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Ultrafast optical processing with photon echoes

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Abstract

We use frequency-selective materials to expand the conventional spatial-domain optical processing into the dimensions of frequency and time. Ultrafast performance on the time scale of 10^{-13} s is achieved by using resonant media with broad inhomogeneous bandwidth of up to 6 THz, in combination with dipole-allowed resonant transitions such as in organic dye-doped polymers at liquid-helium temperature. We discuss experiments on ultrafast bit-to-bit multiplication by photon echo with spectrally shaped ultrashort pulses. We present an implementation of a coherent logic gate performing "controlled, controlled NOT" operation by time-domain interference of ultrafast photon echo. © 1999 Elsevier Science B.V. All rights reserved.

Keywords: Ultrafast holography; Photon echo; Coherent optical transients; Optical processing

1. Introduction

Inhomogeneously broadened frequency-selective materials have a special property of responding to optical wave fronts, in addition to conventional spatial degrees of freedom, also in the dimensions of frequency and time. Recently there has been a considerable interest to apply this special feature to optical holography, leading to novel memory- and processing device. Because of the additional dimensions involved in the light-matter interaction, such devices are potentially able to process data at a much higher speed and store information with much higher density than available with current technologies.

Molecules and atoms embedded in strongly disordered materials such as polymers and glasses are characterized by broad inhomogeneous absorption bands, with a width often exceeding several terahertz. In combination with a narrow width of homogeneous zero-phonon line at low temperatures, such materials can be used for frequency and time-domain storage and processing of broadband ultrafast optical signals with a temporal resolution better than 10^{-13} s.

Holography provides one possible approach how to utilize different degrees of freedom available in the frequency-selective media. Time-and-space-domain holography [1–4] combines the three spatial dimensions with the frequency dimension and correlated to it via Fourier transformation temporal dimension. It allows to implement

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a multidimensional generalization of conventional holography, which serves as a basis for ultrafast convolution and correlation of time–space varying optical wave amplitudes [5–8] (see also Ref. [9] and references therein). Previously, ultrafast analog processing has been performed using dye-doped polymers at low temperatures. Some of the previous demonstrations include associative recall of information [10–12], interferometry of transient images [13,14] and frequency-domain synthesis of arbitrary pulse shapes and delays [15,16].

An alternative approach, described in this paper, is to consider bit-oriented functions such as optical gates and switches. Novel principles for ultrafast logic and switches are of great interest in the field of optical computing [17,18], and most recently in quantum computing [19]. A variety of non-linear optical phenomena, including soliton propagation in fibers [20], frequency-conversion [21], etc. have been proposed in the past to implement ultrafast operations. Coherent optical transients such as photon echo [22] provide means for constructing not only analog devices but also schemes oriented towards processing digital data. This can be achieved if the non-linear character of the photon echo is combined with a proper threshold procedure. Previously, holograms recorded by persistent spectral hole burning were used to implement a XNOR gate [23]. Interference between coherent transients has been used to selectively erase bits in time-domain sequences of data pulses [24–27].

In this paper we explore possibilities of bit-oriented optical processing by using the dimensions of frequency and time. We discuss issues concerning materials properties to achieve ultrafast performance. We present experiments on ultrafast bit-to-bit multiplication by photon echo with spectrally shaped pulses. We also implement a three-port coherent logic gate, which performs as an ultrafast "controlled controlled NOT" by time-domain interference of photon echo.

2. Materials for ultrafast time- and space-domain processing

For efficient multidimensional processing the resonant medium needs to have several specific properties. The first and perhaps the most obvious requirement is that the resonant transition needs to have a sufficiently broad inhomogeneous spectral width. In particular, a pulse with a duration of 100 fs has a typical spectral width of about 150 cm^{-1} (4.5 THz). This determines the smallest acceptable value of the inhomogeneous width. Fig. 1 shows inhomogeneously broadened absorption spectrum of some selected tetrapyrrolic dye molecules embedded in polymer film at low temperature. The inhomogeneous bandwidth of the featured transition from the ground singlet electronic state to the lowest excited singlet state is about 200–250 cm^{-1} (6–7.5 THz). The large inhomogeneous width is a consequence of strongly disordered microscopic structure of the polymer host. Similar inhomogeneous widths are achieved by incorporating chromophores in glasses or specially designed disordered crystals. Another advantage of these materials is that the wavelength of the working absorption band at 770–780 nm overlaps with the tuning range of commercial mode-locked Ti : sapphire lasers. It would be certainly even more preferred, if the electronic transition would be in the fiber communication wavelength range of 1500–1600 nm. Unfortunately, no organic molecules investigated so far has suitable absorption- and emission-properties in the infrared range. This drawback may be due to the fact that in large molecules the excited S_1 states has a tendency to overlap with the vibrational levels of the group state, which can lead to a fast non-radiative decay.

Recently, photon echo in Erbium-doped inorganic crystal was used for storage and processing around 1500 nm wavelength [28]. In general, the inhomogeneous bandwidths of rare earth ions in inorganic crystals are as narrow as 1–10 GHz. At the same time, the homogeneous line width at low temperature can be very narrow, reaching especially in low magnetic moment hosts into sub-kilohertz region. In case of $LiNbO_3$ an inhomogeneous band width of 200 GHz has been observed [29]. However, because of a dipole-forbidden nature of the electronic transition, even for crystals with high dopant concentration, an increase of the inhomogeneous bandwidth leads to a corresponding decrease of the peak absorption coefficient. A practical value of absorption should

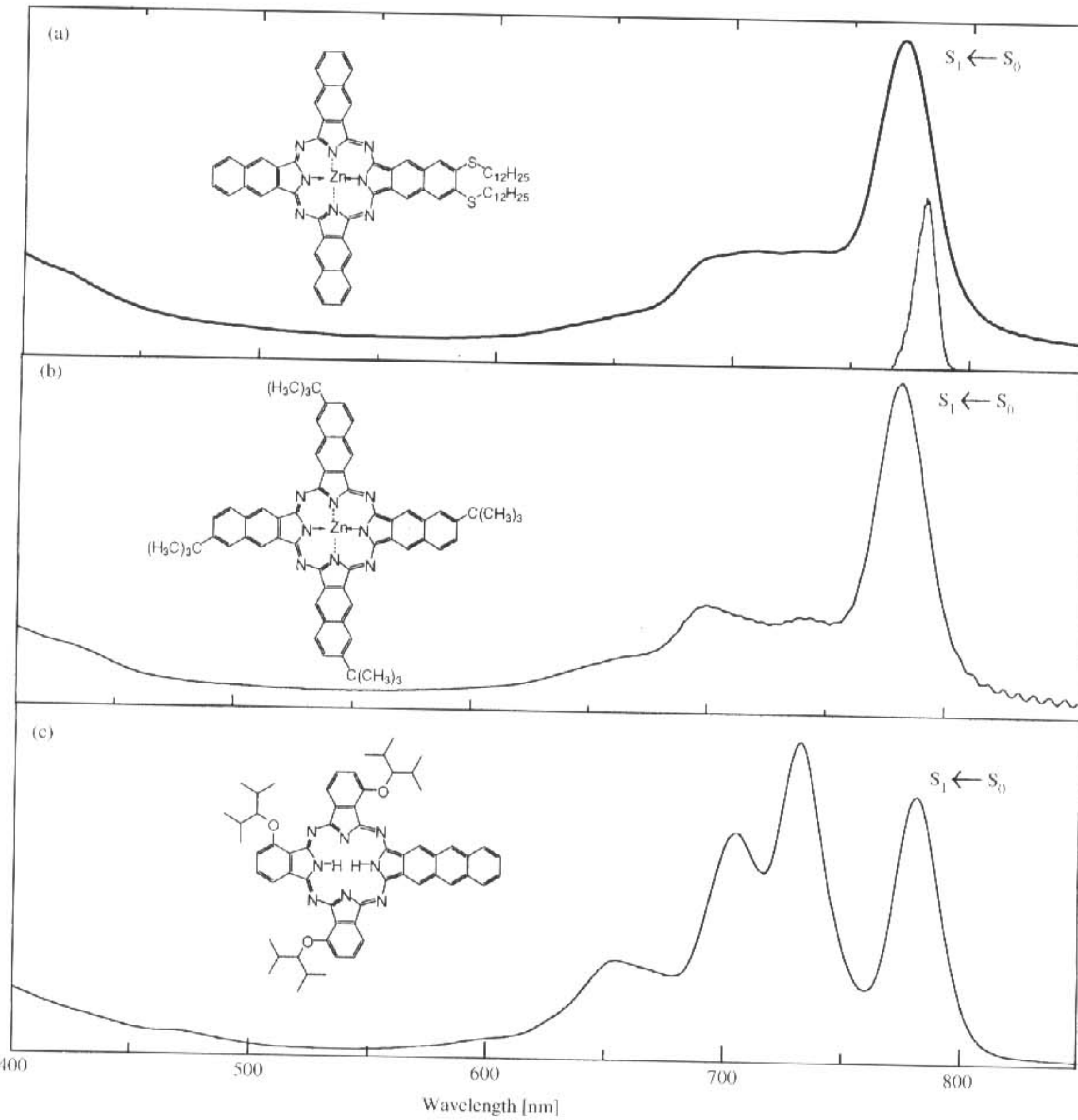

Fig. 1. Low-temperature (10 K) absorption spectra and chemical structure formulas of some phthalocyanine-type dye molecules used in our experiments. (a) Ciba dye HW 3463; (b) Aldrich dye 43,221-0; (c) Ciba dye HW 1009. All three molecules exhibit a dipole-allowed $S_1 \leftarrow S_0$ transition at wavelength around 770–780 nm. A typical intensity spectrum of amplified femtosecond Ti : sapphire laser is shown for comparison.

be above 1 cm^{-1}, which is hard to achieve for weak transitions distributed over a broad frequency range.

This leads us to the second requirement for ultrafast applications, which consists in sufficiently high oscillator strength of the resonant transition. To obtain a photon echo response on an ultrafast time scale, the interaction between the excitation pulses and the resonant medium needs to be sufficiently strong, such that the transition from the ground state to the excited state is accomplished within a very short period of time. This implies that the oscillator strength of the resonant transition should be on the order of $10^{-2} - 1$. These high values are quite common to organic molecules, while for f–f transitions in rare earth ions small values on the order $10^{-8} - 10^{-6}$ are typical. For transitions with small oscillator strength a limiting factor may be the peak intensity of ultrashort excitation pulses, which cannot much exceed the threshold of optical breakdown of the medium.

The third requirement relevant mostly to the molecular systems consists in small coupling between the electronic transition and the vibrational modes of the host matrix. In the materials shown in Fig. 1, a moderate electron–phonon coupling leads to the fact that even at low temperature as much as 50–70% of the pulse energy is absorbed via phonon side band rather than through the zero phonon line. As a consequence, broad-band excitation in the $S_1 \leftarrow S_0$ transition region converts to a large extent directly into heat, rather than into optical coherence. This drawback can limit critically the efficiency of storage and processing, especially if the excitation pulses are applied with a high average power and at a high repetition rate. To overcome this problem, molecular systems with a small electron–phonon coupling must be found, possibly by invoking specialized organic synthesis.

3. Bit-to-bit multiplication in frequency domain

Photon echo can be described as a time-delayed scattering from transient gratings self-induced by a series of excitation pulses in an inhomogeneously broadened resonant media [30]. The gratings are created because of nonlinear interaction of light with the resonant media, usually considered in the fames of a model two-level atom. The nonlinearity of a two-level system is due to the fact that the effect of each excitation pulse depends on the amplitude and phase of the pulses applied earlier in time. The information about the previous pulses is preserved in the phase memory or optical coherence of the molecules. In particular, two-pulse photon echo is created if two pulses with amplitudes, $E_1(t)$, and, $E_2(t-t_p)$, are applied within a time interval less than the optical dephasing time $t_p < T_2$. If the spectral width of the pulses does not exceed the inhomogeneous bandwidth, then the spectral amplitude of the echo can be expressed as

$$E_{echo}(\omega) \propto E_2^*(\omega)E_1^2(\omega), \qquad (1)$$

where $E_1(\omega)$ and $E_2(\omega)$ are the Fourier amplitudes of the first and the second pulse, respectively. Let us assume that some frequency components are selectively removed, e.g. by spectral filtering, from the first pulse. Then, according to relation (1), even if these spectral components are present in the second pulse, the intensity spectrum of the photon echo will not contain these missing components. In a similar way, if components are removed from the spectrum of the second pulse, then the echo will not contain these frequencies. Based on such simple principle, by assigning bit values to different frequencies, we can implement a direct bit-to-bit multiplication operation. To achieve this we need to interpret the presence of a certain frequency in the echo as 1 and the absence of a frequency as 0. An interesting property of such multiplication is that the phase of the pulses is not relevant, because the measured intensity spectrum does not depend on the phase of the excitation pulses. A further interesting point is that such multiplication occurs in parallel at all frequencies. The maximum number of the parallel frequencies is given by the ratio of the inhomogeneous bandwidth to the homogeneous line width. In practice, of course, this ratio will be determined by the spectral resolution with which data is encoded into the excitation pulses.

Recently we have demonstrated a multiplication of two 8-bit binary data vectors in frequency dimension on a time scale less than 10 ps [31]. The multiplication operation was performed by exciting two pulse photon echo in a dye-doped polymer film, similar to that shown in Fig. 1(c). Fig. 2 presents the principle of the experiment. We used 100-fs-duration pulses, which were passed through a two-channel pulse-shaper (for experimental details see Ref. [31]). The pulses illuminated the sample, which was contained in a low-temperature

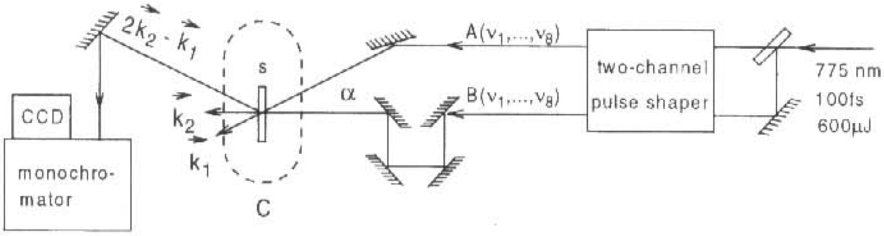

Fig. 2. Experimental arrangement for the frequency-domain multiplication. The output of amplified Ti : sapphire laser is split into two and passed through a two-channel spectral shaping device. Spectrally tailored pulses are directed at the sample inside a liquid-helium cryostat (C) at an angle α. The echo signal is emitted at a complementary angle α and is detected with a spectrometer.

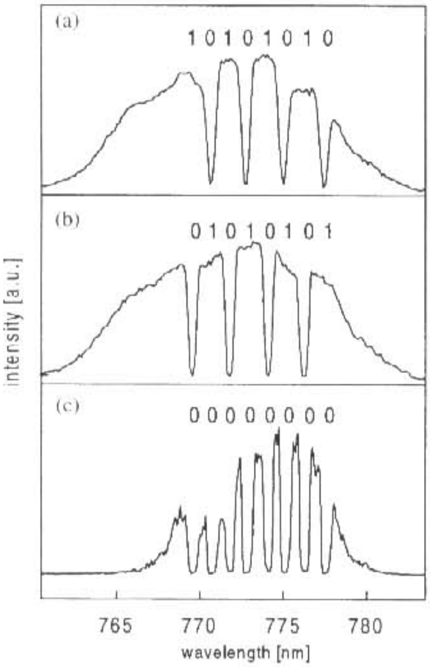

Fig. 3. Intensity spectrum of the first (a) and the second (b) excitation pulses and the resulting two-pulse photon echo spectrum (c). Maxima in the excitation pulse spectra correspond to the frequency components passed through the spectral shaping device. The energy of the excitation pulses was $I_A = 0.5$ and $I_B = 1$ mJ cm^{-2}.

cryostat. A few picosecond time delay was introduced between the pulses in order to avoid temporal overlap. The result of the multiplication was obtained by measuring the intensity spectrum of the echo with a spectrometer. Fig. 3 shows the result, where the intensity spectrum of two pulse echo (c) is seen to the proportional to a product of the intensity spectra of the first (a) and the second (b) excitation pulses. The excitation pulses were coded to present two binary vectors $A = \{1, 0, 1, 0, 1, 0, 1, 0\}$ and $B = \{0, 1, 0, 1, 0, 1, 0, 1\}$. Accordingly, the intensity spectrum of the echo gives a bit-to-bit multiplication, $A \times B = \{0, 0, 0, 0, 0, 0, 0, 0\}$. The duration of the whole multiplication procedure was on the order of a few picoseconds. This experiment shows the advantage of parallel access in the frequency dimension. By these means it is possible, in principle, to perform terahertz-rate processing in one spatial spot. A further step would be to include a carry-over function, such that the result obtained at one frequency can be transferred directly as an input to an adjacent frequency. This would allow addition and multiplication of binary numbers.

4. Logic gate by interference with photon echo

In this section we present a experiment, where we demonstrate an all-optical logic gate by using interference of femtosecond photon echo. The particular type of interference effect discussed here is a consequence of causal nature of photon echo, which prevents pulses applied later in time to change the interference of the pulses applied earlier. Let us consider once again a simple situation, where two-pulse echo is produced by applying two excitation pulses, separated in time by an interval, $\Delta t \ll t_p \ll T_2$, where Δt is the duration of the pulses. The macroscopic polarization, responsible

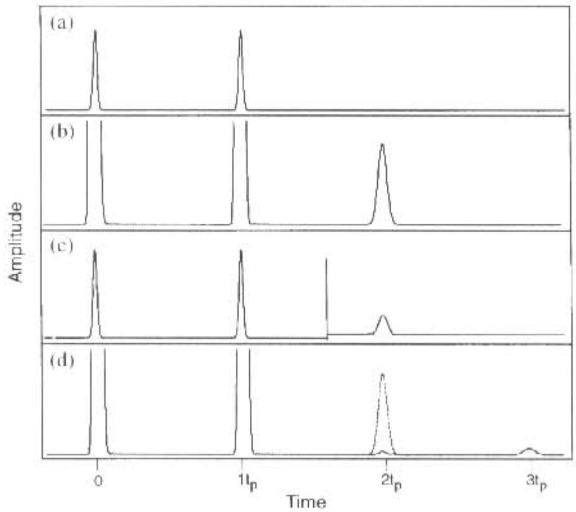

Fig. 4. Simulation of a two-pulse photon echo interference with a third excitation pulse. (a), (c) Excitation field amplitude; (b), (d) macroscopic polarization in an optically thin medium. The first two excitation pulses are applied at time $t = 0$ and $t = t_p$ (a). The third pulse with a lower amplitude is added at $t = 2t_p$ (c). The first two pulses induce a macroscopic polarization at time $t = 2t_p$ (b). Addition of the third pulse reduces the polarization at $t = 2t_p$ at least by one order of magnitude (d). Dashed curve corresponds to the echo polarization in the absence of the third pulse. A weak polarization pulse in (d) at time $t = 3t_p$ is echo due to combined action of the second and third excitation pulses.

Input			Output		
pulse 1	pulse 2	pulse 3	pulse 1	pulse 2	pulse 3
0	0	0	0	0	0
0	0	1	0	0	1
0	1	0	0	1	0
0	1	1	0	1	1
1	0	0	1	0	0
1	0	1	1	0	1
1	1	0	1	1	1
1	1	1	1	1	0

Fig. 5. Truth table of the three-port logic gate. The table is constructed by considering six possible input pulse combinations. The presence or absence of a pulse at the input/output is indicated as 1 or 0. Shaded cell corresponds to the case where output pulse is absent because of the causality condition (see text for discussion).

for the echo signal, will be created at about time $t = 2t_p$ after the first pulse. Fig. 4(a and b) shows a simulation of the two pulse echo polarization under condition, where both pulses have small areas, $\Theta_{1,2} < \pi/10$. Let us assume that a third pulse is applied at the time when the echo is generated. We can choose the amplitude and the phase of third pulse such that the polarization due to this pulse almost exactly compensates the echo polarization. As a result, the photon echo is "erased" by destructive interference with the third pulse. Fig. 4(c and d) show the corresponding model calculation, where the dashed line is the original two-pulse echo polarization. In terms of diffraction, we can say that the third pulse interferes destructively with light amplitude diffracted from a grating created by the first two pulses. It is important to note that the time delay between the pulses guarantees that the grating created by the second and third pulse does not interfere with the first pulse. This is contrast to conventional self-diffraction with temporally overlapping pulses. In the case of a media without a phase memory, no distinction can be made between the pulses based on their temporal arrival time. Consequently, the interference will affect all breams equally. The causal nature of the photon echo allows us to introduce a "arrow of time" as well as it allows to preserve the precise information about the amplitude and phase of the excitation pulses for a time interval, which is much longer than the actual duration of the pulses.

Fig. 5 shows a truth table, which we have constructed by considering the intensity of the three pulses at the output of the echo medium, depending on the intensity of the three input excitation pulses. The presence of a pulse is interpreted as 1 and the absence is interpreted as 0. The first six rows correspond to a situation, where at least one of the first two input pulses is blocked. In this case the output bits repeat the input bits. Note that if the time-delayed photon echo response would be substituted through a conventional non-time delayed diffraction, then the shaded bit in the fourth row would be inverted. The last two rows correspond to a situation, where both first two input pulses are present. In this case, the third bit is inverted. Such truth table corresponds to a logic operation "controlled, controlled NOT".

Fig. 6 shows results of a experiment, where we implemented the interference of the two-pulse echo

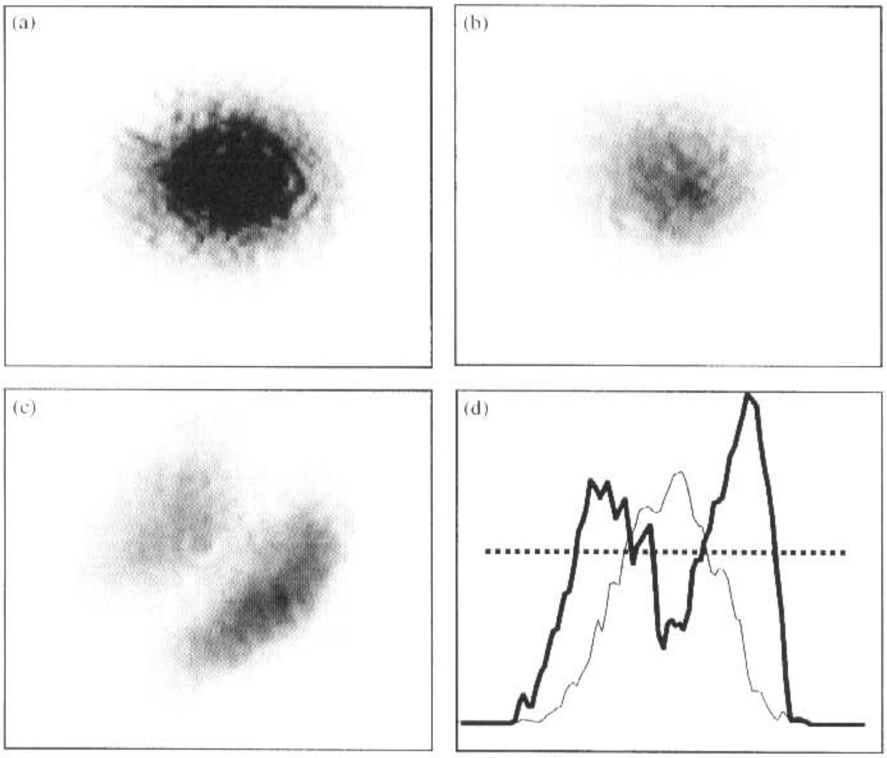

Fig. 6. Spatial intensity distribution measured at the output of the sample in the direction of the third beam for different input pulse combinations. Darker color corresponds to higher intensity. (a) Transmitted third pulse if either first or second (or both) input pulses are blocked; (b) two-pulse echo without interference when the first and second input pulses are present and third pulse is absent; (c) interference picture observed when all three input pulses are present. Both destructive and constructive interference is observed depending on the spatial region of the wave front; (d) one-dimensional intensity profiles measured in horizontal direction for (b) (thin line) and (c) (bold line). Dashed line shows the threshold for discrimination. Intensity below the threshold corresponds to destructive interference.

with a third pulse applied at the time and in the propagation direction of the echo signal. The excitation pulses were derived from a 150-fs amplified Ti : sapphire laser and the interference was detected using a CCD camera. The amplitude of the third pulse was adjusted to maximize the contrast of the destructive interference (for further experimental details see Ref. [32]). Fig. 6(a) shows the spatial image of the third pulse at the output of the sample when the first and the second pulses were blocked at the input. Fig. 6(b and c) show the image of the photon echo spatial intensity without and with the third input beam present. In Fig. 6(c) we can clearly observe a spatial region, where the destructive interference takes place, while at other spatial regions a constructive interference is present. This is due to the fact that the spatial beam profile of the excitation pulses was far from being ideal plane waves. Nevertheless, by selecting an appropriate spatial region of the beam cross-section and by implementing a suitable intensity threshold (Fig. 6d), we can identify the necessary input and output values, which reveal the properties of our truth table. To further discuss our results, let us note that the photon echo logic gate makes use of a coherent superposition of the molecules in the ground and excited state. The resonant medium remains in a coherent excited state for a time T_2. The coherence time may be much longer than the interference process, which is accomplished on a time scale $\sim 2t_p$. We can make use of the long coherence time by applying more than three pulses, which would interfere with corresponding echo amplitude. This would allow us to construct a truth table, which would implement a logic operation with N inputs and N outputs.

The particular truth table considered in our present experiment is known as a Taffoli gate. In recent discussion about quantum computing algorithms Taffoli gate is considered as one of several possible universal gates [33]. A distinctive feature of a quantum computer is that its operations are based on manipulating entangled superposition states of quantum-mechanical systems such as atoms or spins [34]. In our present realization of a Taffoli gate the coherent superposition concerns only one molecule at a time, rather than an entanglement of several molecules. On the other hand, the coherence exists in the medium for time T_2, during which the molecules at different frequencies can interact and build up some sort of mutual coherence. Controlling this coherence would allow to manipulate data coded into a coherence superposition of atoms or molecules at different frequencies. It remains open for a further discussion if the inhomogeneous broadened media can be used for implementing quantum computing algorithms.

5. Conclusions

We have demonstrated that frequency-selective materials can be used to extend the ideas of optical processing into the new dimensions of frequency and time. We have discussed conditions to achieve ultrafast performance on the time scale of 10^{-13} s by using special resonantly absorbing media with broad inhomogeneous band and dipole-allowed transitions, such as organic dye-doped polymers at liquid-helium temperature. We have experimentally demonstrated an ultrafast bit-to-bit multipication by photon echo using spectrally tailored ultrashort pulses. We have also implemented a coherent logic gate based on time-domain interference of ultrafast photon echo, performing an operation of "controlled, controlled NOT".

Acknowledgements

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