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# Fast and ultrafast all-optical control of light in nematic and smectic-A liquid crystals

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## ABSTRACT

We review recent experiments on the fast and ultrafast all-optical control of light in bulk nematic and smectic-A liquid crystals. Ultrafast optical control at sub-picosecond time scale can be achieved via the optical Kerr response of a nematic liquid crystal. We show that the refractive index changes are of the order of  $10^{-4}$  in 5CB nematic liquid crystal and can be optically induced by applying 100 fs pulses of  $4 \text{ mJ/cm}^2$  fluence. We discuss stimulated emission depletion of fluorescence in a smectic-A liquid crystal and demonstrate nanosecond light control of fluorescent pulse shaping. Both methods could be applied to control light by light in future photonic devices based on liquid crystals.

**Keywords:** liquid crystals, optical Kerr effect, STED effect, all-optical control

## 1. INTRODUCTION

Nematic liquid crystals are materials superior for application in large scale optical devices such as flat screens and modulators. Because of their fluidity, softness and strong orientational anchoring at confining surfaces, large scale optical devices with perfect optical quality can be produced. However, their soft elasticity and viscous nature also hinder their response to external electric and magnetic field. This response is collective in nature, meaning that molecules collectively respond to the electric or magnetic torque of external fields and thereby change the orientation of the optical axis. The resulting response times are of the order of milliseconds for reasonable magnitudes of external fields, whereas their turn-off time is determined by viscoelastic relaxation of an elastically distorted liquid crystal.

Whereas the technology of large sized liquid crystal devices is now perfected and is therefore in its mature stage, novel applications of liquid crystals are being considered, among others also the application as an electro-optic material in photonic micro-devices. Photonic micro-engineering is now at the cutting edge of technology research, where microscale devices are being developed akin to electronic microchips. The difference is that in photonic micro-devices, the information is carried by light and devices are being developed, where the optically encoded information will be controlled by light. These devices are therefore conceptually similar to semiconductor microchips, where the information is encoded in the electric charge and the flow of the electric charge is controlled by the electric force. Microelectronic devices based on electric charge transport have reached their optimum performance both in terms of their speed of signal processing and the energy dissipation per bit transaction. The latter is in fact the most hindering disadvantage of electronic devices and new concepts replacing electrons by photons are being intensely investigated. The goal is to process optical signals by light at 100 THz rate and reduce the power transaction per bit to less than femtoJoules per bit transaction.

When considering the application of liquid crystals in future micro-photonic devices, one therefore meets a tremendous gap between the kHz optical response of a liquid crystal and beyond the THz response of future photonic microcircuits. The question therefore arises of how to increase the speed of the nematic liquid crystal response for nine orders of magnitude. There are two answers to this question.

First, by increasing the driving electric field, the electro-optical response of a nematic liquid crystal becomes faster, but also changes its nature. Whereas in the low frequency regime, the response is collective and large, it becomes smaller but faster at higher frequencies, simply because the electric field excites other degrees of freedom in the material. It was shown [1] that by applying an electric field in a small capacitor filled with liquid crystal, one can obtain significant change of the refractive index on a hundred-nanoseconds timescale by using very short and intense electric pulses of several hundreds of  $\text{V}/\mu\text{m}$ . This is also the experimental limit of such a system, because the electric field cannot be

applied any faster. However, the electric field can be applied to the material also via the electromagnetic wave of light. In this case, the refractive index of the material is changed by the strong electric field of light, which is well-known in liquid crystals as an optical Kerr effect [2-5].

Second, for realization of all optical processing of optical signals, it is not necessary to modulate the refractive index which controls the flow of photons. Instead, one could directly modulate and control the production of light. An example of this control of the radiation of light by light is used in super-resolution optical microscopy, known as STimulated Emission Depletion (STED) microscopy [6]. Such an optical microscope is a fluorescent microscope, which effectively narrows the region of fluorescence emission by specially designed and time-synchronized STED pulses. The STED pulses quenches the light signal from a diffraction-limited excitation fluorescence spot by applying a doughnut-shaped STED impulse. The STED pulse takes away the fluorescence light from excited fluorescence molecules by stimulated emission. Because light pulses of sub-nanosecond duration are used in a STED microscope, this is an example of a nanosecond control of light by light. In principle, the STED principle could be used in micro-phonic devices such as micro-lasers and micro-cavities to control the light by light at tens of GHz frequencies.

In this article, we present some of our recent results, demonstrating ultrafast optical Kerr response of a nematic liquid crystal 5CB at a THz [7], and STED experiments in smectic-A liquid crystal 8CB, showing control and optical pulse-shaping at a GHz rate [8].

## 2. ULTRAFAST OPTICAL KERR RESPONSE OF A NEMATIC LIQUID CRYSTAL

It is well-known that in ordinary liquids and liquid crystals as well [2-5, 9-12], the electric field of a strong light impulse modifies the refractive index of the liquid via optical nonlinear Kerr effect on a time scale between 100 fs and 1 picosecond. In this nonlinear process, the electric field of light is directly affecting the molecular electron density distribution, while other molecular degrees of freedom remain practically unchanged. The nonlinear response is orders of magnitude faster than other optical phenomena in liquids. Because of the ultrafast response of the molecular electron density, the liquid can be considered as practically frozen in time, meaning that other degrees of freedom are not important at such short times.

Liquid crystals are orientationally ordered fluids with anisotropic optical properties and consequently polarisation-dependent optical phenomena. When considering the optical Kerr response on a sub-picosecond timescale, the liquid crystal presents a frozen-in-time ensemble of rod-like molecules, collectively aligned in a direction called the director. Because of this collective ordering, the optical Kerr effect should be polarisation-dependent and should be enhanced when compared to the isotropic liquid for a certain polarisation of the excitation (pump) beam. Many studies of the optical Kerr response of nematic liquid crystals have been performed in the past [2-5, 11, 12]. Some of them were concentrated on the optical Kerr response in the isotropic phase, where a Kerr response similar to ordinary liquids was observed. In the isotropic phase, resonant enhancement of the optical Kerr effect was observed on a 100 ps timescale because of the orientational interaction between elongated liquid crystal molecules. Available studies have shown that in the nematic phase, the persistent collective order give rise to complex and much slower orientational dynamics. In our recent work [7], we demonstrated for the first time an ultrafast response of the nematic liquid crystal 5CB on a timescale of 500 fs. The optical Kerr response is polarisation-dependent and is up to 5 orders of magnitude faster than ever observed before.

The experiments were performed on a 23  $\mu\text{m}$ -thick layer of a homogeneously aligned nematic liquid crystal 5CB, sandwiched between two polished quartz glass plates. The liquid crystal was aligned by a rubbed polyimide layer deposited on the quartz glass plates before the assembly of the cells. The optical Kerr effect on a planar nematic liquid crystal cell was measured in a standard pump-probe experiment at different temperatures, using an amplified Ti:Sapphire laser system with the repetition rate of 1 kHz, average output power of 1.2 W, emission wavelength of 800 nm and pulse duration of 100 fs. The laser beam was split with a 85% – 15% beam splitter to set up a non-collinear pump-probe scheme. The pump beam was intensity-modulated by a mechanical chopper operating at 500 Hz, whereas the probe beam was doubled in frequency with a non-linear Barium Boron Oxide crystal. The optical pulses of the two beams were delayed with respect to each other, and both beams were linearly polarised and then superimposed on the sample, as shown in the schematic Figure 1(a). The size of the pump beam in the focal plane on the nematic liquid crystal was set to

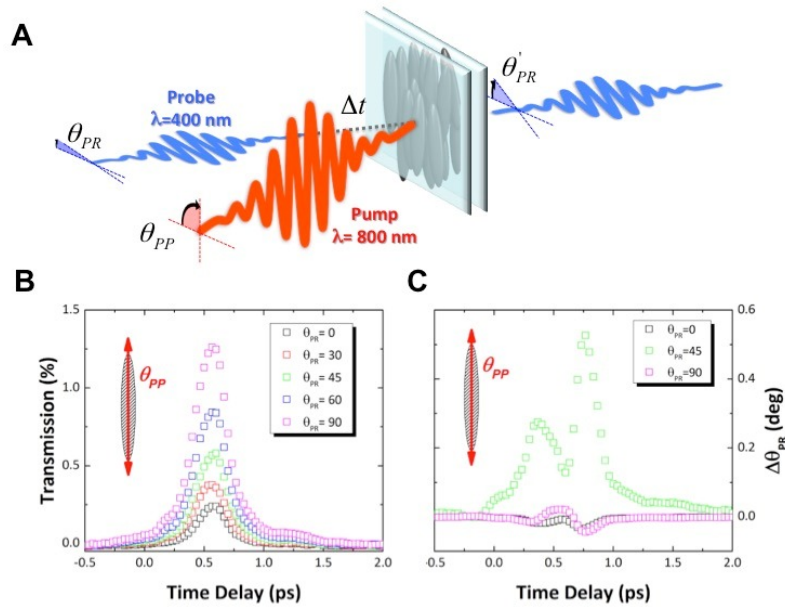


Figure 1. (a) The schematics of the pump-probe experiment measuring the ultrafast optical Kerr response of a nematic liquid crystal 5CB. Note that the polarisation of all beams is measured with respect to the plane perpendicular to the nematic director. (b) Time dependence of the normalised relative transmission of the sample with the pump polarisation along the director and different probe polarisations. The fluence of the pump beam is  $4 \text{ mJ/cm}^2$ , which corresponds to the pulse electric field strength of  $660 \text{ V}/\mu\text{m}$ . (c) Time dependence of the rotation of polarisation of the probe beam for pump polarisation along the director. Note that the polarisation rotation is largest at  $45^\circ$ .

approximately  $120 \mu\text{m}$  whereas the diameter of the focused probe beam was  $55 \mu\text{m}$ . The fluence of the excitation (pump) beam was measured and could be varied in the range  $2\text{--}10 \text{ mJ/cm}^2$  in order to study the optical Kerr effect as a function of the pump beam intensity. After the probe beam was transmitted through the sample, it was detected with a photodetector using a balanced detection scheme. In this scheme, the probe beam first passes through a Wollaston prism, which separates the probe beam with unknown polarisation into two linearly polarised beams with orthogonal polarisations. The intensity of these two polarisations are measured using two separate photodiodes. The sum of the two signals results in the time dependent transmitted probe intensity. On the other hand, the difference between the intensities of the two polarisations gives us direct information on the rotation of the polarisation axis of the probe beam. Both the transmission coefficient and the rotation of the polarisation are measured as a function of the relative delay time between the pump and probe beams, as well as their polarisation dependence.

Figures 1(b) and (c) show the response of a nematic liquid crystal 5CB to a  $100 \text{ fs}$  pump light pulse with the strength of the electric field of  $660 \text{ V}/\mu\text{m}$  oscillating along the nematic director. Well within the first ps of the light pulse, one can clearly observe an ultrafast optical transient, both in the normalised relative transmission of the sample and in the rotation of the probe beam polarisation. The change in the relative transmission is the strongest when the pump beam is also polarised along the director, therefore sensing the change in the extraordinary index of refraction of 5 CB. Obviously, there is an ultrafast change in the transmission of the sample for extraordinary polarisation, which is due to the time dependence of the extraordinary index of refraction. This change of the refractive index instantaneously modifies the transmission of the sample because a different amount of light is reflected at the interface between the quartz glass plate and the 5CB. On the other hand, there is little change when the probe polarisation is sensing the ordinary refractive index with the polarisation normal to the director.

Figure 1(c) shows the time dependence of the rotation of the probe beam polarisation, which is the strongest when the probe beam is polarised at  $45^\circ$  with respect to the director. This is consistent with the major change of the ordinary

refractive index and splitting of the incoming probe beam polarisation into ordinary and extraordinary beams travelling through 5CB. The phase retardation between the extraordinary index, which is strongly modified by the laser pulse, and the ordinary index, which is much less affected, is most influenced when the amplitudes of ordinary and extraordinary waves are equal, which happens at  $45^\circ$ .

To understand the polarisation properties and nature of the optical Kerr response of 5CB, we analysed the polarisation dependence of the OKE in the nematic and isotropic phases. Figure 2(a) shows the time dependence of the relative transmission of the sample in the nematic phase (Figure 2(a)) and in the isotropic phase (Figure 2(b)). Different panels show the relative transmission measured at different probe polarisations and at three different pump polarisations: pump polarisation perpendicular to the director in the first panel, at  $45^\circ$  in the second panel, and along the director in the third panel. In the nematic phase, one can clearly resolve a strong polarisation anisotropy of the OKE. When the pump beam is polarised perpendicularly to the director, the OKE is small and not dependent on the polarisation of the probe beam. The OKE is strongest and most anisotropic with the pump beam parallel to the long molecular axes and therefore the director. In this case, the OKE is strongest with the probe polarisation also parallel to the long molecular axes. This anisotropy disappears when the nematic liquid crystal is heated into the isotropic phase, as shown in Figure 2(b). In all cases of pump beam polarisations, the same OKE response is obtained for exactly the same probe beam polarisation. This can be understood in terms of molecular distribution, which is seen by the ultrafast optical pulse. In the isotropic phase, the liquid crystal is orientationally disordered and both the pump and the probe beams excite and sense a frozen-in-time random distribution of liquid crystal molecules. For a given polarisation of the pump beam, a certain number of molecules will contribute the most strongly to the OKE and this contribution will be measured when the probe beam will be polarised exactly in the same direction as the pump beam.

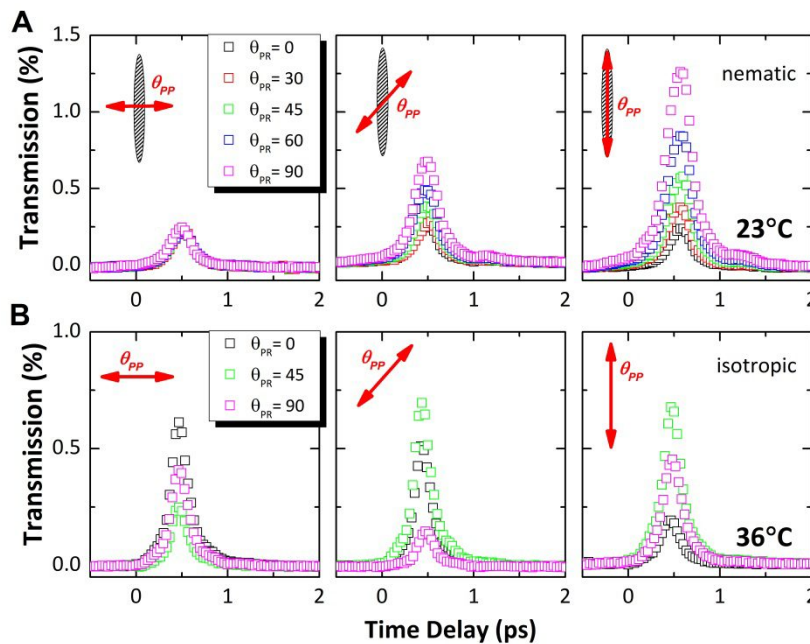


Figure 2. Pump polarisation dependence of the ultrafast response in the nematic liquid crystal 5CB. (a) In the nematic phase of 5CB, at  $23^\circ\text{C}$ , the normalised relative transmission is polarisation-dependent, and the maximum is obtained when both pump and probe polarisations are along the long molecular axes of the NLC. (b) In the isotropic phase, at  $36^\circ\text{C}$ , the response does not depend on the direction of the pump polarisation, and we get maximum response whenever the probe polarisation is rotated into the pump polarisation direction.

The situation is quite different in the nematic phase, when the pump beam will be most efficient with the polarisation parallel to the director and the probe beam will measure the maximum OKE for exactly the same polarisation. By comparing the maximum OKE response in the isotropic and the nematic phase, we see that the OKE is approximately two times bigger in the nematic phase compared to the isotropic phase. The ratio of the OKE in the nematic phase is approximately 4:1, because of the incomplete ordering of the long molecular axes in the nematic phase. In a material with a larger order parameter, this ratio is expected to be higher.

The amplitude of the OKE can be determined either from the measured change of the relative transmission coefficient or by analysing the magnitude of the polarisation rotation as a function of time. For our experiments, we obtain the maximum change of the extraordinary refractive index of 5CB, which is time-dependent, and of the order of  $2 \cdot 10^{-4}$ , as shown in Figure 3.

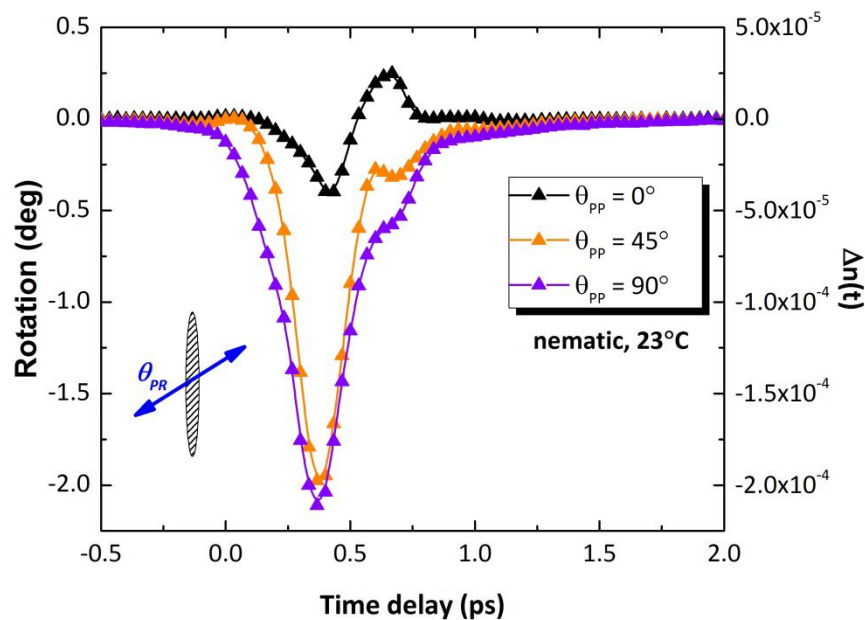


Figure 3. Rotation signal (left axis) and calculated birefringence  $\Delta n(t)$  (right axis) as a function of the pump polarisation axis and time, keeping the probe polarisation fixed at  $45^\circ$ . The measurement has been performed at  $23^\circ\text{C}$  with a pump fluence of  $4 \text{ mJ/cm}^2$ .

The signal of the polarisation rotation, shown in Figure 3, is a mixture of quasi-oscillation and a Gaussian-like offset, which depends on the pump beam polarisation. This behaviour is indeed expected for a Kerr-active sample and is characteristic of the nematic phase, since the oscillation part disappears in the isotropic phase. There, only a simple Gaussian profile remains, with a slightly longer lifetime as compared to the nematic phase.

Based on the observed frequency, magnitude and time duration, the ultrafast Kerr response in the nematic phase of 5CB can be understood in terms of optically induced librational modes, which are intramolecular vibrations excited through a non-resonant channel i.e. by stimulated Raman scattering. The frequency spectrum of the pump optical pulse has a bandwidth of 3 THz and contains both frequencies required to excite the low-frequency vibrational modes of 5CB, collectively oscillating along the pump polarisation. When the pump polarisation is along the long molecular axis, these single-molecule vibrations will be coherently excited and result in an enhanced optical Kerr response of 5CB, oscillating at 2.7 THz.

### 3. SUB-NANOSECOND LIGHT CONTROL BY STED

Depletion of fluorescence of dye molecules by stimulated emission depletion was first studied in 1994 by two different groups in two different contexts. While S. W. Hell and J. Wichmann [6] considered this phenomenon as a way to bypass the diffraction limit of optical microscopy, J. R. Lakowitz et al. [13-15] considered the same phenomenon as a way to modulate fluorescently emitted light on a very fast timescale for fluorometry. Whereas the experimental geometries and targets are quite different in both cases, the underlying physics is the same.

Consider a medium, consisting of fluorescence emitters, which is irradiated with a short (subnanosecond) pulse of light, exciting the electrons in the fluorophore molecules into their excited electronic state. If let free, the electrons will spontaneously transit into their ground state and the molecules will emit fluorescent light within their characteristic fluorescence decay time, which can be as short as 1 nanosecond (Figure 4 (a)). In STED mechanism, the excited molecules are not allowed to spontaneously decay. Instead, before significant fluorescence takes place, another light pulse is sent to the sample, with the frequency resonant with the fluorescent dye electronic transition, as illustrated in Figure 4 (b). These resonant (STED) photons will cause the creation of identical photons via the effect of stimulated emission of light. These new, resonantly created photons will travel along the same direction as the STED beam and the energy. This means that for all other directions of observation, excluding the direction of the STED beam, there will be strongly depressed fluorescence and the sample will be effectively dark. This effect, where the energy of the excited fluorophore is resonantly taken along the STED beam, is also called the depletion of fluorescence or even the quenching of fluorescence. Because the STED effect is related to the electronic transitions in dye molecules, it is very fast, which in turn means that one is in principle able to control light (emitted fluorescence) by light (i.e. STED photons) on a nanosecond or even sub-nanosecond timescale. By using the STED effect, it is therefore possible, in principle, to modulate light beyond the GHz frequencies.

The first time-resolved STED experiments in smectic-A liquid crystals were reported by us in 2015 [8]. The smectic-A liquid crystal 8CB was homogeneously aligned into a thin layer between two glass plates and was previously doped with the Nile red fluorescent dye. This dye is known to align very well in 8CB with the molecular radiative dipole moment parallel to the nematic director.

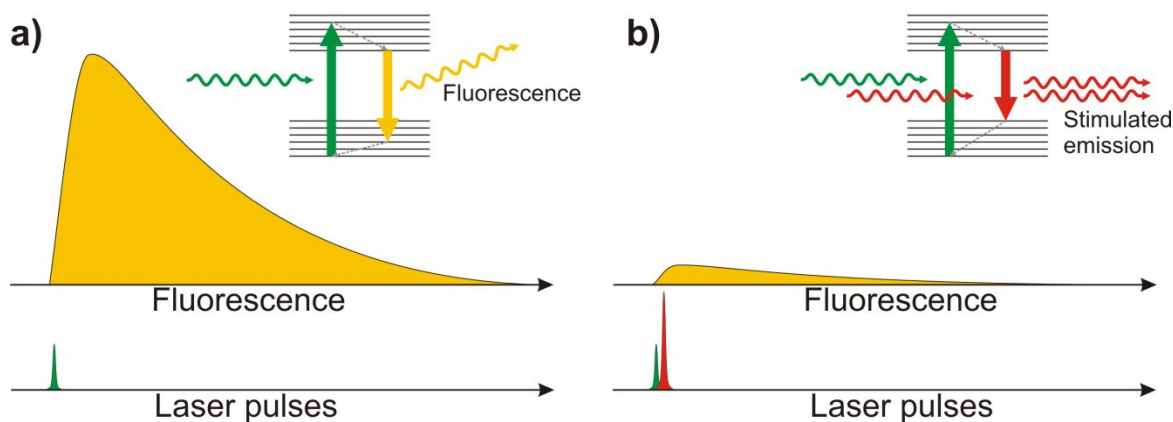


Figure 4. Illustration of the STED principle. (a) A fluorophore gets into an excited state when it absorbs the excitation photon. It then relaxes by spontaneous emission of a fluorescent photon with a larger wavelength into a random direction. Fluorescence lifetime is in the range of nanoseconds. (b) If the STED pulse with a frequency resonant to the dye's electronic transition is applied before the fluorescence has taken place, it causes the system to relax via stimulated emission. In this case the emitted photon is equal to the STED pulse photon, both in wavelength and direction. Spontaneous emission hardly takes place. Because almost all of the light is emitted in a single direction the sample appears dark when viewed from any other direction.



The absorption and emission of dye molecules dispersed in 8CB will therefore be anisotropic, which means that the STED effect will be strongly polarisation-dependent. The STED effect was studied in an optical setup where a pulsed supercontinuum laser emitting white laser light with a continuous spectrum ranging from blue to infrared was used. Two beams, each of 20 nm wavelength band, were extracted from the white spectrum and polarised. They were used as the excitation and the STED beam. In the setup, it was possible to adjust the positions of each of the bands of the two beams, as well as to precisely set the time delay between the two beams with ~3 picosecond resolution.

Both beams were polarised, sent to the microscope via polarisation preserving optical fibres and then precisely overlapped on the focal plane of the microscope objective. The intensity profiles of both beams were Gaussian and their spatial overlapping on a focal plane was adjusted by observing the fluorescence of the Nile red molecules, dissolved in 8CB. In addition to spatial overlapping, precise tuning of the STED wavelength and time delay of the STED pulses with respect to the excitation pulses had to be performed. As a result, one can observe very strong depletion of the fluorescence of the Nile red molecules under an optical microscope. Figure 5(a) shows the microscope image of the fluorescence, as observed under the microscope. When only the excitation beam was present, with the STED beam mechanically blocked, one can see an orange spot on black background, as shown in Figure 5(a).

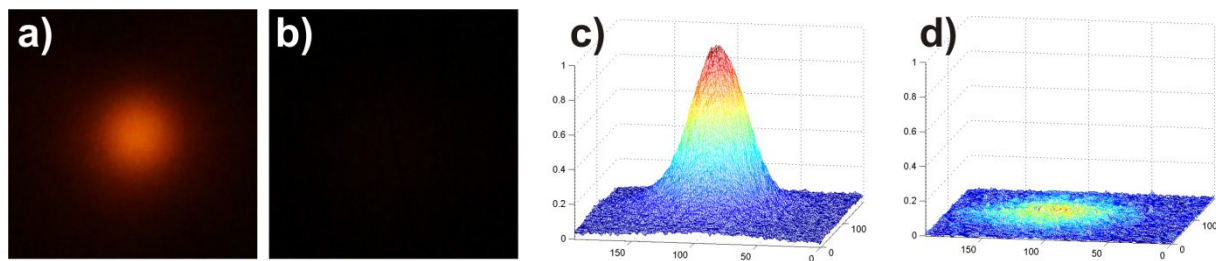


Figure 5. Microscope images and fluorescence profiles of the illuminated area of the 8CB/Nile red mixture. Both polarisations of the excitation and STED beam are parallel to the director. (a) Only the excitation beam (532 nm) is illuminating the sample, with polarisation along the director (and the radiative dipole of Nile red), which results in the observed fluorescence emission. (b) The STED beam (705 nm), polarised along the director, is switched on, which causes a strong decrease of fluorescence emission. (c) Intensity profile of the image in (a), showing fluorescence. (d) Intensity profile of the depleted fluorescence, shown in (b).

When the STED beam is turned on, and both the excitation and the STED beam are active on the molecules, one clearly sees a strong darkening of the previously orange fluorescent spot (Figure 5(b)). The reason for this is simple: the STED beam takes the resonantly produced stimulated photons in a direction away from the eyepiece of the microscope. Practically all the light is therefore taken along the STED beam and there is no fluorescence emission towards the eyepiece and the detectors. Figures 5(c) and (d) show the measured intensities of the fluorescent spot in the case of spontaneous fluorescence (Figure 5(c)) and depleting of the fluorescence (Figure 5(d)). The STED beam lowers the measured fluorescent intensity for more than one order of magnitude (ratio 1:12 was achieved).

Whereas the microscope images in Figure 5(a,b) show the light intensity collected during a very large number of optical pulses from a 1 MHz white laser, it is interesting to see on a real-time scale, what actually happens to individual fluorescence pulses when the STED pulses are delayed in a controlled manner. This is shown in Figure 6, where the time dependence of the emitted light from Nile red molecules is recorded for different delays of the STED pulse, using a 4 GHz bandwidth photodiode and oscilloscope. Figure 6(a) shows the detected fluorescence signal when the STED pulse was delayed for 6 ns with respect to the excitation pulse, arriving to the sample at time  $t = 0$ . The duration of these pulses is 150 ps, which is much shorter than the timescale of our observation. In Figure 6(a) one can observe a sudden increase in fluorescence intensity, which is due to the excitation of the dye molecules with the excitation pulse. This intensity then decays exponentially with the fluorescence decay time of Nile red molecules of the order of 3 ns. Then, 6 ns after excitation, the resonant STED impulse arrives to the sample and takes away all the remaining energy by creating resonant STED photons via stimulated emission. Because of that, the fluorescence is suddenly shut-down, practically to a completely dark level. By decreasing the time delay of the STED pulse, the remaining fluorescent impulse gets shorter and shorter, until it is as short as 1 ns, as shown in Figure 6(d). By further shortening the delay time, there is practically



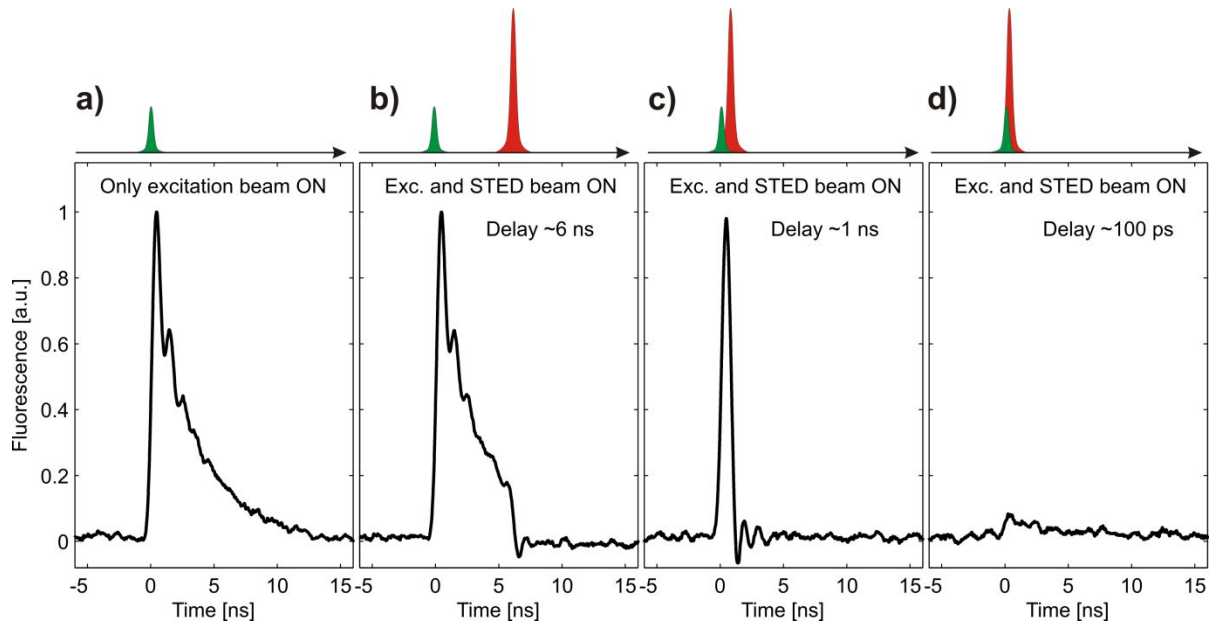


Figure 6. Controlled shortening of the emitted fluorescence by applying a time-delayed STED pulse. In (a), no STED beam has been applied to the sample so the fluorescence intensity decays exponentially. In (b), the STED pulse is delayed for  $\sim 6$  ns with respect to the excitation pulse. Note a sudden decrease of the fluorescence to zero after 6 ns when a 150 ps STED pulse was applied. The delay time of the STED pulse was gradually shortened in (c, d), which resulted in shortening of the emitted fluorescent light (c) or even in its disappearance (d). The delay time was roughly estimated from the optical path measurement with an uncertainty of 50 ps.

no fluorescence detected within the 4 GHz bandwidth of our detection system. However, there is still a possibility that fluorescent pulses could be made even shorter at precisely controlled delay times below 1 ns.

#### 4. Conclusion

In summary, we have demonstrated two different mechanisms to control the flow of light by light in liquid crystals by using very different underlying physics. The results clearly show that it is possible to modulate light in liquid crystals at gigahertz to terahertz rates by using the optical Kerr effect or STED control. There are advantages and disadvantages to each of these two methods. The optical Kerr effect requires extremely strong electric fields of light, which in turn requires large energy density. Whereas this is difficult to achieve in bulk liquid crystal samples, extremely strong local optical fields could be generated in small droplets or fibres made of liquid crystals. On the other hand, the STED technique is well known for extremely large intensities of the STED control beam, which easily exceed the fluorescence excitation intensities by five orders of magnitude. However, this ratio could be made smaller by using resonant enhancement in optical resonators, which remains to be tested in the future.

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