Direct evidence of the flexomagnetoelectric effect revealed by single molecule spectroscopy

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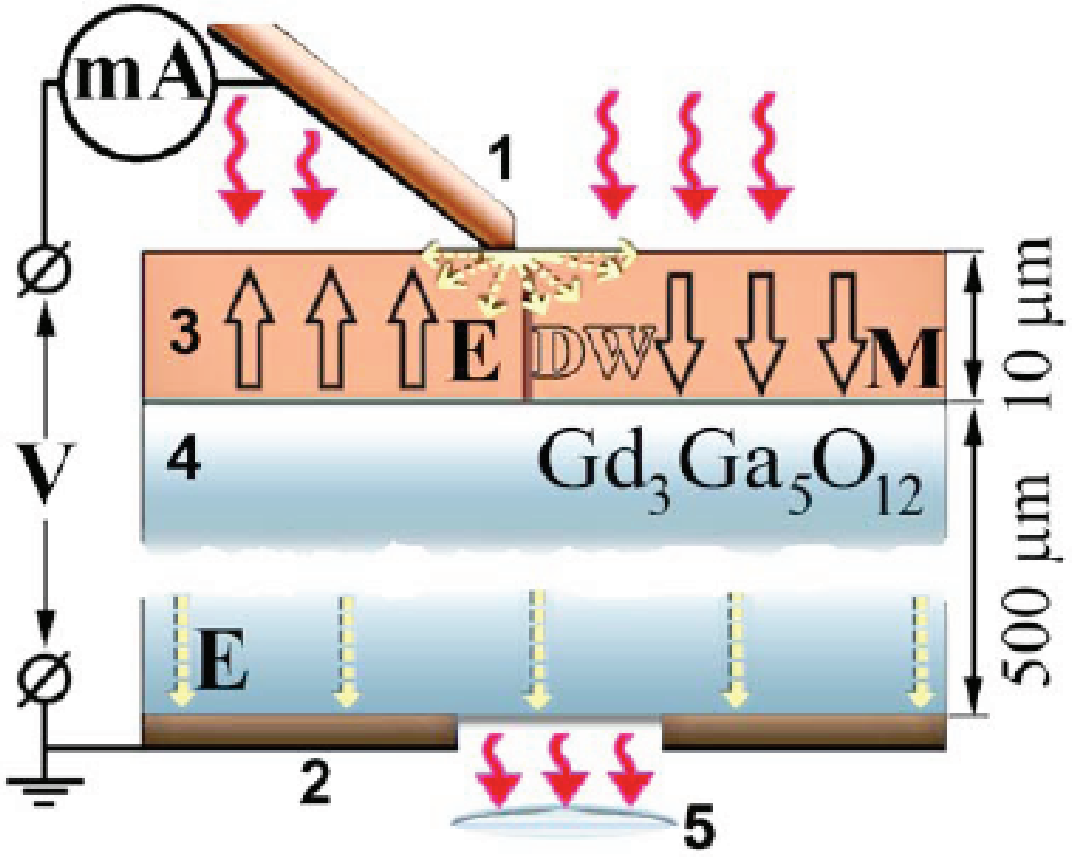
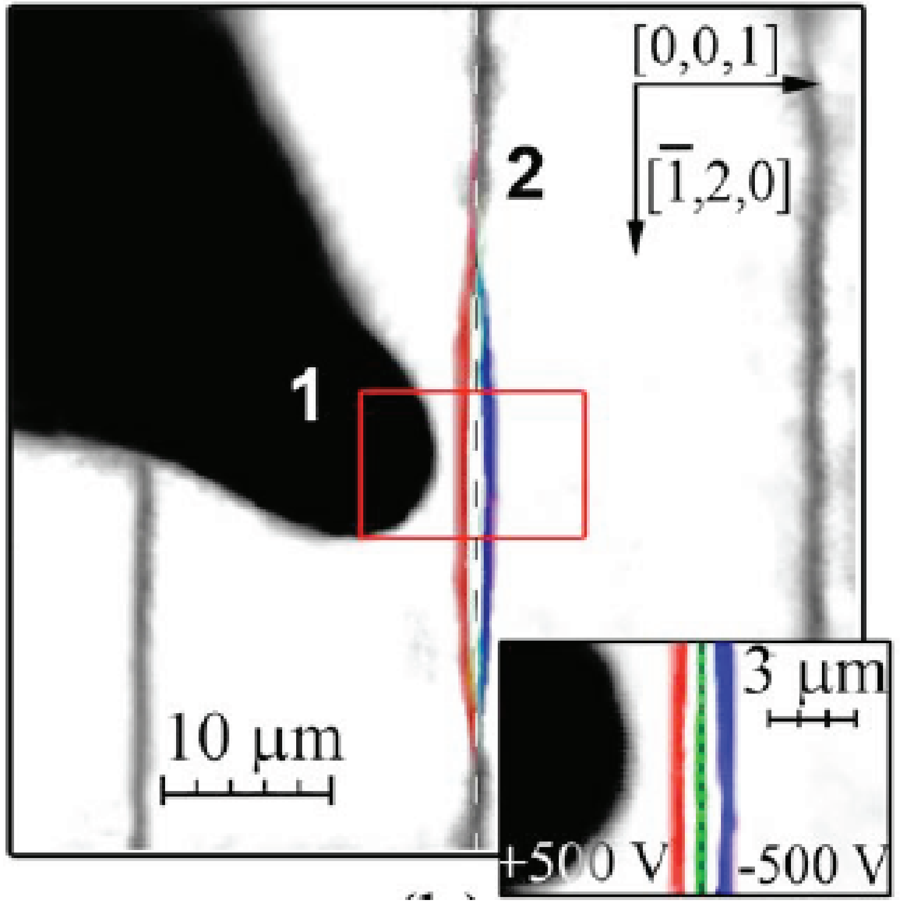
The Magnetoelectric effect

Manipulation of domain walls in iron garnet by electric field E~ 1 MV/m





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Room temperature magnetoelectric control of micromagnetic structure in iron garnet films

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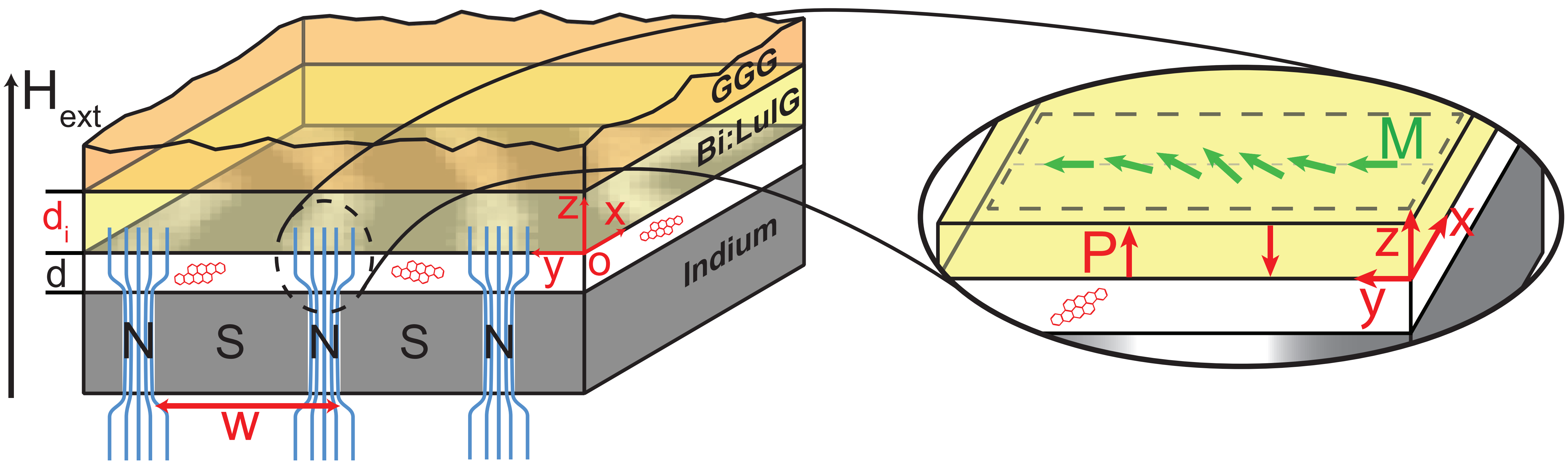
The Flexomagnetoelectric effect

Non uniform magnetization generates electric polarization in systems with broken inversion symmetry :

Bar'yakhtar, V.G. et al. JETP Lett. 37, 673 (1983).

$$\vec{P} = \gamma \chi_e \left[\left(\vec{M} \cdot \vec{\nabla} \right) \vec{M} - \vec{M} \left(\vec{\nabla} \cdot \vec{M} \right) \right]$$



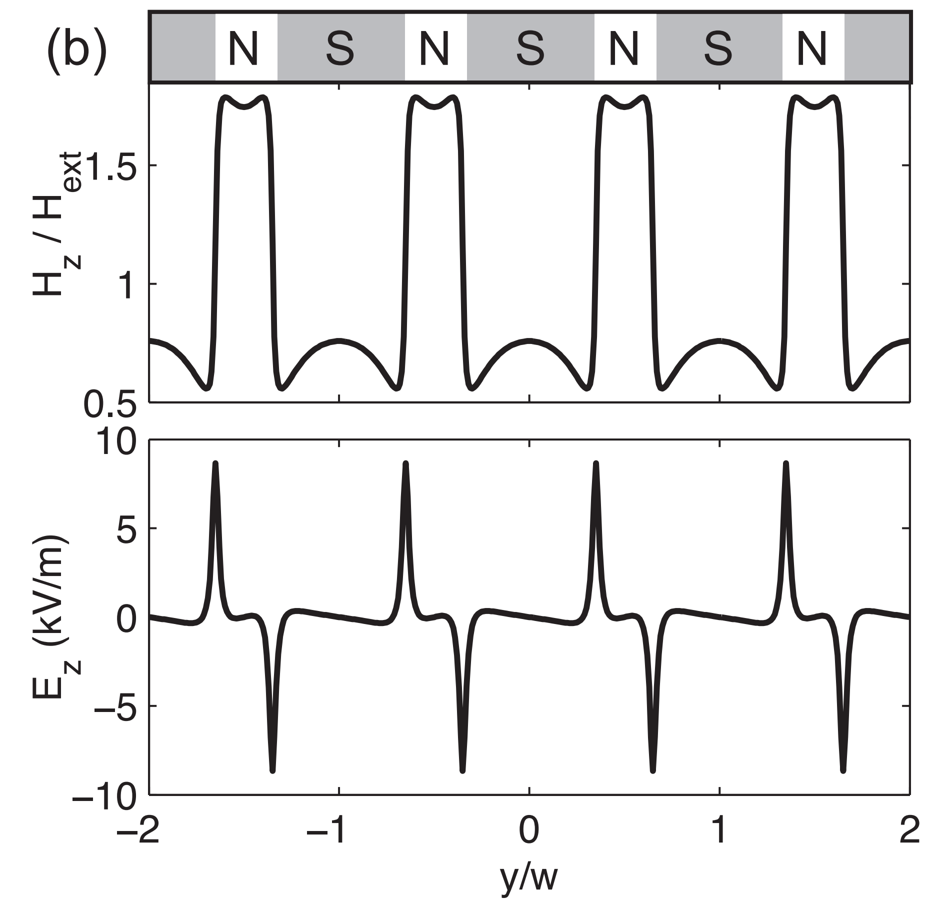
Sample :

- Indium thickness : 1.6 µm
- Molecular film of hexadecane with DBATT fluorescent molecules
- Bi:LuIG layer d_i=2.5 µm

Electric field of the iron garnet

Periodic superconducting domains with magnetization profile :

$$4\pi M_z = \begin{cases} -H_{\text{ext}} & \text{S region} \\ H_{\text{ext}}(d_S/d_N) & \text{N region} \end{cases}$$



Maxwell equations with origin at the surface of the superconductor in the middle of S domain :

$$H_z = H_{\text{ext}} + \sum_{n=1}^{\infty} H_n(z)\cos(q_n y) \qquad H_n(z) = -\frac{2H_{\text{ext}}}{q_n d_N}\left(1 - e^{-q_n D}\right) e^{-q_n z} \sin\left(\frac{q_n d_S}{2}\right)$$

Electric field induced by the polarization P(y,z) in the molecular layer :

$$E_z = \sum_{n=1}^{\infty} E_n(z)\sin(q_n y) \qquad E_n(z) = \frac{4\pi\gamma\chi_e\chi_m M_S H_c}{w}\left(1 - e^{-q_n D}\right)\left(1 - e^{-2q_n d_i}\right) e^{-q_n(2d-z)} \sin\left(\frac{q_n d_S}{2}\right)$$

$E_z \sim 10$ -100 kV/m expected

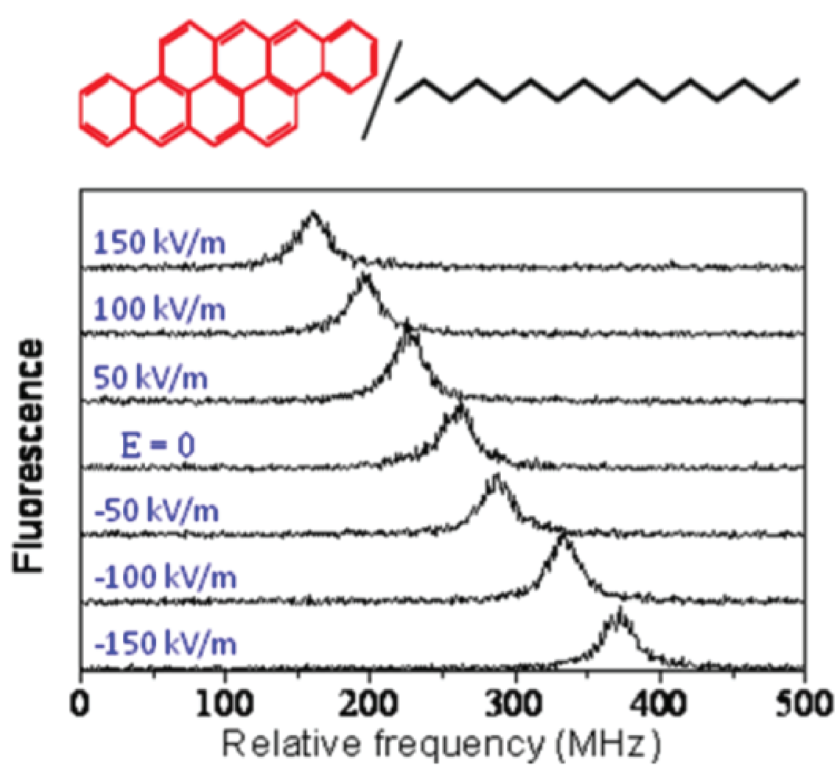
Single molecules as local electric field probe

Properties of single molecules :

- Lifetime-limited optical line width (quality factor 10^8)
- Linear Stark effect δp~0.3 Debyes

$$\Delta\nu = -\frac{1}{h}\left(\delta\vec{p}\cdot\vec{E}_{\text{loc}} + \frac{1}{2}\vec{E}_{\text{loc}}\cdot\delta\vec{\alpha}\cdot\vec{E}_{\text{loc}}\right)$$

- Optical resonance insensitive to moderate magnetic fields (singlet states)
- Molecules are trapped in a thin crystalline matrix

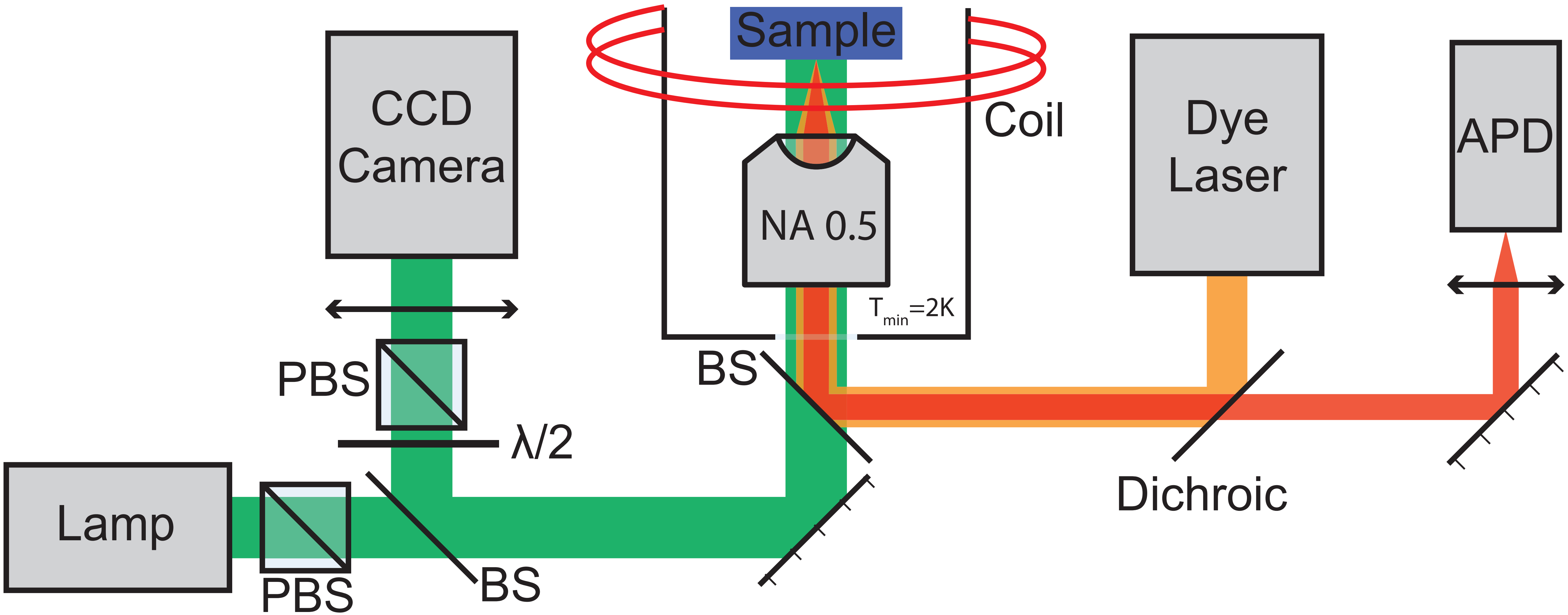


ΔE~10 kV/m will shift ZPL by ~20 MHz

Brunel, Ch. et al. The Journal of Physical Chemistry A 103, no. 14 (1999)

Experimental setup

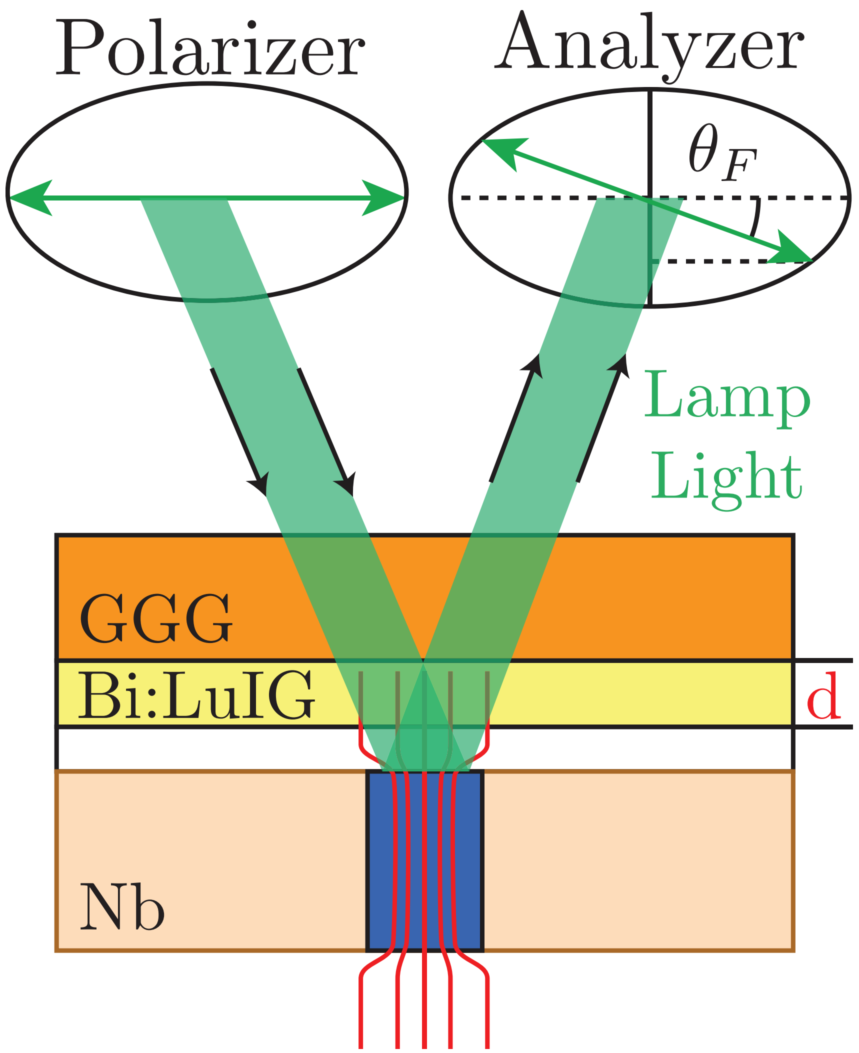
Sample in liquid helium cryostat. $T<T_c$ with $H_{ext}\neq 0$

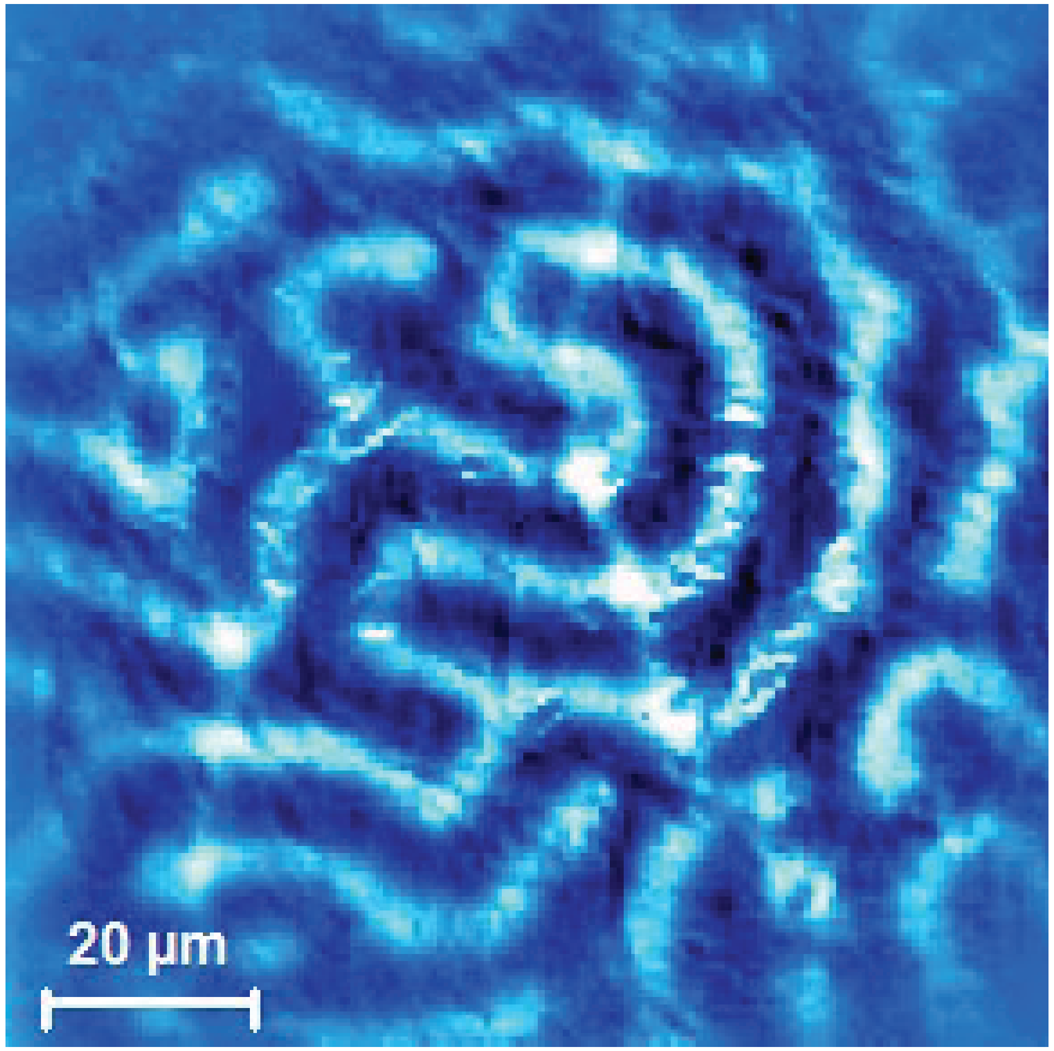


MO imaging setup combined to Single Molecule Spectroscopy

MO image of Indium surface

MO image of Indium with H_{ext}=30 Oe and T=2.6 K



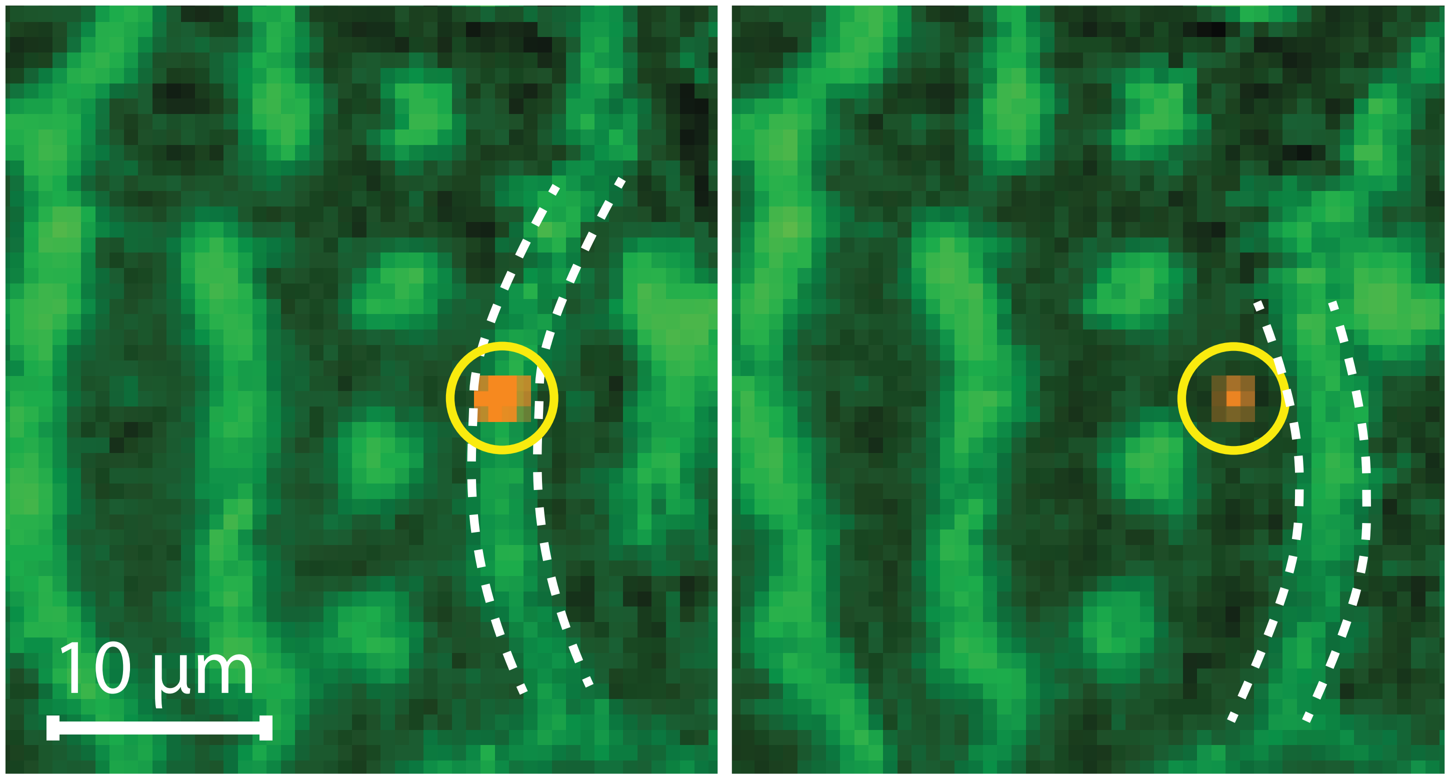


Rotation of the polarization by Faraday effect through a material with Verdet constant V :

$$\theta_F = 2VB_z d \qquad C = \left| \frac{-4Vd(\Phi - \theta_F}{(\Phi - \theta_F)^2 + \varepsilon} \right| = 0.65$$

Probing electric field with a single molecule

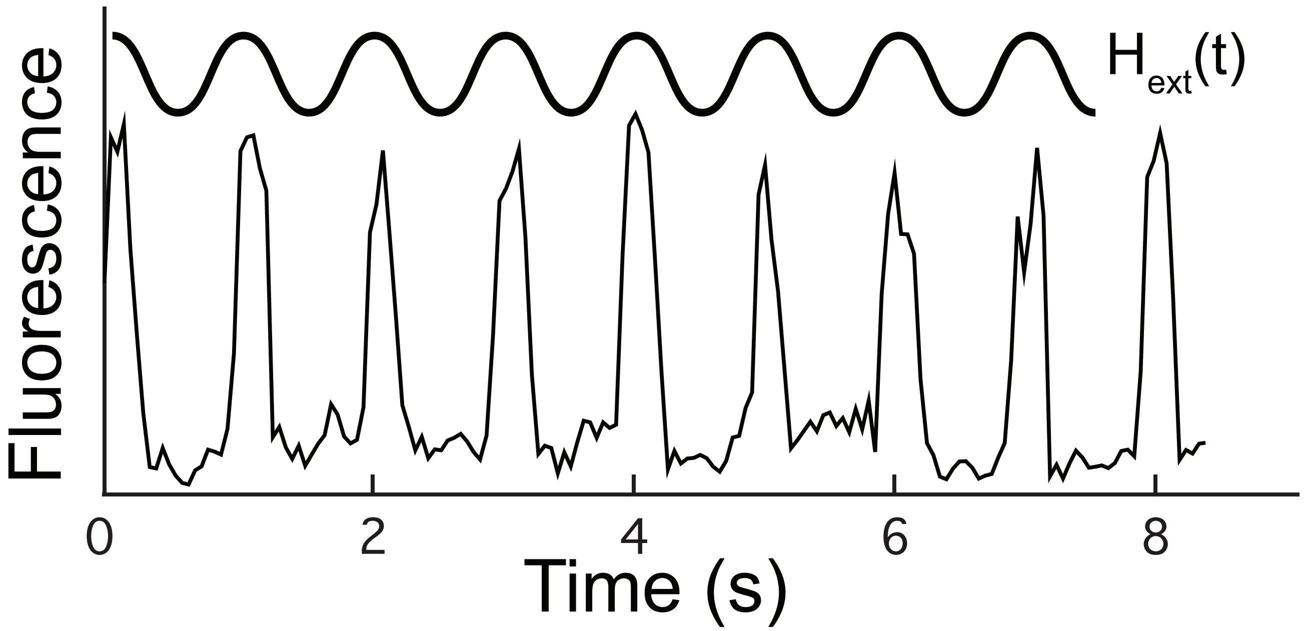
Modulation of H_{ext}



- f_{mod}=1 Hz
- Amplitude 2 Oe aroud 30 Oe

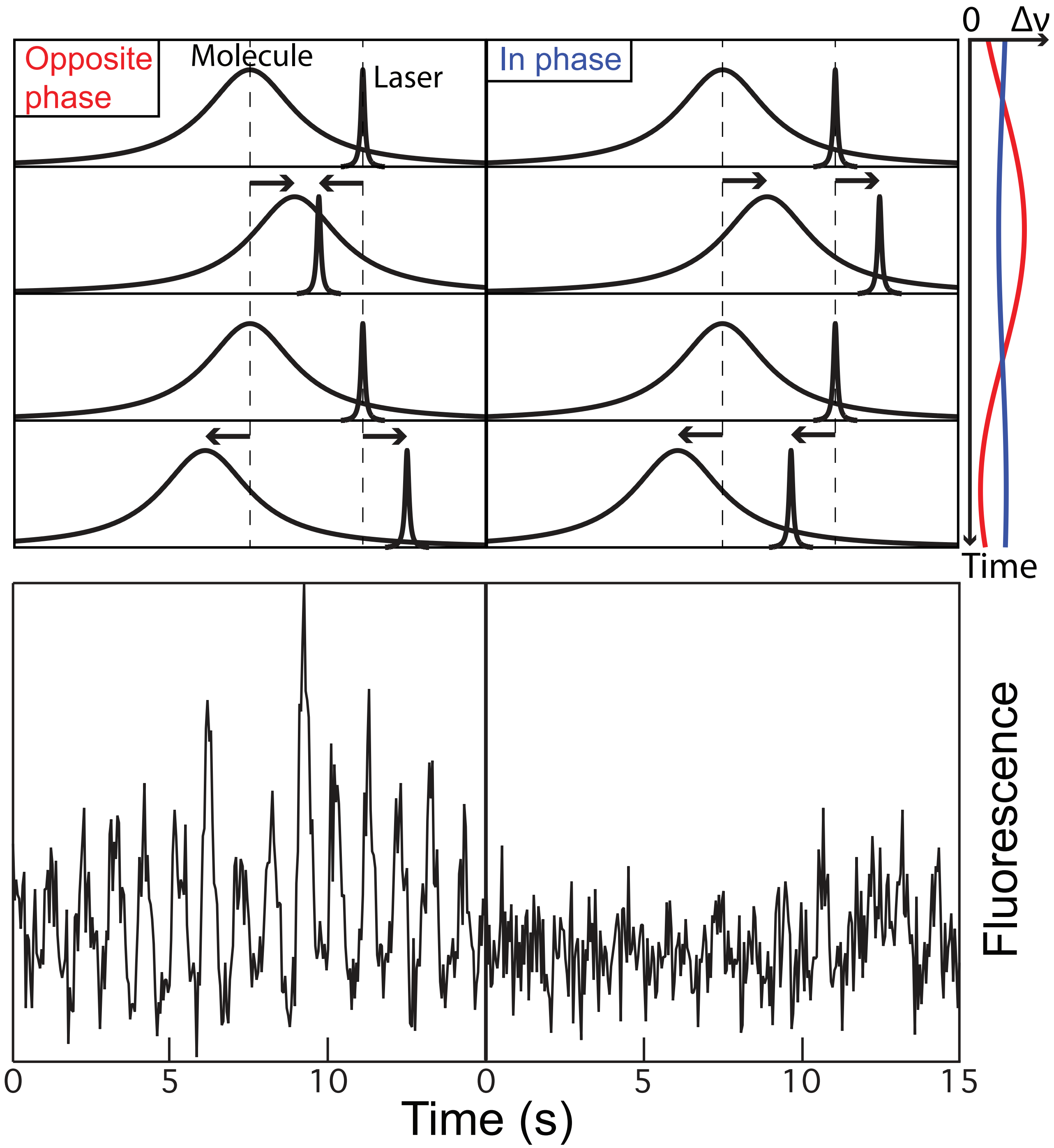
Stark shift ~50 MHz with Stark coefficient 0.8 GHz/(MV/m)

→ E_z~60 kV/m



Modulation of laser frequency and magnetic field

Fluorescence depends on detuning between laser frequency and resonance frequency



$$\delta(t) = \delta_0 + A_L \cos(\omega_{\mathrm{mod}} t + \phi) - A_M \cos(\omega_{\mathrm{mod}} t)$$

- Opposite phase : high modulation amplitude

$$A_L + A_M$$

- In phase : weak modulation amplitude

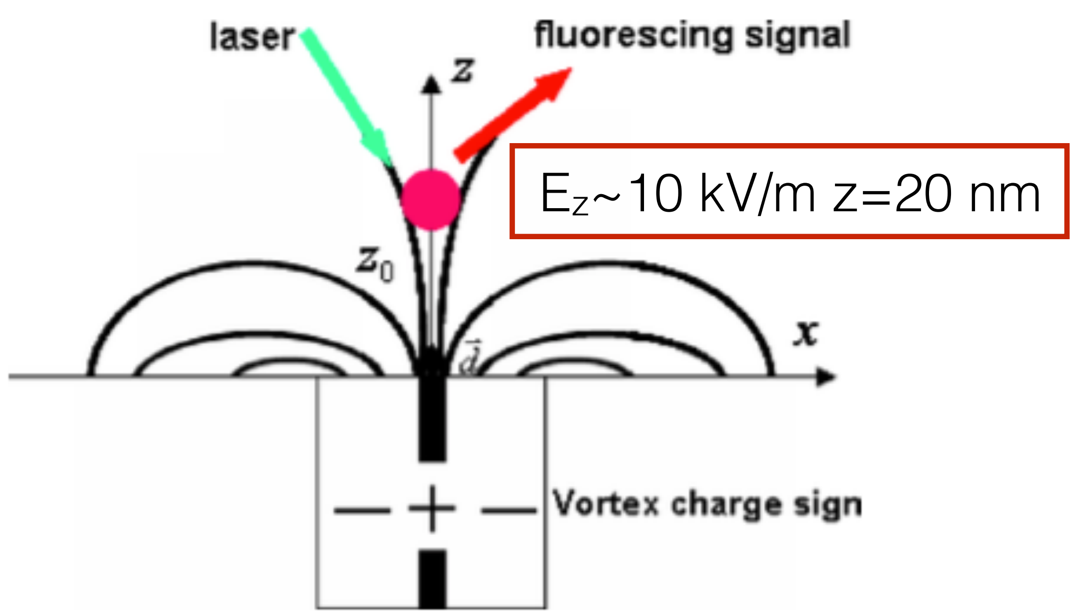
$$|A_L - A_M|$$

Stark shift ~40 MHz with Stark coefficient 1 GHz/(MV/m)

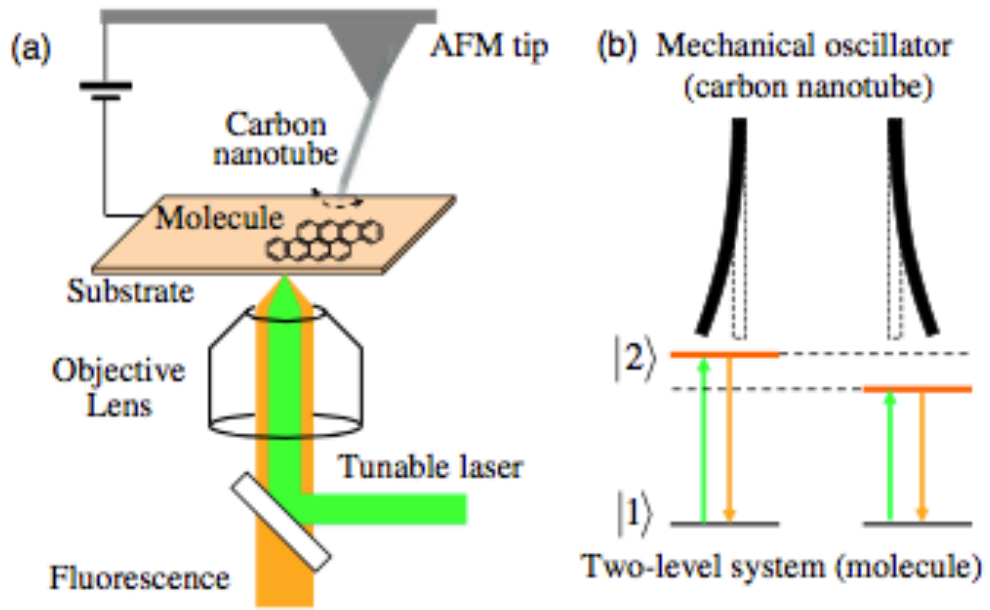
→ E_Z~40 kV/m

Conclusion & perspectives

- Single molecule spectroscopy is used to evidence flexomagnetoelectric effect
- Local electric field variations : from 4 kV/m up to 60 kV/m probed with Stark shift of molecules
- Perspective : use single molecules to probe the charge of the vortex core or motion of charged nano mechanical oscillator



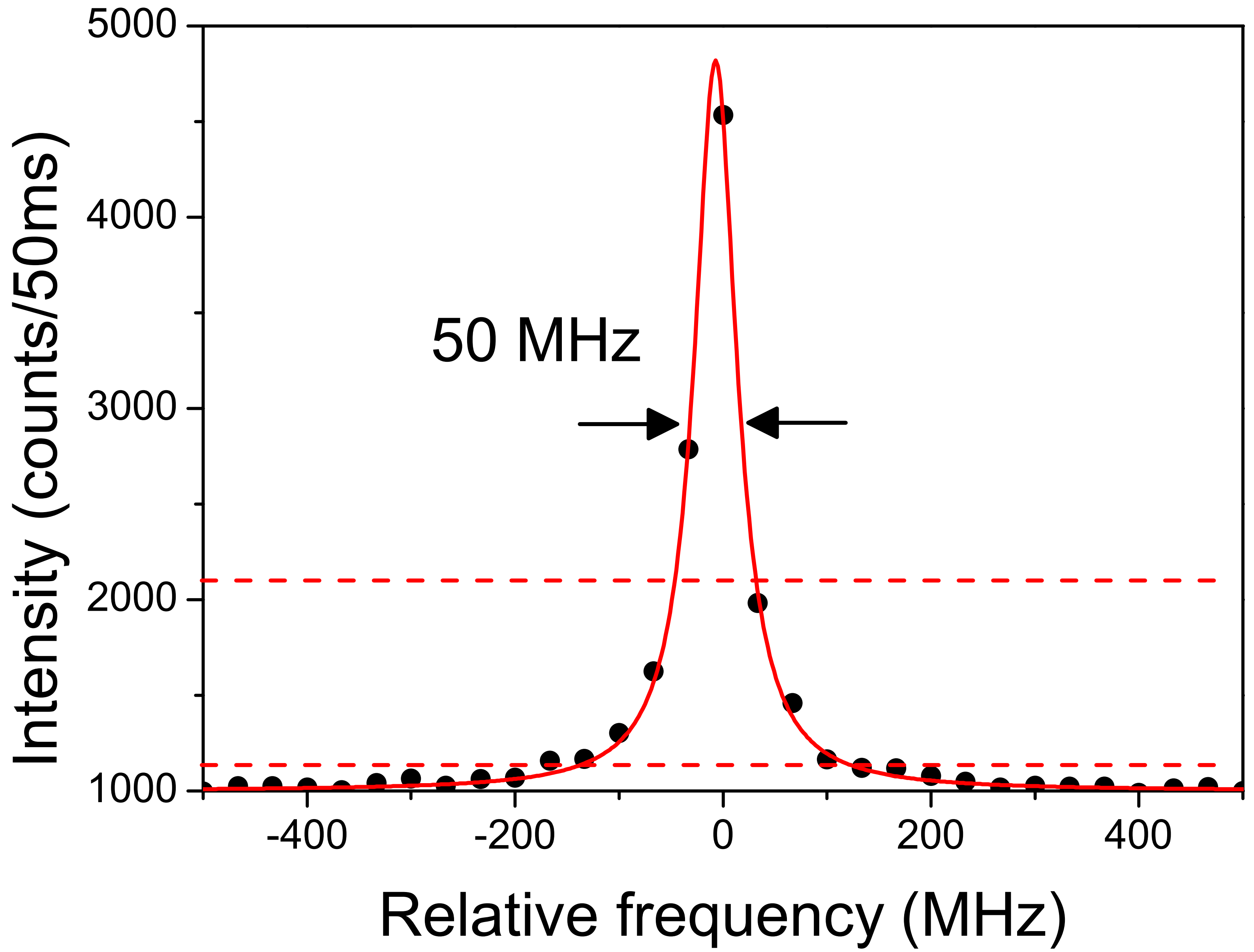
Fauré, M. et al. EPL 77, no. 1 (2007)



Puller, V. et al. PRL 110, no. 12 (2013)

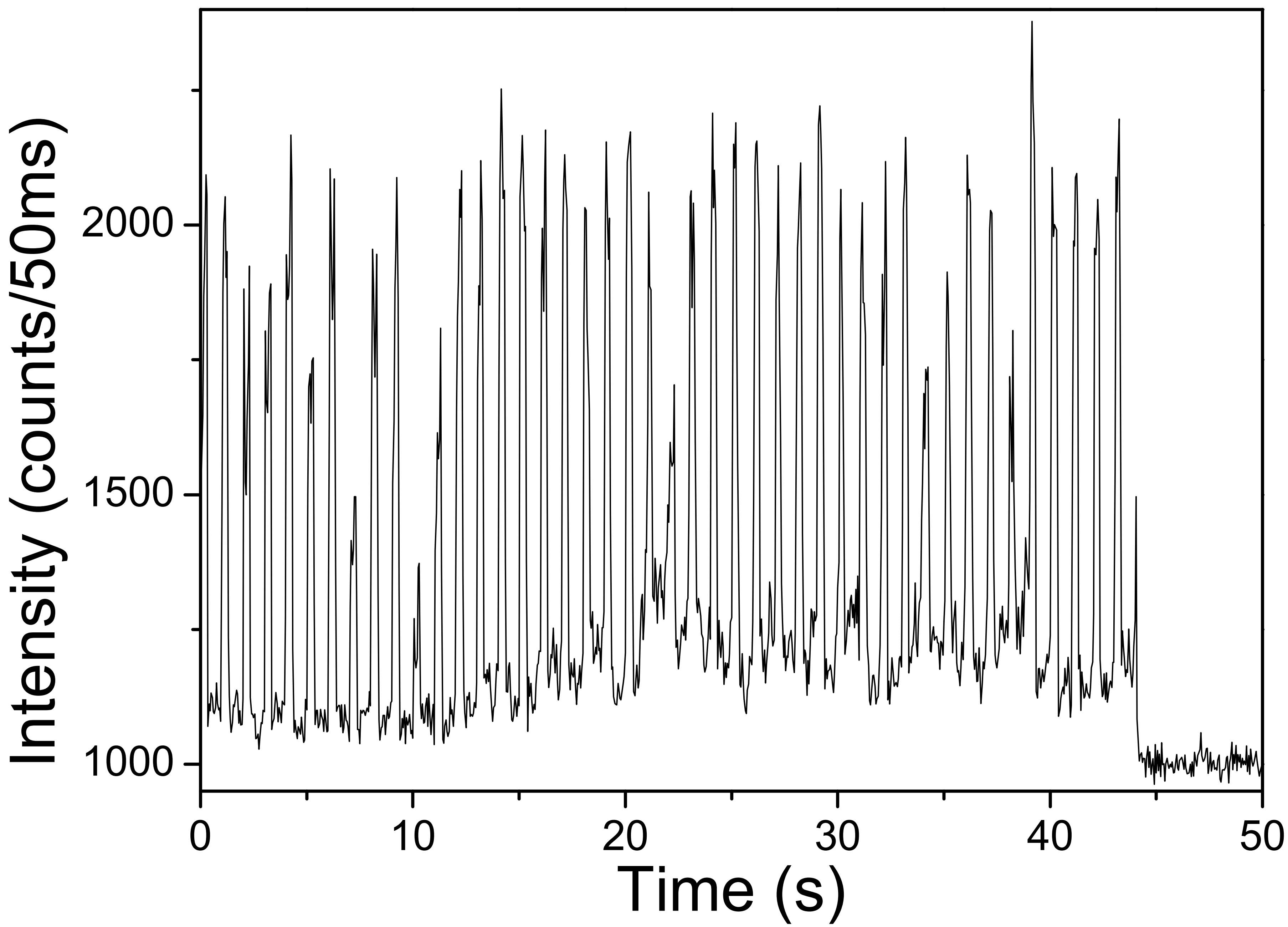
Supplementary materials

Fluorescence excitation spectrum at T = 2.6 K for the single DBATT molecule. The spectrum is recorded at an excitation intensity well below saturation. Lorentzian fit gives a FWHM linewidth of 50 MHz.



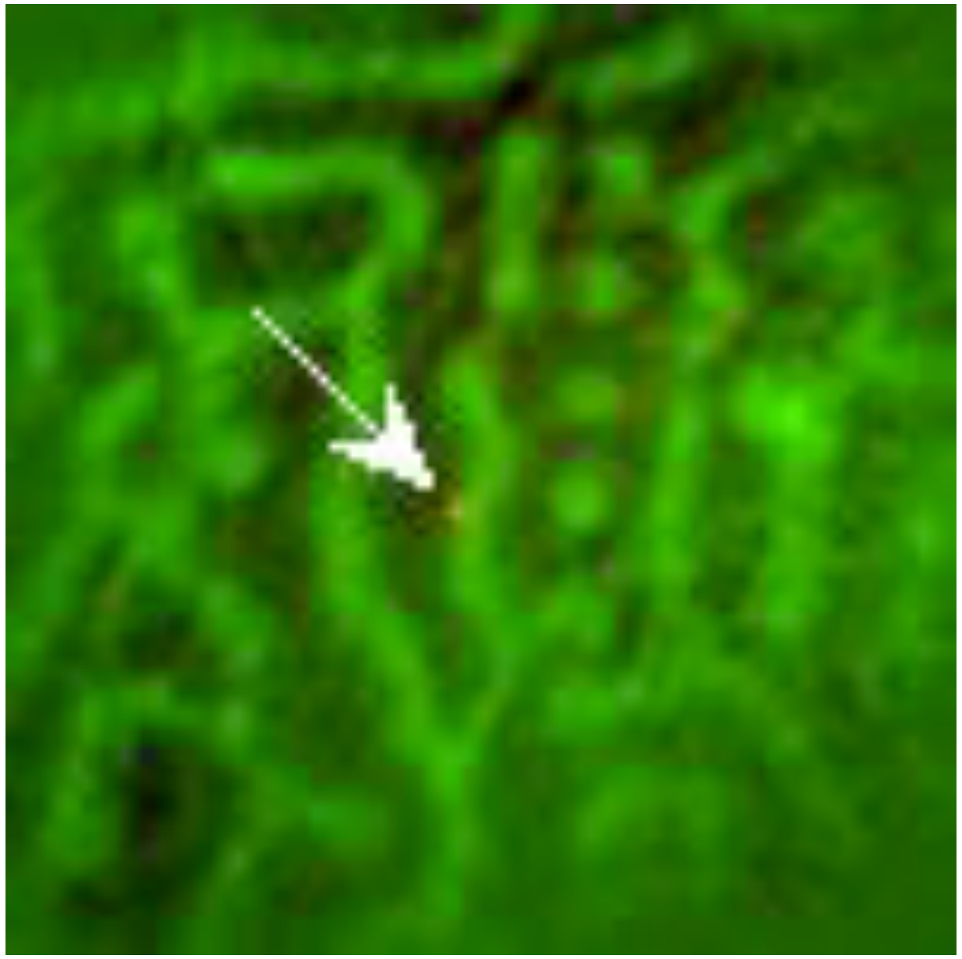
Supplementary materials

Full fluorescence time trace. The laser frequency is fixed on the slope of the Lorentzian-shaped molecular line. After time 44 sec, the laser was tuned far off resonance to determine the background level.



Supplementary materials

Movie of the simultaneous imaging of the periodical shift of superconducting domains in Indium and fluorescence of the molecule.



Supplementary materials

Experimental parameters :

- $w = 10\ \mu m$
- $d = 0.1\ \mu m$
- $d_S = 1.6 \mu m$
- $d_i = 2.5 \mu m$
- $M_S = 50 G$
- $\chi_e = 4/4\pi$
- $\chi_m = 0.11$
- $\gamma = 10^{-8}\ erg^{1/2}\ cm^{-1/2}\ G^{-2}$: Logginov, A. S. et al. JETP Letters 86, no. 2 (2007)
- $H_1(0) \sim H_c(2.6\ K) \sim 11.6\ mT$.