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Terahertz bit-rate parallel multiplication by photon echo  
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## Abstract

We demonstrate multiplication of two 8 bit binary words in less than 10 ps by generating two-pulse photon echo in a cryogenically-cooled polymer film doped with organic dye molecules. Input bits are coded in the intensity spectrum of 100 fs duration excitation pulses and the outcome of the multiplication is obtained by measuring the intensity spectrum of the photon echo signal. We study the dependence of the echo spectrum on the energy of the excitation pulses and investigate excitation conditions when the photon echo has maximum efficiency and minimum distortions. © 1998 Elsevier Science B.V.

## 1. Introduction

Photon echo [1,2] has been long considered as a possible method for future high-speed parallel optical data storage and processing, including holography [3–7]. Practical uses of photon echo phenomena (as well as of related methods of spectral hole burning holography [8,9]) are based on the fact that absorption bands of atoms or molecules trapped in solids are strongly inhomogeneously broadened and comprise at low temperatures very narrow homogeneous zero-phonon lines [10]. Materials like polymers doped with dye molecules and crystals containing rare-earth ions exhibit, especially at liquid-helium temperatures, a ratio between the inhomogeneous and homogeneous line width as large as  $\Gamma_{\text{inh}}/\Gamma_{\text{ZPL}} \sim 10^4$ – $10^6$ . Such highly degree of selectivity in frequency domain allows coherent optical responses or echo to be excited simultane-

ously at a large number of different frequencies using a single focussed laser spot. By virtue of Fourier transformation, the frequency-domain response of zero-phonon lines can be used to process time-domain information and vice versa – coherent superposition frequency-domain response amplitudes can play back time-domain signals. The shortest echo pulse duration is given by the inverse value of the inhomogeneous band width,  $\delta\tau_{\text{min}} \sim (\Gamma_{\text{inh}})^{-1}$ , whereas the maximum duration of the coherent response is determined by the inverse value of the zero-phonon line width (optical dephasing time),  $\tau_{\text{max}} \sim T_2 = (\pi\Gamma_{\text{ZPL}})^{-1}$ .

Photon echo has been used to convolute and correlate time domain optical wave forms [11,12], to perform bit-to-bit multiplication of time domain data trains [13] and to carry out time-domain associative recall and pattern recognition [14]. On this basis it has been suggested that photon echo devices could be used as building blocks for future ultra high speed computers and data routers [15–17].

In Ref. [18] two-pulse photon echo was used to perform bit-to-bit multiplication of two 8 bit words at 1 MHz data rate in a  $\text{Pr}^{3+}:\text{YAlO}_3$  crystal. By using faster modulation techniques and shorter and more energetic excitation pulses

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parallel processing on nanosecond time scale has been demonstrated [19–21]. However, the exploited two-pulse or stimulated photon echo techniques enabled to perform the processing in a single-shot [18–21]. However, because the inhomogeneous band width of rare earth materials is typically limited to a few hundreds of GHz, in order to achieve even faster processing at terahertz data rates, alternative materials with considerably broader inhomogeneous band width are required.

Organic polymers doped with dye molecules have inhomogeneous band width typically in the range of 5–10 THz [22]. By using polymer films containing porphyrin-like dye molecules photochemically accumulated stimulated photon echo has been applied to perform optical storage and processing experiments with femtosecond time resolution [23–25]. In this paper we carry out real-time multiplication of two 8 bit binary words at a terahertz data rate by generating two pulse photon echo in an organic dye-doped polymer film. In contrast to earlier femtosecond experiments, which used accumulated gratings and low intensity high repetition rate laser pulses on the time scale of several seconds, in the present experiment we apply energetic femtosecond laser pulses and carry out parallel processing in a single shot. We code the input bits into the intensity spectrum of the excitation pulses by using a two-channel pulse shaper and read out the result of the multiplication by measuring the intensity spectrum of the photon echo signal. We also carry out measurements to determine optimal conditions for the multiplication process where photon echo efficiency is maximal and distortions of the echo spectrum are minimal.

## 2. Theory

The origin of photon echo is in coherent optical recall or scattering from an inhomogeneously broadened ensemble of molecules as a response to resonance excitation at frequency  $\nu_0$  of the transition from the ground state to an excited electronic state. Different types of echoes such as two-pulse photon echo [26], three-pulse or stimulated photon echo [27] and various accumulated photon echoes [28,29] are observed depending on the time scale and on the physical nature of the nonlinearity of the optical response [30]. In the case of two-pulse photon echo, if the first excitation pulse is applied at moment  $t_1 = 0$  propagating in the direction of wave vector  $\mathbf{k}_1$ , and the second pulse is applied at moment,  $t_2 < T_2$ , in the direction of wave vector  $\mathbf{k}_2$ , then the echo is observed at time  $t = 2t_2$  in the direction of wave vector  $\mathbf{k}_{2PPE} = 2\mathbf{k}_2 - \mathbf{k}_1$ . If the intensity of the excitation pulses does not exceed

$$I_{\max} = \frac{\varepsilon_0 c}{2} \left( \frac{\hbar \Delta \nu_p}{|d|} \right)^2, \quad (1)$$

where  $d$  is the dipole moment of the electronic transition,

$\hbar$  is Planck's constant,  $\Delta \nu_p$  is the spectral width of the pulses and  $\varepsilon_0$  is the vacuum permeability, then the echo signal amplitude can be approximated by a linear superposition of the response amplitudes produced by each frequency component of the incident light. If the spectral width of the inhomogeneous absorption band is large compared to the spectral width of the pulses and provided that the angle  $\alpha$  between the excitation beams is small enough to satisfy the wave matching condition [31], then the amplitude of the echo can be presented in the frequency dimension as [4,15,30]

$$E_{\text{echo}}(\nu) \propto E_1^*(\nu) E_2(\nu), \quad (2)$$

where the frequency-domain amplitudes are defined by Fourier transformation of time-domain pulse amplitudes ( $i = 1, 2$ )

$$E_i(\nu) = \int E_i(t') e^{-i2\pi\nu t'} dt', \quad (3)$$

Let us consider two pulses with intensity spectrum  $A(\nu)$  and  $B(\nu)$  incident upon a sample with a broad inhomogeneous absorption spectrum,  $\Delta \nu_p < \Gamma_{\text{inh}}$ . The intensity spectrum of the two-pulse photon echo is then proportional to

$$I^{\text{echo}}(\nu) \propto A(\nu) B^2(\nu). \quad (4)$$

This relation reflects the fact that at low excitation intensities, if a certain frequency component is not present in the intensity spectrum of one or both excitation pulses, then this frequency will be absent also from the intensity spectrum of the echo signal. We divide the spectrum of the excitation pulses into  $N$  frequency components, where we assign bit value '1' to a frequency component that is present and bit value '0' to a frequency component that is absent. If the echo is excited with pulses corresponding to two  $N$ -bit words,  $A = (\nu_1, \nu_2, \dots, \nu_N)$  and  $B = (\nu_1, \nu_2, \dots, \nu_N)$ , then the  $i$ th frequency component of the echo intensity spectrum will be

$$I_i^{\text{echo}} = A_i B_i, \quad i = 1, 2, \dots, N, \quad (5)$$

i.e. the echo represents the result of bit-to-bit multiplication of the two input words. In this paper we use two-pulse photon echo to carry out bit-to-bit multiplication on ultra-fast time scale. In order to determine the range of applicability of Eq. (5) we investigate the relation between the photon echo intensity and spectrum to the intensity of the excitation pulses.

## 3. Experimental

As a laser source we use a commercial regenerative Ti:sapphire laser amplifier (Clark MXR CPA-1000). The system comprises a self mode-locked femtosecond Ti:sapphire laser oscillator, a grating pulse stretcher, a Ti:sap-

phire regenerative amplifier and a grating pulse compressor [32]. The oscillator is pumped by a CW Ar-ion laser (Spectra-Physics, BeamLok 2060) and provides 70 fs duration pulses with a repetition rate of 100 MHz at a wavelength of 770–780 nm. The regenerative amplifier is pumped by a 1 kHz repetition rate Nd:YAG laser. The compressed output pulses have a duration of 80–120 fs and an energy of 0.6–0.9 mJ per pulse.

A scheme of the experimental set-up is shown in Fig. 1a. The beam from the femtosecond laser is split into two equal parts which are then directed through a two-channel pulse shaper which encodes data bits into the respective intensity spectrum of the two pulses. The frequency-coded pulses are then superimposed on the sample positioned in an optical liquid-helium cryostat. The angle between the excitation beams is  $\alpha = 4^\circ$ . To measure the spectrum of the echo signal and of the excitation pulses we use a grating monochromator SPEX 1412 equipped with a CCD-array parallel detector and a digital oscilloscope Tektronix TDS 520A to read out the data. To obtain absolute energy measurements, we use a pyroelectric joulemeter Moletron J3S-10. The temporal shape of the echo and of the excitation pulses is monitored by measuring second-harmonic cross-correlation in a 0.5 mm thick  $\text{LiIO}_3$  crystal.

The sample is a polyvinylbutyral film doped with Zn-naphthalocyanine-type dye molecules (Ciba dye HW3463) at a concentration of  $10^{-4}$  mol/l [33]. The geometric thickness and cross-section of the film are  $100 \mu\text{m}$  and  $18 \times 22 \text{ mm}^2$ , correspondingly. The optical density of the sample is  $\text{OD} = 1.7$  at the wavelength of 770 nm absorption maximum corresponding to the  $S_1 \leftarrow S_0$  transition of

the dye molecules. The inhomogeneous band has a nearly Gaussian shape with a width of 30 nm ( $\sim 500 \text{ cm}^{-1}$ ), which corresponds to an ultimate time resolution for Gaussian-shaped pulses of about 30 fs. For our purpose it is important also that the absorption band overlaps with the wavelength of the femtosecond excitation pulses and that in this type of dye molecules mechanisms for permanent hole burning are suppressed.

Fig. 1b shows the details of the pulse shaper device. It involves two identical diffraction gratings (2000 lines/mm) and two identical cylindrical lenses ( $f = 150 \text{ mm}$ ) positioned symmetrically with respect to the spectral plane [34]. In this plane, the frequency components of the laser pulses are spread out in space in horizontal direction. The two laser beams pass at different vertical heights. This allows us to place two different masks, MA and MB in the spectral plane so that we can cut out different frequency components of each of the beams independently.

#### 4. Results and discussion

Our first experimental task is to measure the spectrum of the photon echo signal. The excitation is carried out with two spectrally-coded pulses,  $A = (1, 0, 1, 0, 1, 0, 1, 0)$  and  $B = (0, 1, 0, 1, 0, 1, 0, 1)$ , each containing one 8 bit word. For this purpose the spectrum of the laser pulses is modulated at 8 prefixed frequencies. To minimize distortions, only the central flat portion of the laser spectrum is used. The bits are coded by two masks made out of thin opaque ribbons positioned in the spectral plane of the pulse shaper. Frequencies of the excitation pulses corre-

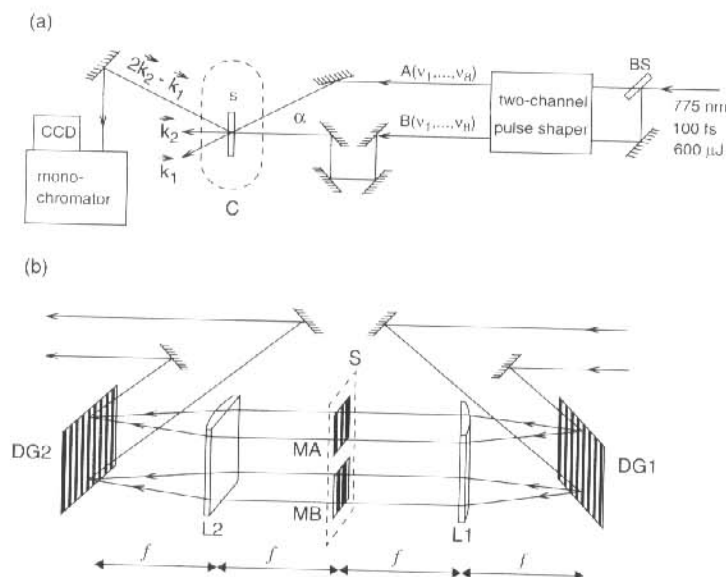


Fig. 1. Scheme of the experimental set-up. (a) Optical layout. BS: 50% beam splitter; C: cryostat; CCD: detector array. (b) Scheme of the pulse shaper. DG1, DG2: diffraction gratings; L1, L2: cylindrical lenses; MA, MB: spatial transmission masks; S: spectral plane.

sponding to bit value '0' are blocked, while the frequencies corresponding to bit value '1' are transmitted. Figs. 2a and 2b show the intensity spectrum of the first and the second excitation pulse at the output of the pulse shaper. The intensity contrast between the minima and maxima is better than 1:10. At the sample the energy of the excitation pulses is  $20 \mu\text{J}$  and the total illuminated area is about  $1 \text{ cm}^2$ . We note that in the present setup the frequency resolution of the grating pulse shaper is about  $2 \text{ cm}^{-1}$ . The spectral width of the modulated frequency channels is mainly determined by the width of the opaque ribbons which is about  $10 \text{ cm}^{-1}$ . To avoid overlap between neighboring bits we leave ample intervals of  $40 \text{ cm}^{-1}$  between the channels.

Fig. 2c shows the intensity spectrum of the photon echo signal. In agreement with Eq. (5), the spectrum contains only those frequency components, which are present in both excitation pulses. The excitation pulse spectra overlap only in the narrow intervals between the bits and at the far wings of the laser spectrum. Accordingly, in all 8 specified frequencies the resulting bit value is '0' and we obtain as result of multiplication following 8 bit word,  $I^{\text{echo}} = (0, 0, 0, 0, 0, 0, 0, 0)$ .

To demonstrate the speed of the multiplication process, we measured the time-domain intensity profile of the excitation pulses and of the photon echo signal. Fig. 2d shows that the overall duration of the pulse sequence consisting of the photon echo and the excitation pulses is about 25 ps, while the duration of individual pulses is about 4–6 ps. Because 8 bits are processed in parallel, we can estimate that the bit rate in this multiplication process

is over 1 THz. As compared to the previous bit-to-bit multiplication experiments performed with rare-earth materials [18], our result represents an increase of 6 orders of magnitude in the speed of processing per bit in one spatial channel.

The energy density of the excitation pulses at the sample is  $20 \mu\text{J cm}^{-2}$ . The maximum relative efficiency of the echo observed at frequencies where the spectra of the excitation pulses overlap is about 0.1–0.2%. Since our present experiment is carried out with unfocused beams, we can estimate that the energy needed to excite the echo with the same efficiency on an area of  $\lambda^2$  is about  $10^{-12} \text{ J}$ . The other interesting point to notice is that as long the homogeneous line width ( $F_{\text{ZPL}} \sim 0.01 \text{ cm}^{-1}$  at  $T = 2 \text{ K}$ ) is much less than the width of the frequency interval assigned to one bit, the efficiency of the echo remains constant if the number of bits is increased. This means that, in principle, we can multiply  $10^3$ – $10^4$  bits in a single shot by using the same amount of laser energy, provided that we are able to efficiently modulate the spectrum of the pulses with a precision of  $0.01 \text{ cm}^{-1}$ . We can estimate that the minimum amount of energy needed to multiply two bits in this case will be as low as  $10^{-15}$ – $10^{-16} \text{ J}$ , which corresponds to about  $10^3$ – $10^4$  photons.

The multiplication procedure by photon echo incorporates a time delay that needs to be introduced to prevent the excitation pulses from overlapping in time. In the present experiment the peaks of the excitation pulses are separated by 12 ps, while the trailing end of pulse A and the front end of pulse B have a separation of a few picoseconds. If the pulses A and B overlap in time, then

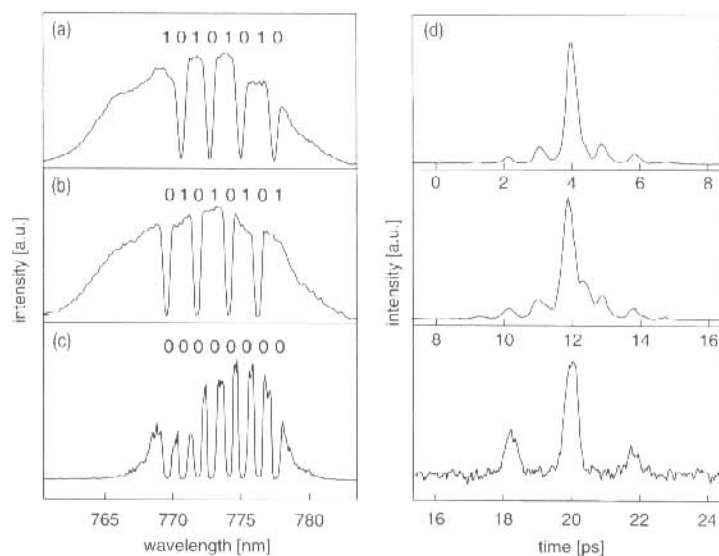


Fig. 2. Bit-to-bit multiplication by photon echo. (a) Intensity spectrum of excitation pulse A in the direction  $\mathbf{k}_1$  (first pulse). (b) Intensity spectrum of excitation pulse B in the direction  $\mathbf{k}_2$  (second pulse). (c) Intensity spectrum of the photon echo signal in the direction  $2\mathbf{k}_2 - \mathbf{k}_1$ . The energy of the excitation pulses is  $I_A = 20 \mu\text{J cm}^{-2}$  and  $I_B = 20 \mu\text{J cm}^{-2}$ . Temporal intensity profile of the excitation pulses A (d) and B (e) and of the echo (f) measured by cross-correlation with the original laser pulse.

some part of the echo signal propagates in a conjugated diffraction direction instead of the present detection direction,  $2\mathbf{k}_1 - \mathbf{k}_2$  [35]. This leads to a loss of signal intensity and, more important, causes distortions of the echo intensity spectrum (see Ref. [24] for more detailed discussion). Another reason why temporal overlap has to be avoided, is because at small delays  $t < 0.5$  ps not only the zero-phonon line, but also the accompanying phonon side band contributes to the echo signal [35,36]. If the delay is too short, then the echo signal originating from the broad phonon side band gives rise to a broad background in the measured intensity spectrum which can eventually distort the outcome of the bit-to-bit multiplication.

It can be noticed from Fig. 2c that the overall echo intensity drops at the shorter wavelength side of the spectrum. Although in the present experiment this small distortion does not affect the fidelity of the multiplication, we can explain this asymmetry by noting that in our sample the relative contribution of zero-phonon lines to the overall absorption is larger at the longer wavelength side of the spectrum, whereas at the shorter wavelength side the largest contribution comes from the phonon side bands (composition of inhomogeneous absorption bands in dye-doped polymers is discussed in Ref. [37]). Although in our experiment the echo signal was restricted to the zero-phonon lines, the absorption by phonon side bands is still responsible for the slight distortion of the spectrum at the short wavelength side of the inhomogeneous band.

Further we investigate the dependence of the photon echo signal on the energy of the excitation pulses. In order to guarantee uniform intensity over the illuminated area, we restrict the cross-section of the excitation beams by placing a  $0.1 \text{ cm}^2$  pinhole in front of the sample in the center of the beam. The excitation pulse energy is varied by using a set of neutral-density filters. We measure the energy of the echo pulses as we change the energy density of one of the excitation beams by keeping the energy density of the other beam constant. In this measurement we remove the spectral shaping masks so that both excitation pulses have smooth spectral profile with  $170 \text{ cm}^{-1}$  width and a duration of 100 fs.

Fig. 3 shows the result of the measurement. The data points marked by asterisks are obtained by varying the energy of the first excitation pulse and open circles correspond to data points, where the energy of the second pulse was varied. If the energy of the second pulse is  $1200 \text{ mJ cm}^{-2}$ , then below  $100\text{--}200 \text{ } \mu\text{J cm}^{-2}$ , the dependence of the echo signal on the energy density of the first excitation pulse is close to a linear function. At the same time, if the energy of the first pulse is  $300 \text{ } \mu\text{J cm}^{-2}$ , then at energy densities below  $500\text{--}1000 \text{ } \mu\text{J cm}^{-2}$ , the dependence of the echo on the second pulse is closer to a quadratic function. These observations are qualitatively in agreement with Eqs. (1) and (4), which predict this type of dependence for low- and moderate-intensity excitation pulses. By inserting into Eq. (1) the estimated value of the transi-

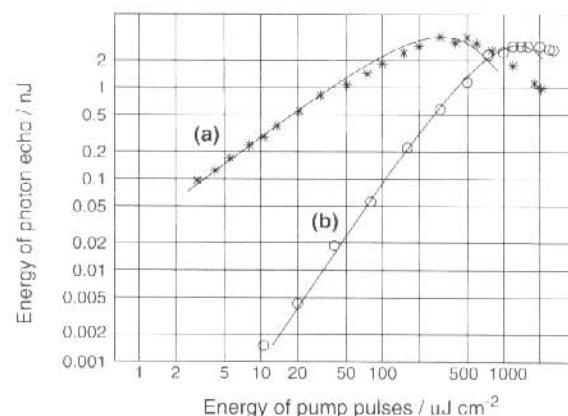


Fig. 3. Energy of the photon echo pulse as a function of the energy of the excitation pulses. (a) The energy of the first pulse is varied, while the energy of the second pulse is a constant,  $I_B = 1200 \text{ } \mu\text{J cm}^{-2}$ . (b) The energy of the second pulse is varied, while the energy of the first pulse is a constant,  $I_A = 300 \text{ } \mu\text{J cm}^{-2}$ . The solid curves are the results of the simulations based on numerical solution of the optical Bloch equations.

tion dipole moment of the dye molecules,  $d = 10^{-28} \text{ C m}$ , we obtain for the critical maximum illumination intensity,  $I_{\text{max}} \approx 0.15 \text{ GW cm}^{-2}$ . Fig. 3 shows that the experimental dependence on the intensity of the first excitation pulse begins to deviate from linear at a slightly higher value,  $I_{\text{max}} \approx 1.0 \text{ GW cm}^{-2}$ , which is still in reasonable agreement with the approximate formula of Eq. (1). Fig. 3 also presents the results of the theoretical calculations of the echo intensity that we performed based on the numerical solution of optical Bloch equations. Considerable deviation from the theory occurs at very high excitation intensities, where the echo signal starts to decrease with increasing excitation energy density. To account for these effects more elaborate calculations will be carried out in the future.

Fig. 4 shows the spectrum of the echo, when we use again the spectral modulation of the excitation pulses described above, but with the energy density increased to  $80 \text{ } \mu\text{J cm}^{-2}$ . We see that at high excitation intensities we start to observe frequency components in the intensity spectrum of the echo, which were not present if the intensity of pulses was low. This result shows that at high intensity of the excitation pulses the behavior of the photon echo considerably deviates from the dependence described by Eq. (4) in the sense that the echo spectrum contains frequency components that are not present in both excitation spectra. From our present results we can conclude that optimum conditions for high-speed bit-to-bit multiplication in the present dye-doped polymer are achieved at spectral excitation energy density of  $20 \text{ } \mu\text{J cm}^{-2}$  per  $170 \text{ cm}^{-1}$  frequency interval.



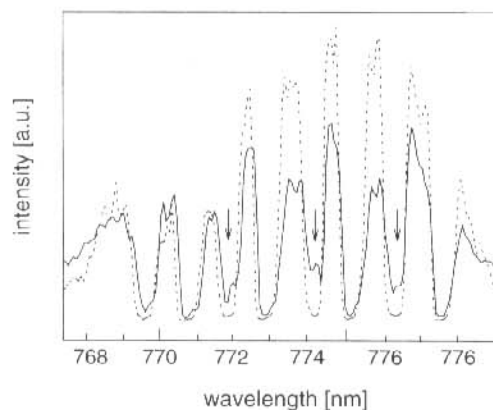


Fig. 4. Solid line—intensity spectrum of the photon echo at high excitation intensity  $I_A = I_B = 80 \mu\text{J cm}^{-2}$ . Dashed line: intensity spectrum at low excitation intensity  $I_A = I_B = 20 \mu\text{J cm}^{-2}$ . Arrows indicate frequency components that are not present in the intensity spectrum of both excitation pulses.

## 5. Conclusions

We have performed an experiment where we demonstrate parallel bit-to-bit multiplication of two 8 bit binary words in less than 10 ps, which corresponds to a bit rate of about 1 THz. This is achieved by generating a two-pulse photon echo in a cryogenically-cooled polymer film doped with organic dye molecules using intense 100 fs duration pulses for excitation. We have shown that information can be coded by modulating the intensity spectrum of the excitation pulses and the result of the multiplication procedure can be read out by measuring the intensity spectrum of the photon echo signal.

We have also studied the dependence of the echo spectrum on the energy density of the excitation pulses. At low energy density, the echo intensity is a linear function of the energy of the first excitation pulse and a quadratic function of the energy of the second excitation pulse. At high excitation energy density, above  $80 \mu\text{J cm}^{-2}$ , saturation and decrease of the echo intensity is detected. We also show that at high excitation intensity the echo spectrum contains frequency components that are not present in the intensity spectrum of the excitation pulses. We determine that optimum conditions for parallel bit-to-bit multiplication are achieved at energy densities of about  $20 \mu\text{J cm}^{-2}$ .

Our results demonstrate that the low-temperature dye-doped polymers are suitable materials for ultrafast parallel frequency-domain optical data processing on the time scale of  $10^{-12}$ – $10^{-13}$  s per bit.

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