*;
- *single polymer molecules*;
- *single-wall carbon nanotubes*
Current challenges in dye-based SPSs*)

- emitter bleaching and blinking;
- low collection and excitation efficiencies;
- scattered-photon background;
- nondeterministic polarization state of photons.

We propose

- To use liquid crystal hosts (including liquid crystal oligomers/polymers) to align the dopant along the direction preferable for excitation efficiency (along the light polarization).

Deterministic molecular alignment will provide deterministically polarized photons.
We propose

- To use *chiral* liquid crystal hosts with their 1-D photonic band-gap tuned to the chromophore fluorescence band.
Planar-aligned **cholesteric** liquid crystal has a 1-D photonic band-gap structure

\[ \lambda_0 = n_{av} P_o, \quad \Delta \lambda = \lambda_0 \Delta n/n_{av}, \]

where pitch \( P_o = 2a \) (\( a \) is a period of the structure);

\[ n_{av} = (n_e + n_o)/2; \quad \Delta n = n_e - n_o. \]

The polarizing microscope periodic line pattern is due to the helical structure of the cholesteric phase, with the helical axis in the plane of the substrate*).

*) From I. Dierking
1-D photonic bandgap structures in cholesteric liquid crystals possess an advantage over conventional 1-D micropost technologies which are used with heterostructures containing semiconductor quantum dots.

Because the refractive index $n$ varies gradually rather than abruptly in cholesteric liquid crystals, there are no losses into the waveguide modes, which in the case of micropost technology arise from total internal reflection at the border between two consecutive layers with different $n$. 
We prepared 1-D photonic crystal structures in chiral liquid crystals.

A perspective view of the AFM-topographical image of a planar-aligned layer of this Wacker liquid crystal. The scan was made perpendicular to a planar-aligned surface (1120 nm x 1120 nm scan).

Photograph of the samples with 1-D photonic bandgap chiral liquid crystal structures.
We propose

- To use 2-D and 3-D photonic crystals made of liquid crystals [1];
- 2-D and 3-D photonic crystals [2-3] and/or photonic crystal fibers infiltrated with liquid crystals [4]

We prepared 2-D photonic crystal crystal structures in liquid crystals

Near-field optical image (left) and AFM images (center and right) of 2-D photonic crystal self-assemblies in defects of a planar-aligned glassy cholesteric liquid crystal structures (5 μm x 5 μm scan).
Scanning electron micrographs of the cubic photonic crystal formed in holographic polymer dispersed liquid crystals. (From Reference 1).

Micrograph of the guided modes in a photonic crystal fiber filled with chiral nematic liquid crystal at four different temperatures: green - $T = 77^\circ C$; yellow - $T = 89^\circ C$; nothing – phase transition; blue - $T = 94^\circ C$. The bandgap location sensitivity is $\sim 1$nm/$^\circ C$ and 3nm/$^\circ C$ in visible and infrared region respectively. (From Reference 2).

We are using **nematic** and **cholesteric** liquid crystals in two forms:

- **Low molecular weight**;
- **Oligomer (glassy liquid crystals)**

Molecular structure of University of Rochester (Prof. S.-H. Chen) glassy oligomer **cholesteric** and **nematic** liquid crystals (top) and Wacker glassy oligomer **cholesteric** liquid crystals (bottom).
Glassy nematic liquid crystal layers doped with single dye molecules show good planar alignment (layer thickness is ~ 100nm)
Deterministically polarized fluorescence from single dye molecules doped in planar-aligned glassy nematic liquid crystal host: Experimental set up
Deterministically polarized fluorescence of single dye molecules in glassy **nematic** liquid crystal host

\[ P = \frac{I_{\text{par}} - I_{\text{perp}}}{I_{\text{par}} + I_{\text{perp}}} \]

Perpendicular \hspace{1cm} Parallel

\[ \rho = \frac{I_{\text{par}} - I_{\text{perp}}}{I_{\text{par}} + I_{\text{perp}}} \]

10 μm x 10 μm scan

Molecular structure of DiIC$_{18}(3)$ absorbing and emitting dipoles are parallel to the bridge (perpendicular to two alkyl chains) $^2$.

38 molecules

Spectrofluorimeter measurements of a polarized fluorescence of DiIC$_{18}$(3) dye doped in planar-aligned glassy nematic liquid crystal (~0.5% concentration by weight, 4.1 μm layer thickness).

Dye fluorescence shows polarization anisotropy

\[ P = \frac{I_{\text{par}} - I_{\text{perp}}}{I_{\text{par}} + I_{\text{perp}}} = -0.5 \]
Cholesteric (chiral nematics) liquid crystals: low molecular weight and Wacker glassy oligomers
Selective reflection from photonic band-gap structures of cholesteric liquid crystals with different handedness: Wacker cholesterics (top plot) and low-molecular weight mixture of CB15 and E7 (bottom plots).
Circularly polarized photoluminescence of DiIC$_{18}(3)$ in planar aligned Wacker cholesteric liquid crystals
(~ 0.5% weight concentration, 4.1 μm layer thickness)

Experimental setup for photon antibunching correlation measurements

- **CW laser 532nm**
- **Microscope objectives**
- **Sample**
- **Interference filter**
- **Fiber optical 50:50 nonpolarizing beamsplitter**
- **Avalanche photodiodes**
- **Time-to-digital converter**
- **Start**
- **Stop**
Transmission confocal microscopy setup

- Upper objective
- 532 nm, cw laser
- Avalanche photodiodes
- Fiber optical 50/50 beam splitter
- Lower objective (not visible)
- Interference filters
Antibunching in the fluorescence of single terylene molecule in a Wacker glassy cholesteric liquid crystal host

Left histogram exhibits a dip at $\tau = 0$ indicating photon antibunching in the fluorescence of the single molecule; no antibunching is observed in the right histogram in the fluorescence of the clusters of the molecules.
Source efficiency

The estimated efficiency $p_\alpha$ of our current SPS is $\approx 4\%$, where $2N_{\text{out}} = 0.95N_{\text{inc}} p_\alpha DQ$.

$N_{\text{out}} = 3kc/s; N_{\text{inc}} = 1.2 \times 10^6 \text{photons/s-mol}; D = 0.2; Q = 0.64.$

Source efficiency can be increased up to the value of $\sim 20-40\%$

- by aligning the dye molecules along a direction preferable for maximum excitation efficiency (by $2.6 - 4.3$ times [1])
- by tuning a 1-D photonic-bandgap microcavity of planar-aligned cholesteric liquid crystal to the dye fluorescence band (at least by $2 - 3$ times [2]).

The rate of pulses with two photons

The probability of emission of more than one photon \( P_2 = C_N(0) P_1^2/2 \), if \( P_2 \ll 1 \),

\( P_1 \) is the probability for 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 terylene molecule fluorescence in a Wacker oligomer liquid crystal host \( C_N(0) = g^{(2)}(0) = 0.25 – 0.33 \).

It means that the rate of pulses with two photons is three – four times lower than for Poissonian light.

\[ * \) The source efficiency \( p_\alpha \) introduced earlier, \( p_\alpha = \alpha P_1 \). Here \( \alpha \) is a collection efficiency including losses in filters.
Over the course of more than one hour, no dye bleaching was observed in the oxygen-depleted liquid crystal host (upper curve) in difference with the host without treatment (lower curve).
The main results are as follows:

• Demonstration of a robust SPS based on a single-dye-molecule fluorescence in a liquid crystal host (fluorescence antibunching);

• First demonstration of deterministically polarized single photons from fluorescence emitters;

• Avoided bleaching of the terrylene dye molecules over cw, > 1-hour-excitation by special preparation of liquid crystals.
Future Steps

• Increasing the source outcoupling efficiency, a narrowing of the fluorescence bandwidth (to ~1-10 nm), a decrease in the fluorescent lifetime from a few ns to hundreds of ps by use of various photonic bandgap nanostructures in liquid crystals (1-D in chiral nematics, 2-D/3-D in polymer-dispersed liquid crystals);

• Preparation of tunable SPS using photonic crystals/microstructured fibers infiltrated with liquid crystals;

• Using various fluorescence emitters (dye molecules, colloidal semiconductor quantum dots, rods, carbon nanotubes and rare-earth ions) to extend the working region of the source from the visible to communication wavelengths (1.55 μm).
A confocal microscope with a pulsed laser source was built recently with 75 MHz pulse repetition rate, ~6 ps pulsed-laser excitation at 532 nm.
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