Deterministically polarized single-photon source

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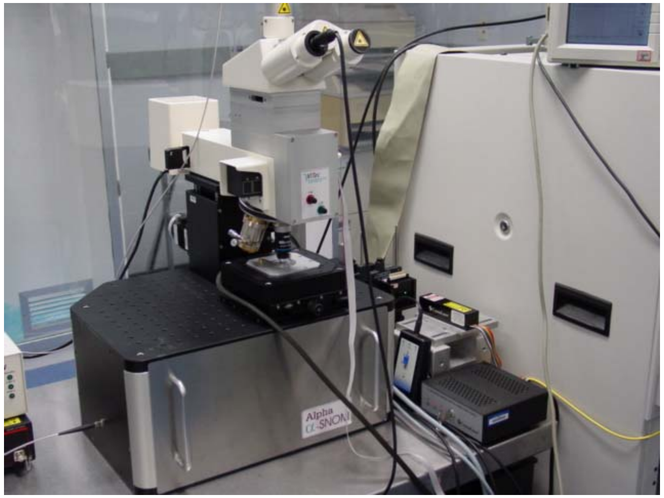
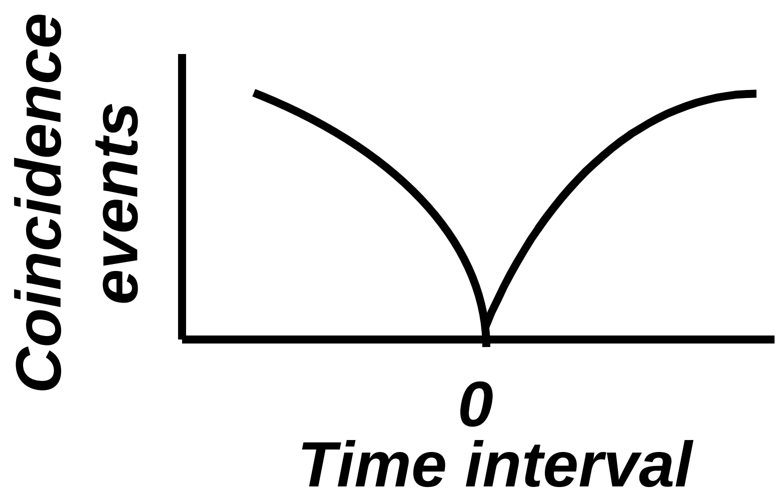


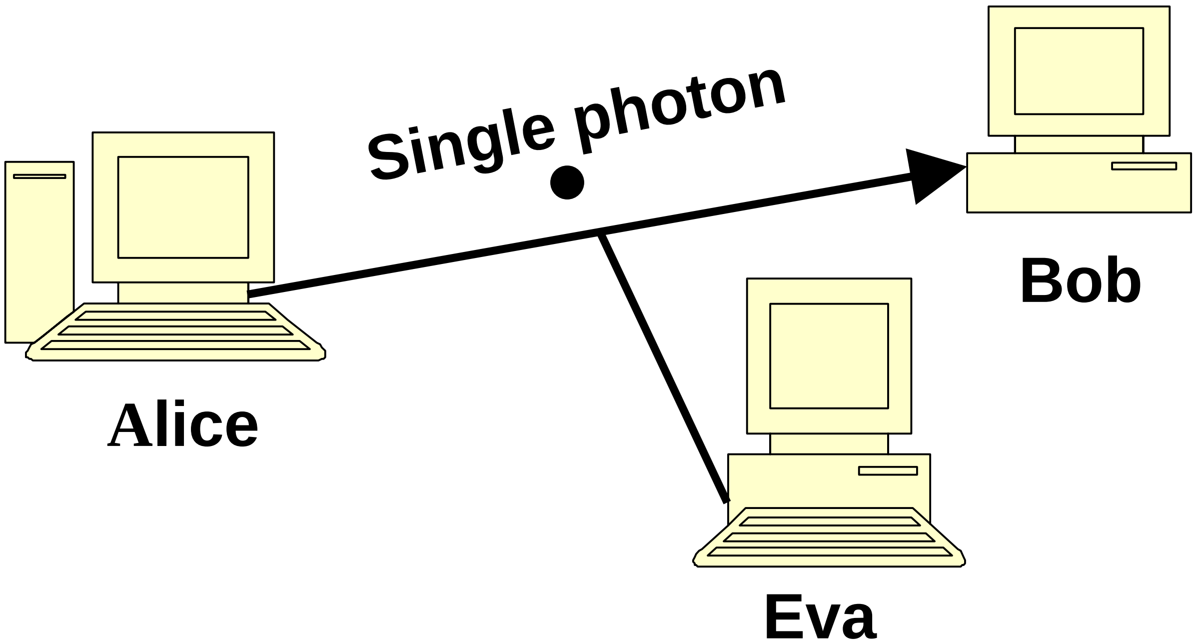
A single-photon source (SPS)

- For single photons, a second order correlation function $g^{(2)}(0) = 0$ indicating the absence of pairs, i.e., antibunching.
- It was proved experimentally, 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. This justifies our assumption that $n(\tau)$ is proportional to the autocorrelation function $g^{(2)}(\tau)$.

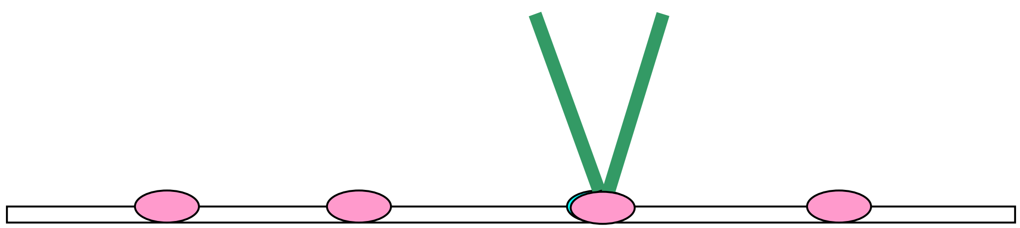


Practical applications of SPSs

- Secure quantum communication;
- Quantum computation with linear optical elements and photodetectors



To produce single photons a laser beam is tightly focused into a sample area containing a very low concentration of emitters, so that only one emitter becomes excited. It emits only one photon at a time.



To date, most SPSs operate only at liquid He temperature.

Room-temperature SPSs:

- **single-dye-molecules;**
- **single color centers in diamond;**
- **colloidal quantum dots**

Current challenges in dye-based SPSs

- **dye bleaching;**
- **low collection and excitation efficiencies;**
- **scattered-photon background;**
- **nondeterministic polarization state of photons**

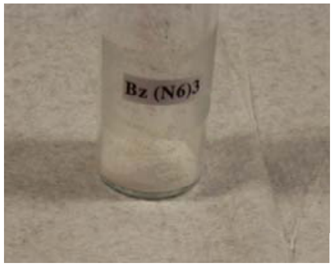
We propose

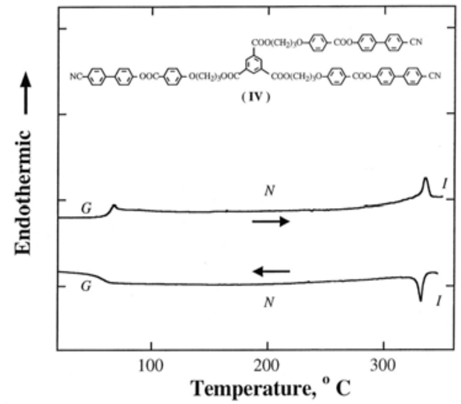
- To use *__liquid crystal__* hosts (including liquid crystal oligomer/polymers) to align the dopant along the direction preferable for excitation efficiency (along the light polarization).

Deterministic molecular alignment will provide <u>deterministically polarized photons.</u>

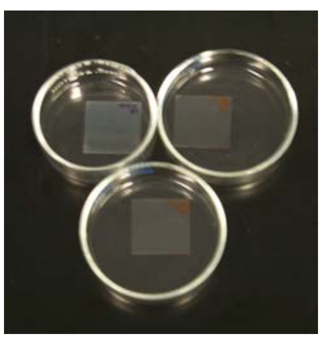
- To use *__chiral__* liquid crystal hosts with their 1-D photonic band-gap tuned to the chromophore fluorescence band.

Polarizing microscope images of S.H. Chen's glassy nematic liquid crystal layers





Photoalignment with polarized UV-light

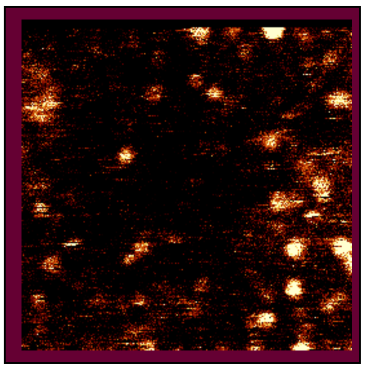


Rotation of 45°

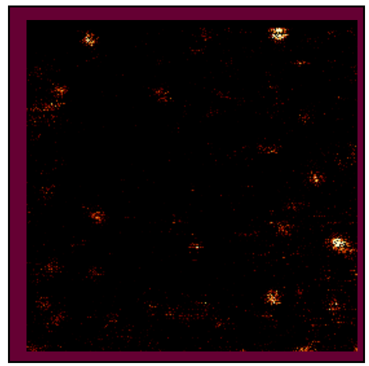
Glassy nematic liquid crystal layers doped with single dye molecules show good planar alignment (layer thickness is ~ 100nm)

Deterministically polarized fluorescence of single dye molecules in glassy nematic liquid crystal host

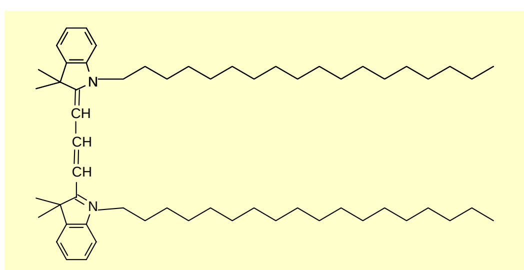
Perpendicular



Parallel



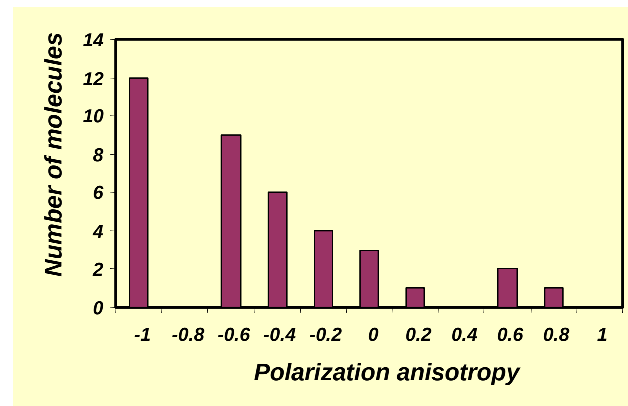
10 μm x 10 μm scan



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

Polarization anisotropy[1)]

$$\rho = (I_{par} - I_{perp})/(I_{par} + I_{perp})$$

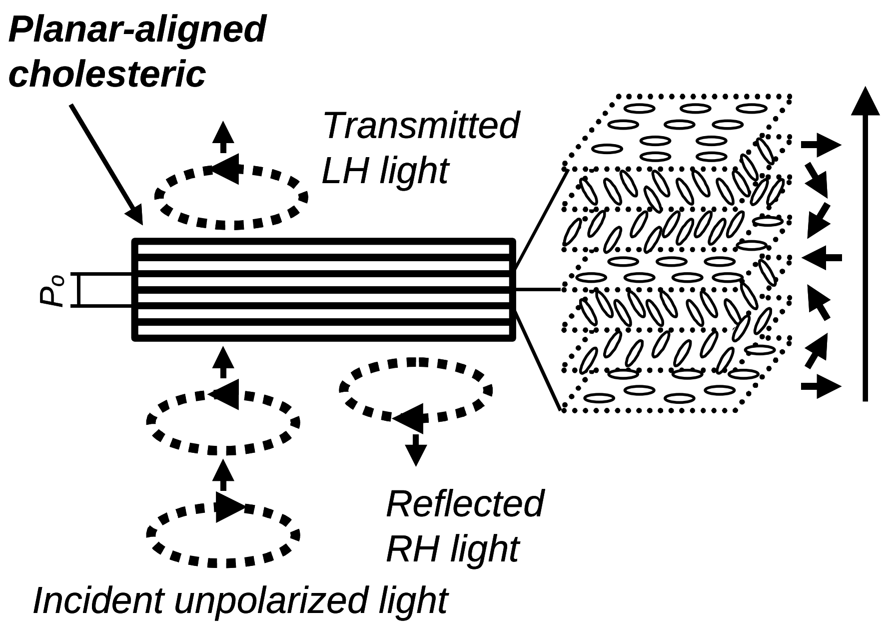


38 molecules

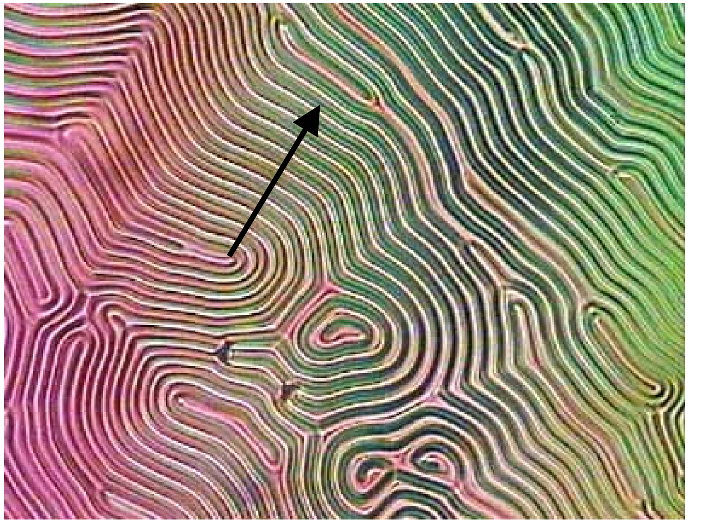
[1)] I. Chung, K.T. Shimizu, M.G. Bawendi, *PNAS,* vol.100, 405 (2003).

[2)] B. Stevens and T. Ha, *J. Chem. Phys.*, vol. 120, 3030 (2004). See also D. Axelrod, *Biophys. J.*, vol. 26, 557 (1979).

Planar-aligned cholesteric liquid crystal has a 1-D photonic band-gap structure

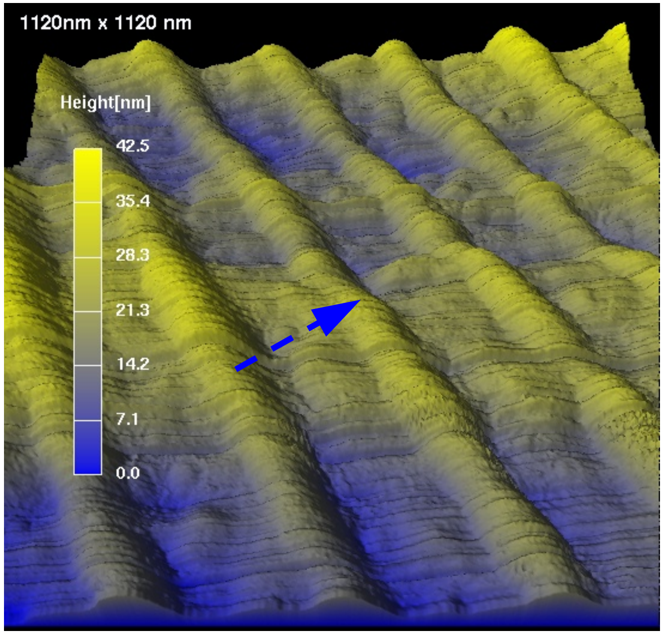


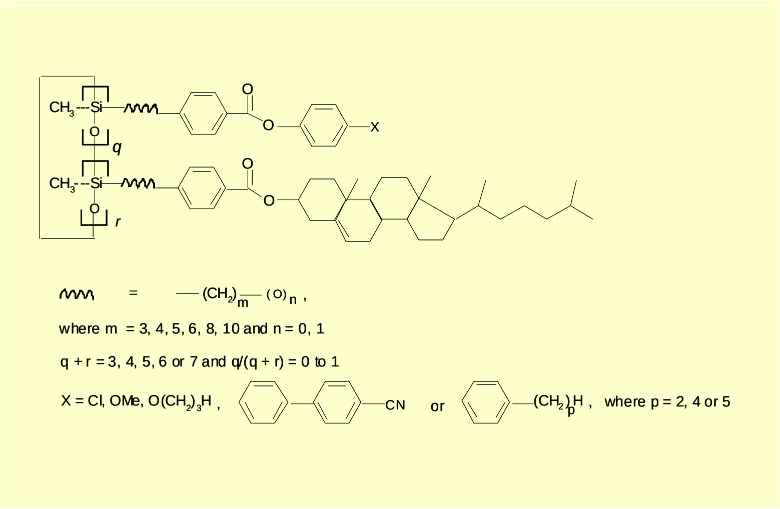
$\lambda_o = n_{av}P_o$, $\Delta\lambda = \lambda_o\Delta n/n_{av}$,
where pitch $P_o = 2a$
(a is a period of the structure);
$n_{av} = (n_e + n_o)/2$; $\Delta n = n_e - n_o$.



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

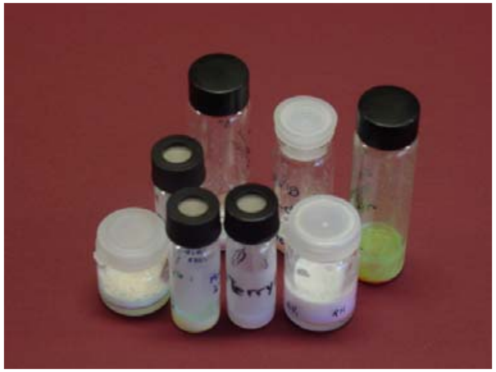


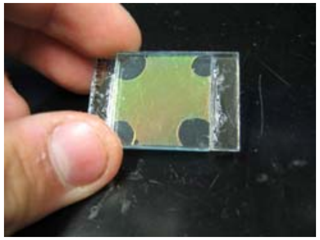


Perspective view of the AFM-topographical image of a planar-aligned Wacker OCLC (1120 nm x 1120 nm scan).

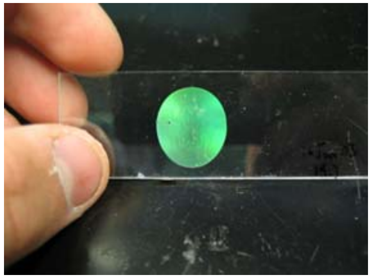
Molecular structure of Wacker siloxane oligomer cholesteric liquid crystal (OCLC)

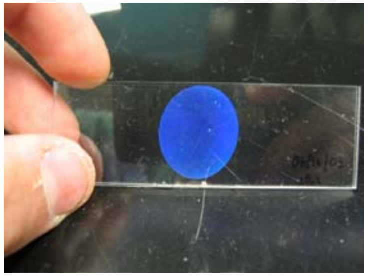
Samples prepared from the low molecular weight liquid crystals

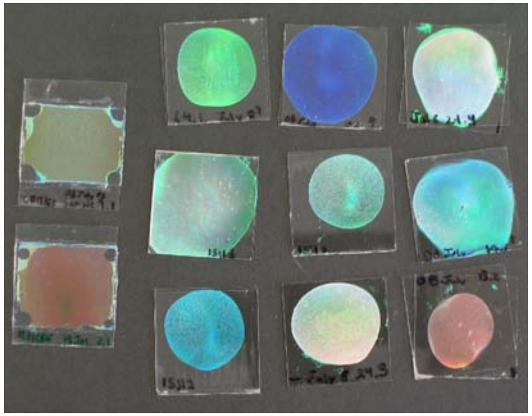




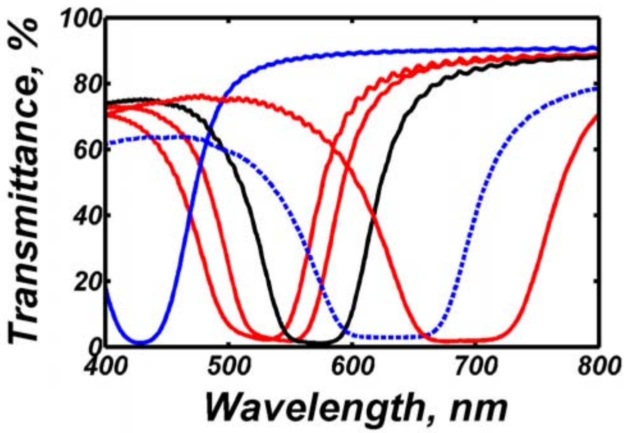
Samples prepared from Wacker oligomer liquid crystals





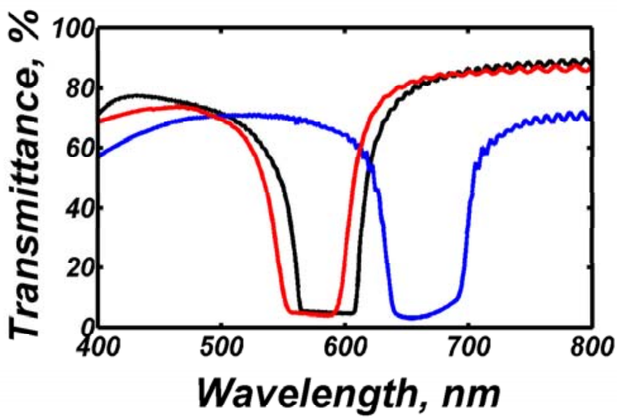


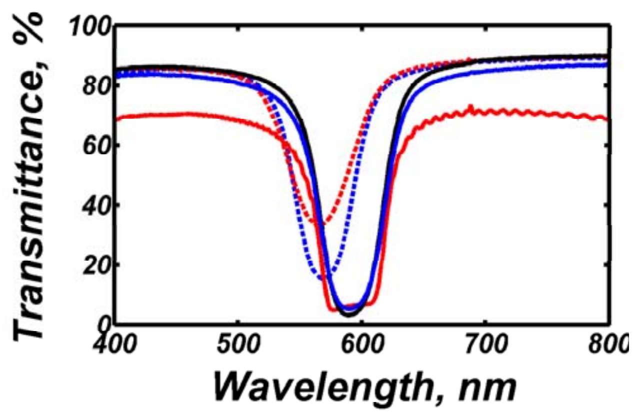
Selective reflection from photonic band-gap structures of Wacker OCLCs (top plot) and low-molecular weight mixture of CB15 and E7 (bottom plots)



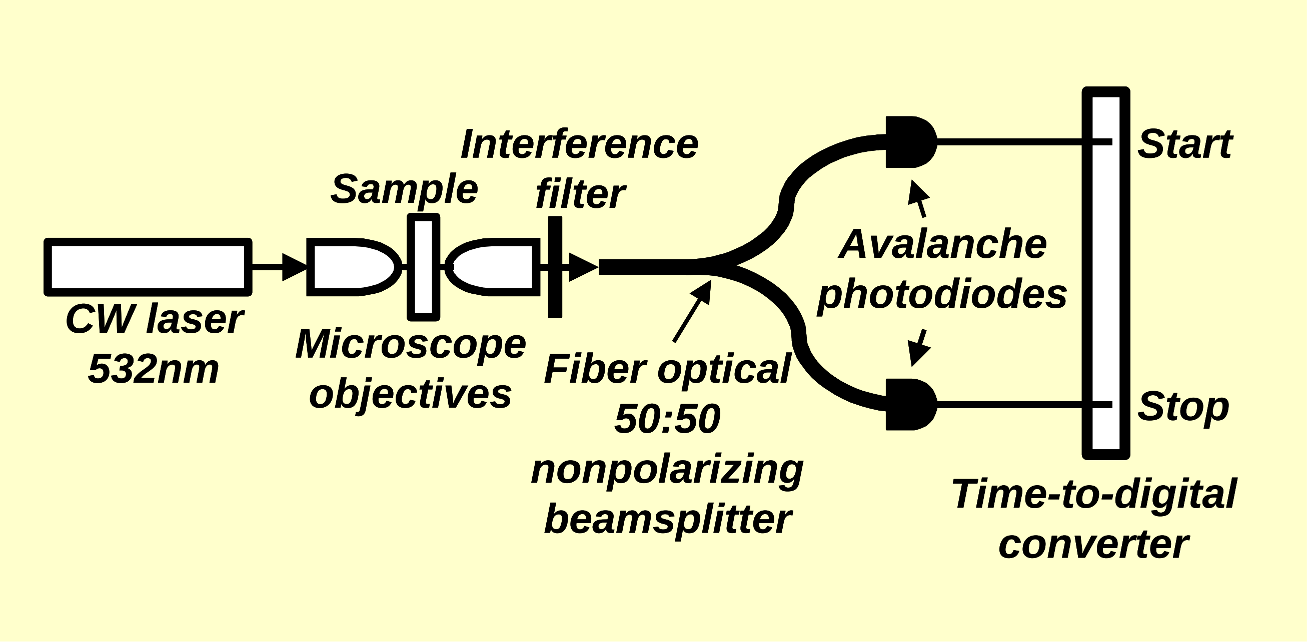
Left-handed circularly polarized light

Right-handed circularly polarized light



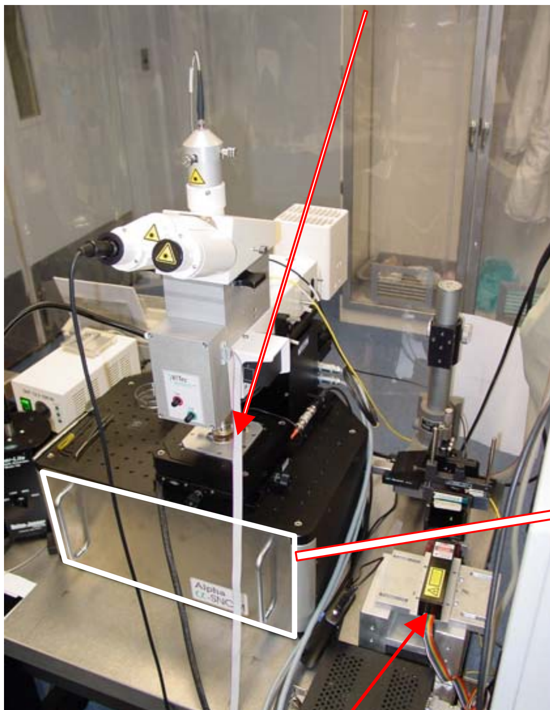


Experimental setup for photon antibunching correlation measurements



Transmission confocal microscopy setup

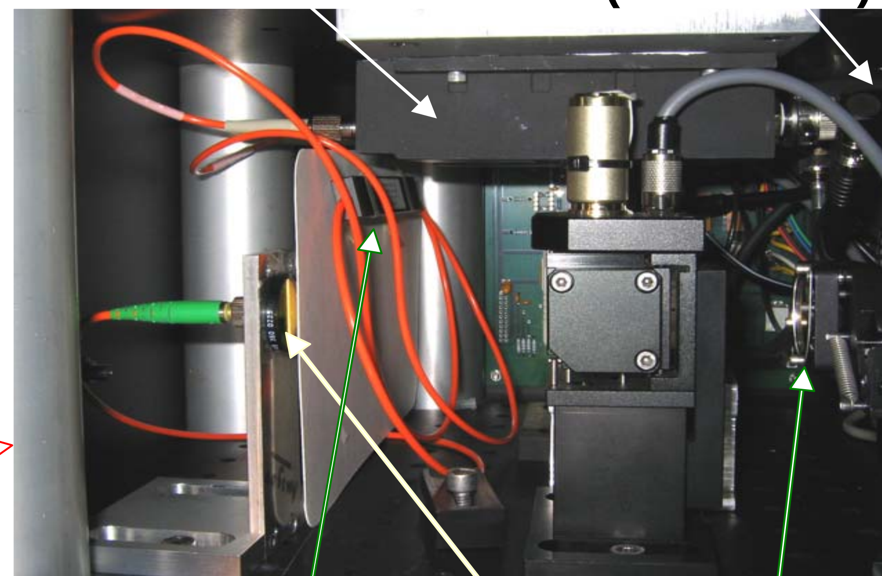
Upper objective



532 nm, cw laser

Avalanche photodiodes

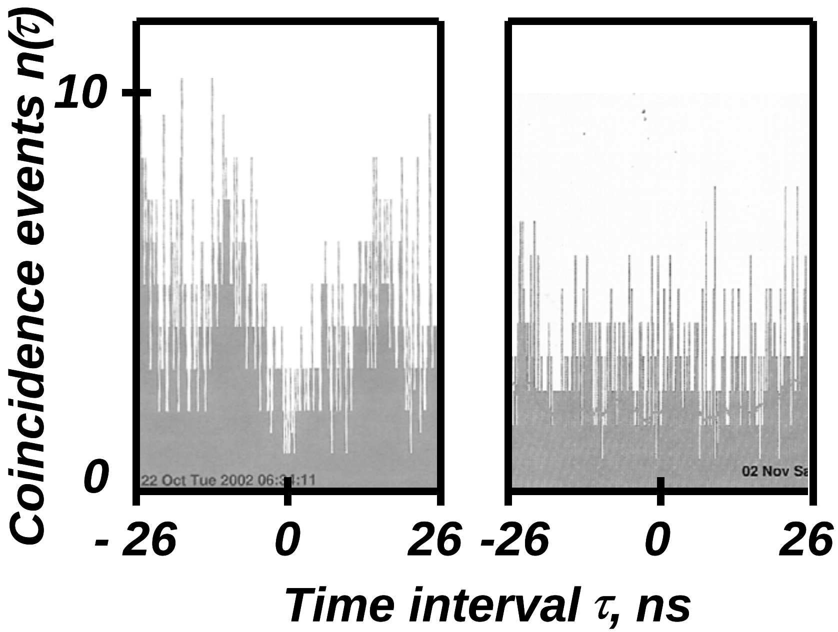
Lower objective (not visible)



Fiber optical 50/50 beam splitter

Interference filters

The histograms of coincidence events of single-terrylene-molecule-fluorescence in a Wacker OCLC host (left) and of an assembly of several uncorrelated molecules (right).



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.

Source efficiency

The estimated efficiency p_α of our current SPS is $\approx$ 4%,

where $2N_{out} = 0.95 N_{inc} p_a DQ$.

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

Source efficiency can be increased up to ~ 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]).**

[1] M. Croci, H.-J. Müschenborn, U.P. Wild, p. 73, in Th. Basché, W.E. Moerner, M. Orrit, U.P. Wild (Eds), *Single molecule optical detection, imaging and spectroscopy,* Weinheim: VCH, 1997, 250 pp.

[2] S.C. Kitson, P. Jonsson, J.G. Rarity, and P.R. Tapster, *Phys. Rev. A*, vol. 58, pp. 620 –627, 1998.

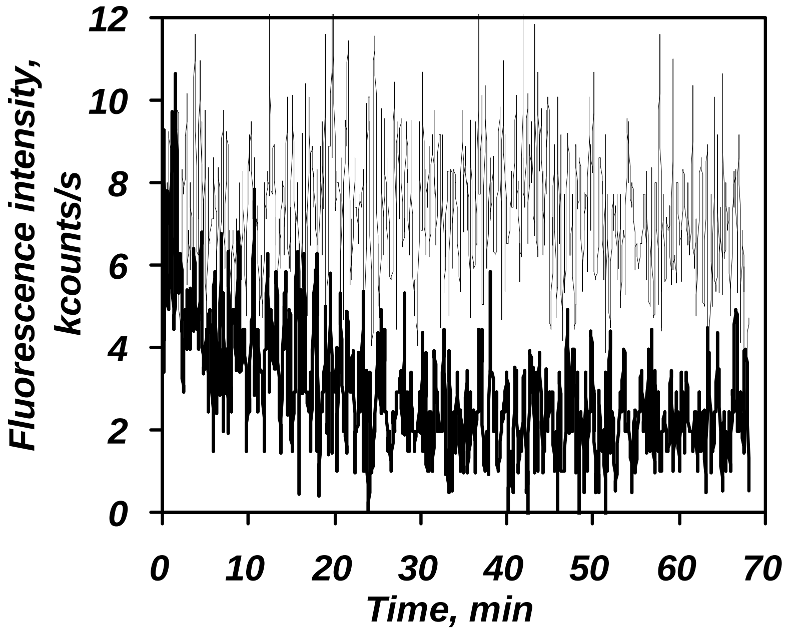
Probability of two-photon emission

The probability of two-photon emission $P_2 = C_N(0)\, P_1^2/2$, 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 terrylene 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 two-photon pulses is three – four times lower than for Poissonian light.

*) The source efficiency p_α introduced earlier, $p_\alpha = \alpha P_1$. Here α is a collection efficiency including losses in filters.

Preventing terrylene dye bleaching in liquid crystal host



Over the course of more than one hour, no dye bleaching was observed in *the oxygen - depleted* liquid crystal host (upper curve).

Summary

The main results are as follows:

- **First demonstration of a robust SPS based on a single-dye-molecule fluorescence in a *<u>liquid crystal host</u>* (fluorescence antibunching);**

- **First demonstration of deterministically polarized single photons from fluorescence emitters;**

- **Avoided bleaching of the terrylene dye molecules over > 1-hour-excitation by special preparation of liquid crystals;**

- **Preparation of different planar-aligned 1-D photonic band-gap structures with bandgaps from 400 to 2200 nm in dye-doped cholesteric liquid crystal oligomers and low molecular weight liquid crystals.**

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