Dye-doped cholesteric-liquid-crystal single photon source

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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.**
- **Among known *room-temperature* SPSs, only those based on single-dye-molecule fluorescence can be used *in high-speed* systems (~ 100 MHz repetition rate operation).**
- **Alternatives such as color centers in diamond and colloidal quantum dots possess too long fluorescence lifetimes.**

In dye-based SPSs*), current challenges are as follows:

(1) dye bleaching,

(2) low collection and excitation efficiencies;

(3) scattered-photon background;

(4) nondeterministic polarization state of photons.

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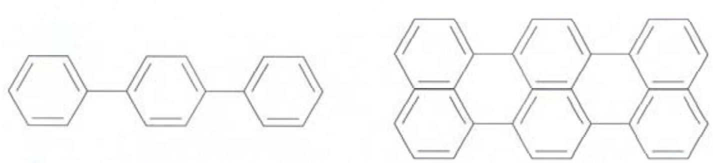
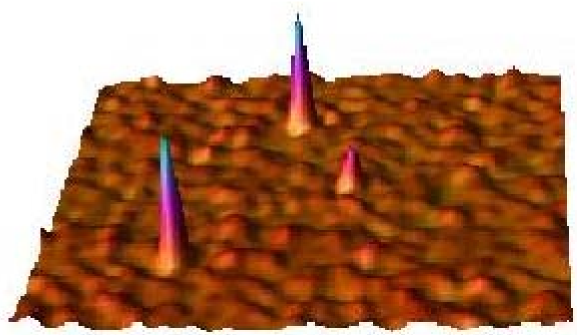
*) 1. W. P. Ambrose et al., *Chem. Phys. Lett.*, **269**, 365 (1997).

2. B. Lounis and W.E. Moerner, *Nature*, **407**, 491 (2000).
3. L. Fleury et al., *Phys. Rev. Lett.*, **84**, 1148 (2000).
4. F. Treussart et al., *Opt. Lett.*, **26**, 1504, 2001.

B. Lounis and W.E. Moerner *(Nature*, 407, 491, 2000) spurred new interest in dye-based SPSs on demand:

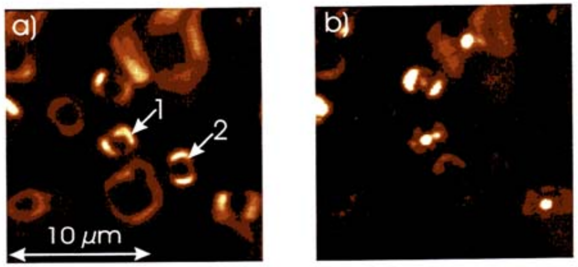
<u>Terrylene</u> dye molecules did not bleach during several hours of *<u>room temperature,</u>* pulsed excitation of <u>p-terphenyl</u> microcrystal flakes doped with terrylene.





Restrictions on practical use of terrylene/p-terphenyl SPS:

- source fragility and stress-sensitivity;
- poor efficiency (Poor match between molecular dipole orientation and light polarization);
- 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.*



P-terphenyl microcrystal flakes doped with terrylene (from J. Michaelis, *Ph.D. Thesis*, Konstanz, 2000).

Noncrystalline, amorphous hosts, e.g., polymers, do not:
(1) provide spectral stability of single molecule emission even in the case of terrylene, and
(2) provide long time (e.g., several hours) emission of excited molecule without bleaching.

On the other hand, no crystal hosts other than fragile sublimated p-terphenyl flakes are known to be proposed in single-molecule *room-temperature* experiments.

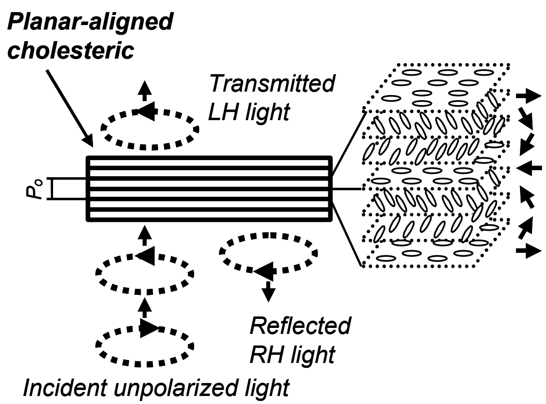
T We propose

- ❖ 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 deterministically polarized photons.

We propose

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

Chiral nematic (cholesteric) liquid crystals



For cholesteric planar layers > 10μm, the reflectance of normally incident, circularly polarized light with electric-field vector-rotation opposite to the rotation of molecules in the helical structure, approaches 100% within a band centered at $\lambda_o = n_{av}P_o$ 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 $\Delta\lambda = \lambda_o \Delta n/n_{av}$, where $\Delta n = n_e - n_o$.

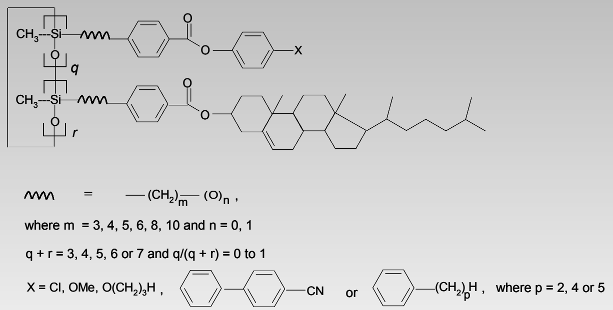
The polarizing microscope line pattern is due to the helical structure of the cholesteric phase, with the helical axis in the plane of the substrate*).

*) From I. Dierking "Textures of Liquid Crystals"(Wiley-VCH, 2003).

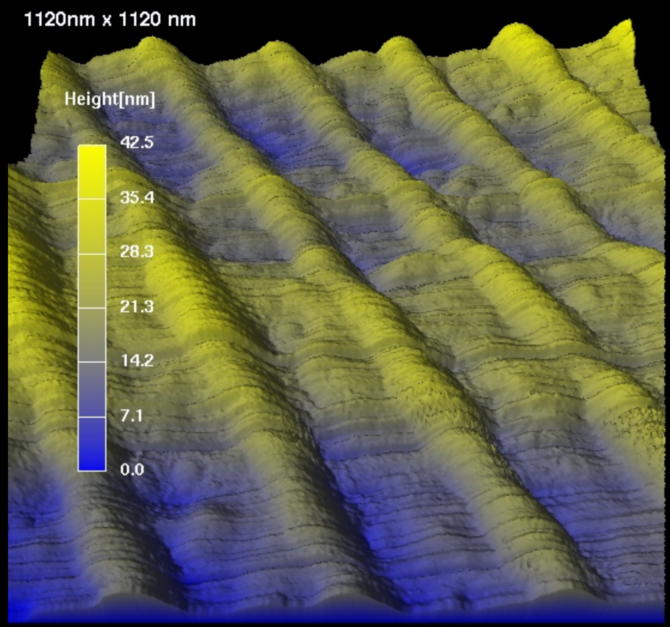
We are using cholesteric liquid crystals in two forms:

(1) Low molecular weight (fluid-like);

(2) Oligomer

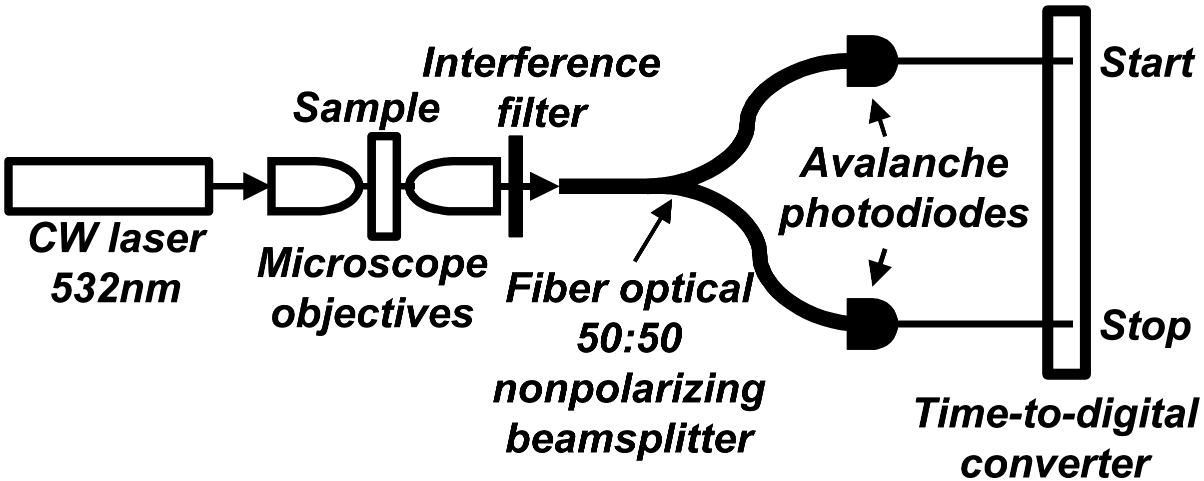


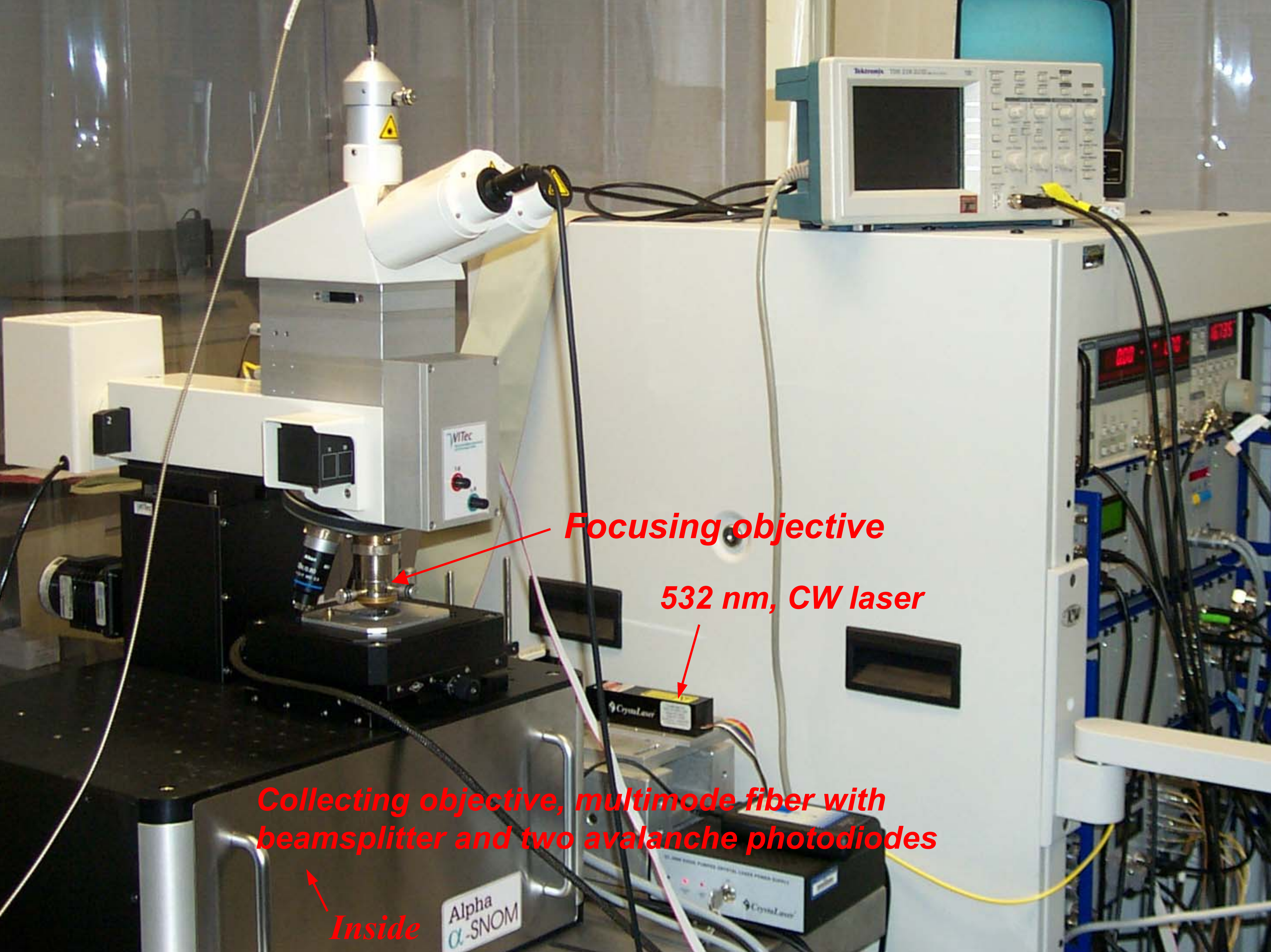
Molecular structure of Wacker siloxane oligomer cholesteric liquid crystal (OCLC)

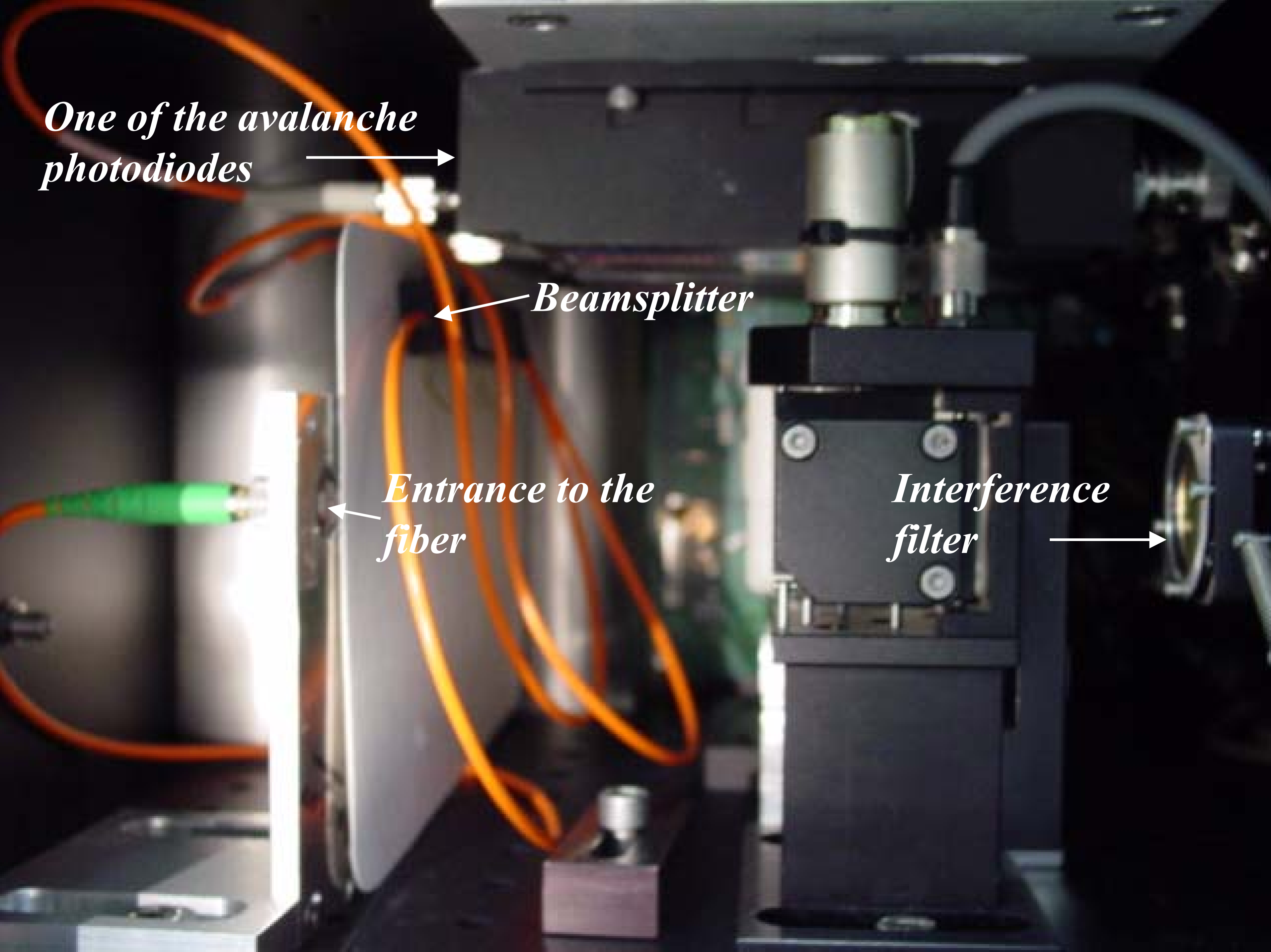


Perspective view of the AFM-topographical image of a planar-aligned Wacker OCLC . 1120 nm x 1120 nm scan.

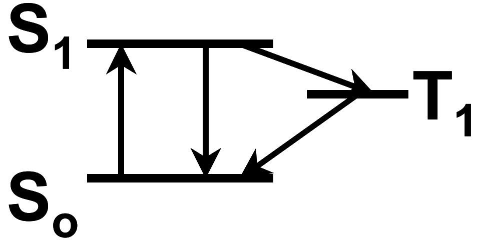
Experimental setup

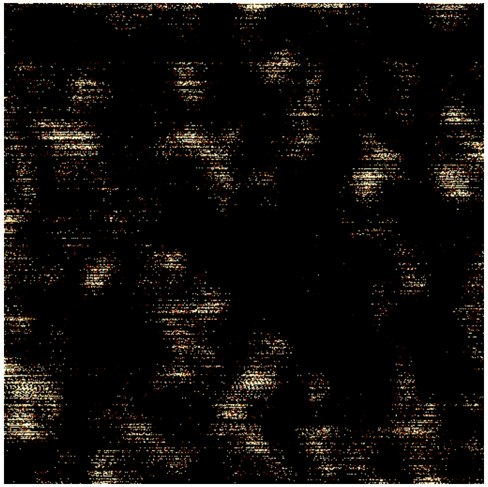



Focusing objective
532 nm, CW laser
Collecting objective, multimode fiber with beamsplitter and two avalanche photodiodes
Inside
Alpha α-SNOM


One of the avalanche photodiodes
Beamsplitter
Entrance to the fiber
Interference filter

Confocal fluorescence microscopy of terrylene-dye molecules in a Wacker OCLC host. *Single molecules* show "blinking" behavior.

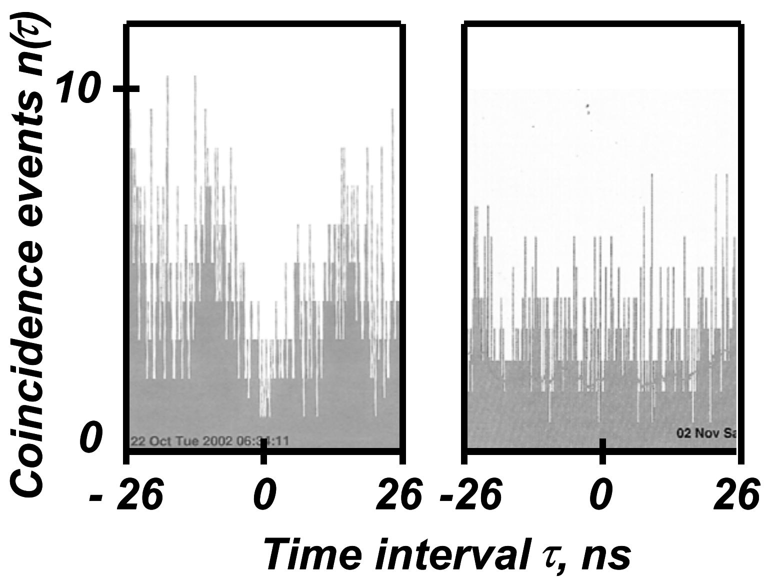




10 μm x 10 μm

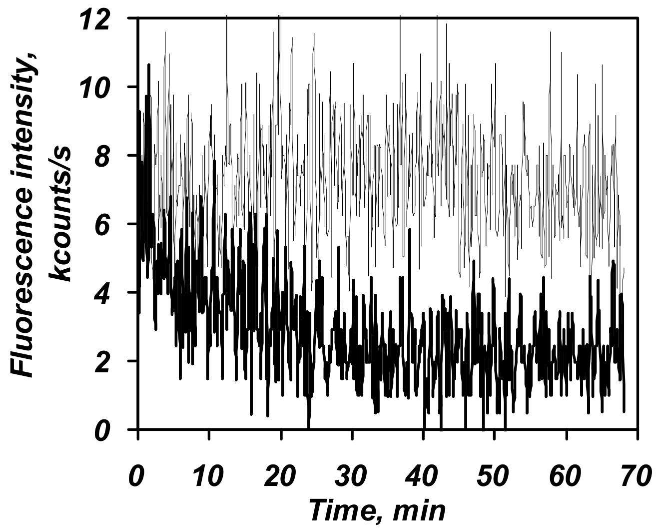
Resolution of optical system is ~ 0.5μm

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.

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 and Future Steps:

The main results are as follows:

- Demonstration of a robust single photon source based on fluorescence from a single-dye-molecule in a ***liquid crystal host*** (fluorescence antibunching);
- Avoiding bleaching of the terrylene dye molecules over > 1-hour-excitation by special preparation of liquid crystals;
- Preparation of planar-aligned 1-D photonic band-gap structures in dye-doped cholesteric liquid crystal oligomer.

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 single-photon source on demand.

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