



Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals

Publication details, including instructions for authors and
subscription information:

<http://www.tandfonline.com/loi/gmcl19>

Spectral and Time Domain Studies of Accumulated Photon Echo

I. Zeylikovich^{a b}, G. Bai^{a b}, A. Gorokhovskiy^{a b c} & R. R. Alfano^{a b}

^a Institute for Ultrafast Spectroscopy and Lasers, New York State
Center for Advanced Technology for Ultrafast Photonic Materials
and Applications

^b Department of Physics, The City College and The Graduate
School of The City University of New York, New York, NY, 10031
Phone: (212) 650-5531 Fax: (212) 650-5531

^c Department of Applied Sciences, The College of Staten Island,
The City University of New York, 2800 Victory Boulevard, Staten
Island, NY, 10314

Version of record first published: 04 Oct 2006.

To cite this article: I. Zeylikovich, G. Bai, A. Gorokhovskiy & R. R. Alfano (1996): Spectral and Time Domain Studies of Accumulated Photon Echo, Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals, 291:1, 277-285

To link to this article: <http://dx.doi.org/10.1080/10587259608042757>

PLEASE SCROLL DOWN FOR ARTICLE

Full terms and conditions of use: <http://www.tandfonline.com/page/terms-and-conditions>

This article may be used for research, teaching, and private study purposes. Any substantial or systematic reproduction, redistribution, reselling, loan, sub-licensing, systematic supply, or distribution in any form to anyone is expressly forbidden.

The publisher does not give any warranty express or implied or make any representation that the contents will be complete or accurate or up to date. The accuracy of any instructions, formulae, and drug doses should be independently verified with primary sources. The publisher shall not be liable for any loss, actions,

claims, proceedings, demand, or costs or damages whatsoever or howsoever caused arising directly or indirectly in connection with or arising out of the use of this material.

SPECTRAL AND TIME DOMAIN STUDIES OF ACCUMULATED PHOTON ECHO

I. ZEYLIKOVICH, G. BAI, A. GOROKHOVSKY^{a)} and R. R. ALFANO
Institute for Ultrafast Spectroscopy and Lasers, New York State Center for
Advanced Technology for Ultrafast Photonic Materials and Applications,
Department of Physics, The City College and The Graduate School of The City
University of New York, New York, NY 10031. TEL. (212) 650-5531. FAX. (212)
650-5530.

Abstract Multishot or single-shot retrieval of femtosecond echo signal without substantial spectral distortion was experimentally demonstrated for a H₂-octaethylporphine doped polystyrene sample. Upto 10⁴ single-shot readout cycles using a femtosecond pulse were observed without substantial echo signal degradation. The echo decay time was determined for this sample to be (50 ± 15) ps.

INTRODUCTION

Persistent hole burning (PHB) media^{1,2} has been proposed for ultrahigh-density optical data-storage applications because of their high spectral selectivity and long storage times. Frequency^{3,4} and time-domain⁵ approaches have been proposed for writing and reading streams of data bits. In the frequency domain, monochromatic light is employed to write and read data sequentially into frequency channels. The upper limit for readout speed of a data bit is determined by the minimum width of a frequency channel or the homogeneous linewidth (FWHM) Γ_H , typically 1 kHz - 1GHz at liquid-helium temperature. In the time-domain, data bits are written and recalled in parallel by a spectrally broad laser pulse. In this case, a read-out speed of a data bit is much higher than in the frequency-domain approach and is limited by the width of the

a) Current address: Department of Applied Sciences, The College of Staten Island, The City University of New York, 2800 Victory Boulevard, Staten Island, NY 10314.

inhomogeneous absorption band Γ_1 of the storage material, typically of 10 GHz -10 THz.

Utilization of the highest possible readout speed requires femtosecond optical pulses. Femtosecond PHB holography was demonstrated for storage, recall and conjugation of accumulated echo signals. Signals as short as 100 fs were detected using a standard multishot second-harmonic generation technique.⁶ To increase the readout speed of femtosecond accumulated photon echo signals, an ultrafast single-shot interferometric crosscorrelation technique was proposed.⁷ A data readout speed as fast as 27 Tbits/s was experimentally demonstrated for a H₂-octaethylporphine (OEP) doped polystyrene sample. The observed amplitude correlation time (ACT) of the echo signal was approximately twice as long as the ACT of the readout pulse. ACT broadening could be caused by a distortion of the echo spectrum.⁸

In this work, the PE spectra were measured and analyzed to minimize spectral distortion and to improve the time resolution of the crosscorrelation technique for PE signal from a OEP-doped sample. High intensity pulses, can introduce distortion and degradation of the PE signal. The dependence of PE intensity on the number of the single-shot readout cycles and PE decay time were measured.

EXPERIMENTAL METHOD

A schematic of the experimental arrangement is shown in Figure 1. The optical pulses were generated by a colliding-pulse mode-locked (CPM) laser operating at a repetition rate of 125 MHz, 6-mW average power, with 100 fs pulse duration. The amplifier system was pumped by a Q-switched ND:YAG laser at 20-Hz repetition rate and produced an amplified single pulse with an energy of 3 μ J/pulse.

A OEP- doped polystyrene film at $T = 1.4$ K ($OD = 0.96$ at $\lambda = 618$ nm) was used as the holographic medium. The signal pulse was stored in the sample after exposure by superimposed reference and signal beams. Average CPM laser power of 1 mW and a 0.8 mm - diameter spot were used. The echo signal was recalled using the multishot CPM

pulses with average readout intensity of about 0.04-mW, or by a single amplified pulse of 150 fs duration with an energy of 0.7 μ J. The two spectra from PE and

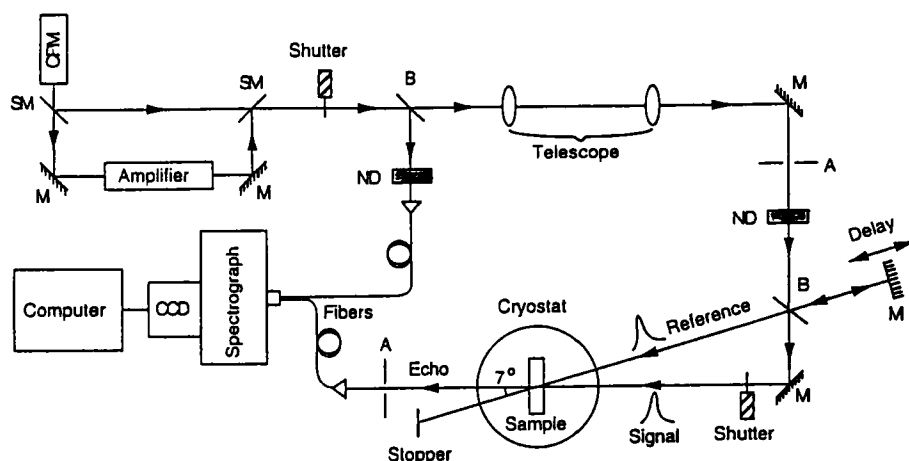


FIGURE 1 Experimental arrangement for hole-burning holography using CPM laser pulses: SM's, transfer mirrors; M's, mirrors, ND's, neutral density filters; B's, beam splitters; A's, aperture.

readout pulses were recorded simultaneously by a spectrograph and a CCD sensor (Photometrics Ltd., Model LC 200). The spectrum of transmitted pulses was measured using the signal beam (reference beam was blocked).

EXPERIMENTAL RESULTS

Hologram Writing

The dependence of PE spectral intensity on writing exposure is shown in Figure 2. At low exposure level, PE intensity depends on exposure almost linearly, and saturates at the level of 6 J/cm². This behavior is related to the saturation of the narrow hole shape at high exposures in hole-burning materials,⁹ which causes hole broadening, a decrease of spectral grating contrast, and shortening of the PE decay time.¹⁰ To minimize such

effects, a low exposure level of 1 J/cm^2 was used in the experiments, corresponding to an exposure time of about 5 s for the CPM laser pulse energy of the order of 40 pJ with 6×10^8 writing pulse sequences.

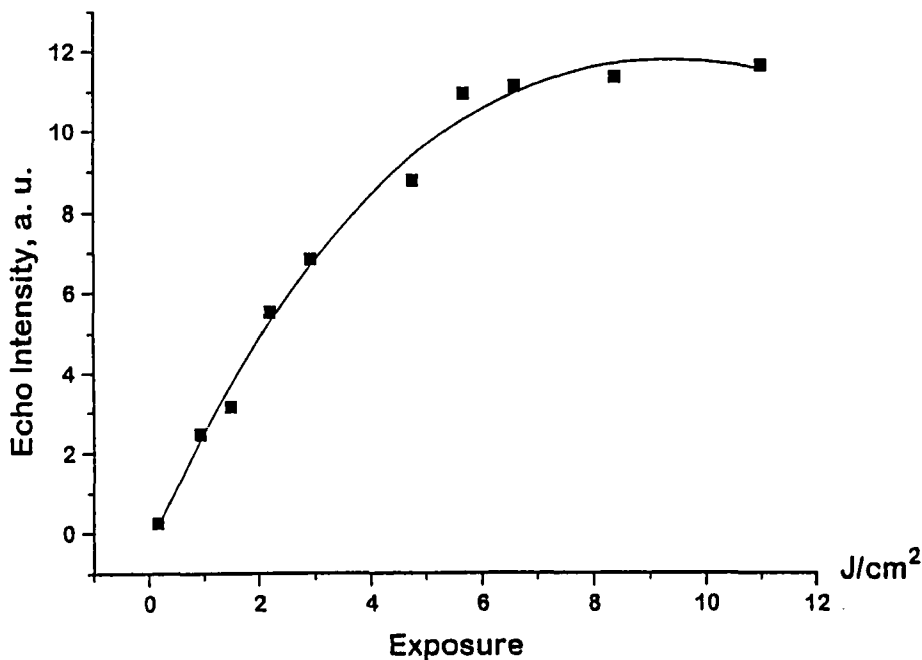


FIGURE 2 The dependence of the normalized multishot echo intensity from exposure.

PE Spectra

To determine spectral distortion from the OEP-doped sample, PE spectra for holograms written by CPM laser at two wavelengths, 620 nm, or 616 nm, were studied. Figs. 3a and b show the spectra of the PE (solid) and readout pulses (dashed line) for the CPM multishot readout mode. For laser wavelength of 620 nm, the PE spectrum consists of two bands. The most intense band at 622 nm is more than twice as narrow ($\text{FWHM} \approx 2.5 \text{ nm}$) as the readout pulse spectrum. As a result, the ACT of the PE signal became twice as long. The PE spectrum changes (see Fig. 3b) when writing and readout were performed at 616 nm. The spectrum ($\text{FWHM} \approx 4 \text{ nm}$) became smooth and only about

10 % narrower than the readout pulses spectrum. Figs. 3c and 3d show the spectra of the PE and readout pulses (solid and dashed lines, respectively) for single-shot readouts.

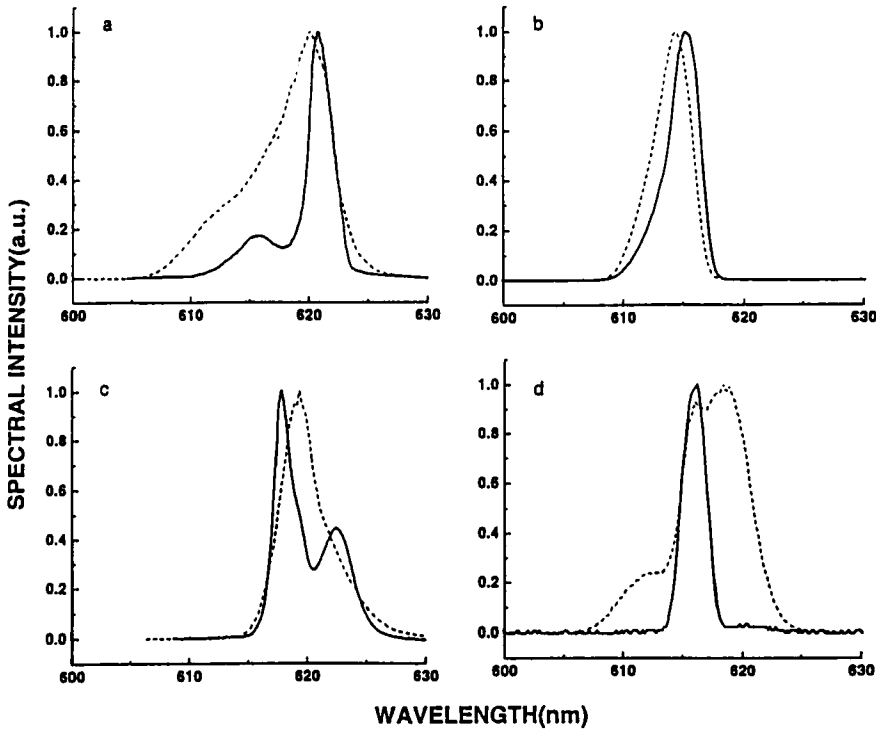


FIGURE 3 Spectra of the echo and readout pulses (solid and dashed lines, respectively). (a) and (b) for readout multishot CPM pulses, and (c) and (d) for readout single.

These spectra show the basic features of the multishot PE spectra.

PE Decay

The capacity of time-domain storage depends on the accumulated PE decay time, τ_{PE} . This time was determined using the multishot readout mode. The echo intensity was measured at the maximum of the echo spectrum and normalized to the intensity of the laser pulses for different delay times between the data and reference pulses.

Experimental data are shown in Fig. 4. The echo signal decays exponentially with the time constant $\tau_{PE} = 50 \pm 15$ ps.

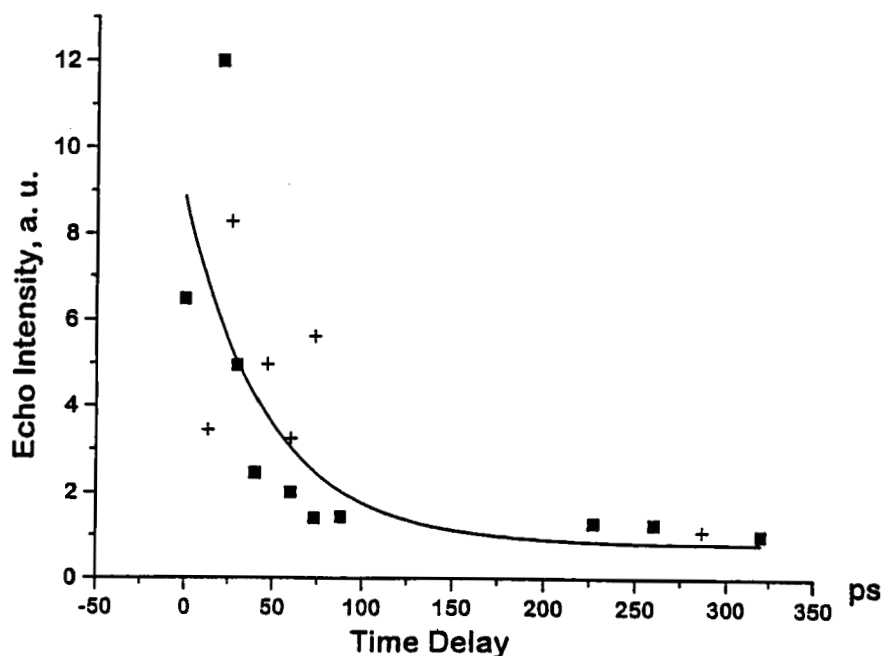


FIGURE 4 The normalized multishot echo intensity decay curve versus the time delay τ .

The accumulated echo in PHB media is a 3 pulse stimulated PE. Its decay is typically expressed as $I(t_{12}) = I(0)\exp(-4t_{12}/T_2^{3PE})$, where t_{12} is the delay time between the data and reference pulse, and T_2^{3PE} is the dephasing time.¹¹ For persistent hole-burning materials with long storage lifetime, the delay between reference and readout pulses, $t_{23} \sim 1-10$ s, and T_2^{3PE} includes optical dephasing and spectral diffusion effects over time ($t_{12} + t_{23}$).¹² We may compare T_2^{3PE} to the dephasing time determined from persistent hole-burning experiments which have the same time scale on the order of seconds, $T_2^H = 2(\pi\delta_v)^{-1}$, where $\delta_v = 2\Gamma_H$ is the holewidth (FWHM). For OEP-PS at temperature of 1.4 K, $\delta_v = 900$ MHz,¹³ what gives $T_2^H = 710$ ps. Therefore, on the basis of hole-burning data a decay time of 177 ps is expected for this material, more than $3\times$ longer than measured. This discrepancy is most likely related to a high peak

intensity of femtosecond pulses used in PE experiments ($\approx 10^8 \text{ W/cm}^2$), which is much higher than for CW lasers typically used for narrow band hole-burning ($\approx 0.01 - 1 \text{ W/cm}^2$). This effect may cause an excessive dephasing. Phonon-induced¹⁴ or instantaneous¹⁵ spectral diffusion, or nonequilibrium optical heating¹⁶ may be responsible for this difference.

Single-shot PE Readout Mode

The PE intensity dependence was investigated for a number of readout pulses (N in single-shot readout mode). The hologram was written by CPM laser pulse sequences and readout was performed by a single 150 fs pulse with energy of $0.3 \mu\text{J}$. The results are shown on Fig. 5 a.

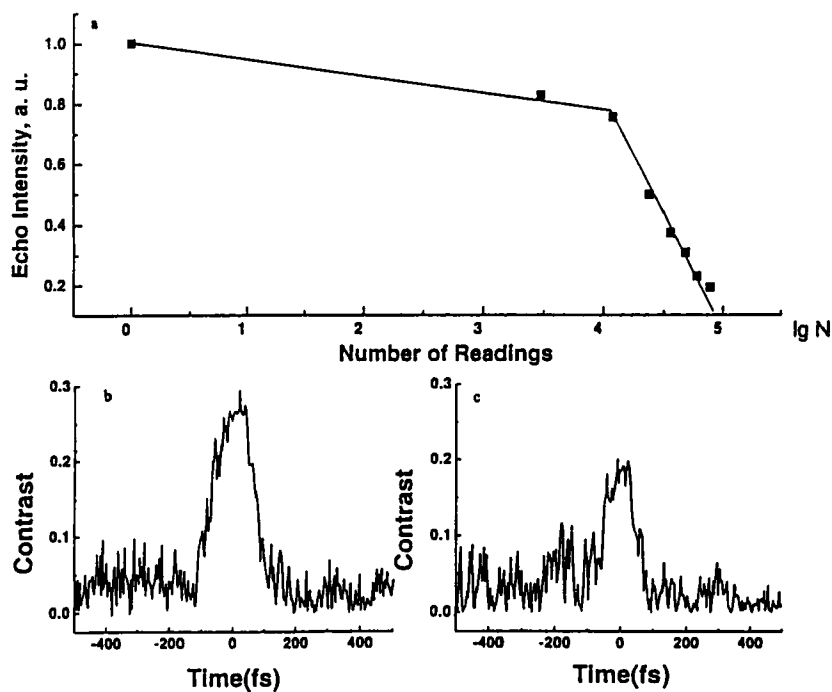


FIGURE 5 a) The decay of normalized single-shot echo intensity from the number N of the single-shot readout pulses (logarithmic scale) and b), c) crosscorrelation traces for $N = 1$ and $N = 2.4 \times 10^4$.

The echo intensity demonstrates a very slow degradation with increase in the number of readout pulses up to 2×10^4 , and much faster after that. The corresponding crosscorrelation traces for $N = 1$ and $N = 2.4 \times 10^4$ are shown in Fig. 5, b and c. Despite a poorer signal to noise ratio the data pulse can be easily separated from a background, even after 7.2×10^4 readouts. The “safe” number $N = 2 \times 10^4$ of readout pulses corresponds to a total reading exposure of 1.2 J/cm^2 , which is close to a writing exposure of 1 J/cm^2 . The interpretation is clear: the destructive readout gradually decreases the contrast of the hologram in the frequency domain. When accumulated reading exposure reaches the writing exposure, the hologram contrast became close to zero and the PE signal rapidly decreases. Thus, due to the high detection sensitivity, our single shot readout crosscorrelation technique allows for more than 10^4 readouts with reasonable signal to noise ratio.

CONCLUSION

These experiments demonstrate storage and multishot or single-shot retrieval of femtosecond echo signal without substantial spectral distortion for a OEP-sample. Upto 10^4 single-shot readout cycles with femtosecond pulses was observed without substantial echo signal degradation.

The authors wish to thank of A. Turukhin for help in carrying out this work. This research is supported in part by the U.S. Air Force Office of Scientific Research under contract F49620-931-0046 and by the New York State Technology Foundation.

REFERENCES

1. A. A. Gorokhovskiy, R. K. Kaarli, and L. A. Rebane, *JETP Lett.* **20**, 216 (1974).
2. B. M. Kharlamov, R. I. Personov, and L. A. Bykovskaya, *Opt. Commun.*, **12**, 191 (1974).
3. A. Szabo, *U. S. patent* 3,896,420 (July 22, 1975).
4. G. Castro, D. Haarer, R. M. Macfarlane, and H. P. Trommsdorf, *U. S. patent* 4,101,976 (July 18, 1978).
5. T. W. Mossberg, *Opt. Lett.*, **7**, (1982) 77.
6. A. Rebane, J. Aaviksoo, and J. Kuhl, *Appl. Phys. Lett.*, **54**, 93 (1989).

7. I. Zeylikovich, G. Bai, A. Gorokhovsky, and R. R. Alfano, Opt. Lett., **20**, 749 (1995).
8. P. Kaarli, A. Rebane, K. Rebane and P. Saari, J. of Luminescence., **45**, 401 (1990).
9. L. A. Rebane, A. A. Gorokhovskii, and J. V. Kikas, Appl. Phys. B., **29**, 235 (1982).
10. H. Sonajalg and P. Saari, J. Opt. Soc. Am. B., **11**, 372 (1994).
11. W. H. Hesselink and D. A. Wiersma, J. Chem. Phys., **75**, 4192 (1981).
12. M. Berg, C. A. Walsh, L. R. Narasimhan, K. A. Littau, and M. D. Fayer, J. Chem. Phys., **88**, 1564 (1988).
13. A. A. Gorokhovsky, Bull. Acad. Sci. USSR, Phys. Ser. **52**, 8 (1988).
14. Y. S. Bai and R. Kahru, Phys. Rev. B, **46**, 13735 (1992).
15. J. Huang, J. Zhang, A. Lezama, and T. W. Mossberg, Phys. Rev. Lett., **63**, 78 (1989).
16. A. A. Gorokhovsky, G. S. Zavt, and V. V. Palm, JETP Lett., **48**, 369 (1988).