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13. ABSTRACT ***(Maximum 200 Words)***

We have accomplished the principal goal of this project: we have performed an experiment which shows that capturing a holographic image of a unique ultrafast event on subpicosecond time scale in a single recording exposure is indeed practically possible. This was achieved, on one hand, by investigating spectral hole burning properties of a range of organic materials, and on the other hand, by carefully optimizing the experimental conditions. After several unsuccessful attempts, finally success was achieved by modifying a previously known spectral hole burning materials in way, which increased the recording efficacy by more than one order of magnitude. This allowed us to record image holograms with 150-fs duration pulses without need to accumulate the SHB effect from many exposures. Results of this research show that it is possible to perform optical recording of data in frequency-domain on ultrafast time scale. These results can be used also as a new diagnostic tool for femtosecond dynamics in various ultrafast optical interactions.

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ULTRAFAST HOLOGRAPHIC IMAGE RECORDING BY SINGLE SHOT FEMTOSECOND SPECTRAL HOLE BURNING

A. Rebane, Montana State University

1. Highlights

- *Our results on ultrafast recording & storage materials featured in ACST Newsletter*
- *Single-shot recording with femtosecond pulses achieved for the 1st time;*
- *Two-photon spectral hole burning with femtosecond pulses demonstrated for the 1st time;*
- *Technology of IR detection card licensed to a local optoelectronics company*

2. Summary of Results

2.1. Personnel.

The success of this project is largely due to our ability to attract qualified research associates and able students to MSU. In 1999 we hired a **Ph.D. scientist** *Dr. Mikhail Drobizhev* , originally from the world-renown Lebedev Physical Institute in Moscow. His specialty is in photophysics and photochemistry of low-temperature organic impurity solids. He is an expert on organic spectral hole-burning material, energy- and electron transfer in porphyrin complexes and aggregates. Since January 1999 he has been a very valuable addition to my group. Part of his salary is being paid by this grant, and part by other current grants. During the period of this project I had **two graduate students**. The first student worked until he fulfilled requirement for a Master's Degree and left for a position in IT-related industry. The second student has passed successfully qualification for Ph.D., and continues to working in my group. Both students have been trained in advanced techniques of femtosecond lasers, ultrafast holography, low-temperature spectroscopy and photochemistry.

2.2. New Lab Equipment

During this project we have essentially built up a new femtosecond laser laboratory starting from scratch. At the end of this grant period, the laboratory has a well-functioning femtosecond

amplified laser system with ample wavelength tuning capability. The system consists of a Ti:sapphire master femtosecond oscillator (Coherent Mira900) pumped by an Coherent Verdi 5 W CW frequency-doubled Nd:YAG laser. The pulses are amplified 1 kHz rate to 0.9 - 1.0 millijoule energy by a Ti:sapphire regenerative amplifier (Clark MXR CPA-1000), which was installed in summer 1998. An important improvement of the laser system was replacing the Ar-ion pump laser (Coherent Innova 400-25) with the solid-state pump laser. Big advantage of the solid state laser is that it has a much better beam pointing stability, and also a better TM00 spatial mode structure for better focusing on the Ti:sapphire crystal. To achieve necessary wavelength tuning in infrared and in visible, we have purchased and installed an optical parametric amplifier (Quantronix Topas). For hologram detection, a cooled high-resolution CCD camera has been purchased and installed.

2.3. Results of the Experiments

2.3.1. Increased efficiency of spectral hole burning.

One of most difficult problems in this project was how to increase the quantum efficiency of spectral hole burning in an organic dye-doped polymer material. So far, all attempts to achieve single-shot recording in femtosecond regime have failed, basically, for this very reason. Our approach was to improve sample preparation and to look for new photochemical- and photophysical effects, which would allow to improve the recording efficiency.

We have developed new ways for chemical sample preparation for hole burning experiments. We have learned how to work with a variety of dyes and polymers to prepare thin polymer films with good optical quality, low scattering and with variable chromophore concentration. We have learned to prepare samples pressed between glass plates for improved stability and better optical quality. We have found a new class of photochromic materials, where the spectral hole burning is possible instead of usual stable photo-tautomer form, in the unstable photo-tautomer form. The advantage of the unstable tautomer is that we can now avoid spectral overlap between the initial and final (after phototransformation) forms of the molecule, which usually poses a serious difficulty for broad-band holographic recording. The reason why we can achieve this in this new system is in a curious fact that the oscillator strength of the unstable form is at least an order of magnitude larger than that of the stable form. Therefore, the low oscillator strength form absorbs only weakly, and practically does not appear in the frequency response of the medium at all. This

molecule is free-base Br-tetraethylporphyrin (BrTEP). We have performed detailed spectroscopic studies of this new material. We have measured Debye-Waller factor $\alpha > 0.7$ at 4K, burning quantum efficiency $\varphi > 10^{-3}$ and thermal stability up to 100K. We have observed for the first time that narrow-band illumination in the educt band leads to formation of vibronic holes in the photoproduct band. This is an unusual observation, which may be perhaps be used for crating absorption bands with special spectral profiles. The results of the spectroscopic studies were reported at the International Meeting on Spectral Hole Burning and Related Spectroscopies (HBRS'99), which took place in France in September 1999. A similar effect in a phthalonaphthalocyanie dye was used in out successful demonstration of single-shot recording of SHB hologram.

As a by-product of this work, we have prepared a new compound, which consists of strongly aggregated cyanide dye molecules generally known as J-aggregates. The special property of this compound is that it has resonance absorption peak in far-red wavelength around 810 nm. J-aggregates have been investigated earlier as potential materials for nonlinear optical- and electro-optical materials. All demonstrations so far have been at shorter wavelengths around 600nm, where no efficient diode lasers operate. To our knowledge our new material is the first demonstration of a J-aggregate system at a wavelength accessible with diode lasers and Ti:sapphire laser. A paper about this new material was published in *Chem. Phys. Lett.*

The second new result that was obtained in the last year is about novel interference effects in ultrafast photon echo.

Further, we have performed an experiment, where we are using interference of femtosecond laser pulses with two-pulse photon echo in dye-doped polymer films at low temperatures to demonstrate an ultrafast three-port all-optical coherent logic gate. The gate performs an operation equivalent of "controlled controlled NOT" on the time scale of less than 1 ps. We showed that an important condition for this gate is the causal nature of the coherent transient response, because only causality allows to implement the relevant truth table. The other special property of the gate is its coherent phase memory. On the basis of our results, we suggest that if the excited electronic state coherence time is much longer that the duration of the pulses, then it may be possible to implement a coherent logic gate with a large number of inputs, something similar to logic operations considered in quantum computing. A paper describing our results was published in *Laser Physics*.

2.3.2. Single-shot hologram recording experiment

In fall of 1999, we have been able to find a suitable material. This material consists of a polyethylene polymer film activated with antrhraceno-phthalocyanine (AnPc) molecules (see Figure below). SHB mechanism in this material consists in photo-tautomerization by switching the position of two protons in the center of tetratpyrrolic ring structure. As such, this material was known before, however, because the SHB efficiency in the stable tautomer (solid curve) was low (0.1 %), this material didn't work out for single-shot exposure recording. The crucial improvement came when we noticed that in the unstable photo-tautomer (dashed curve) form this molecule has a much higher SHB efficiency of about 10%. Therefore, we decided to try to "photo-sensitize" the material by switching the AnPc into its unstable photo-tautomer form first. This was achieved by cooling the polymer film down to liquid helium temperature, and then illuminating the material for about 1 min. with broadband light at the wavelength 780, which coincides with the resonant absorption of stable form of AnPc. As a result, a new absorption band appeared at a shorter wavelength 764 nm, corresponding to inhomogeneously broadened zero-phonon transition of the unstable photo-tautomer form. As the next step, we successfully

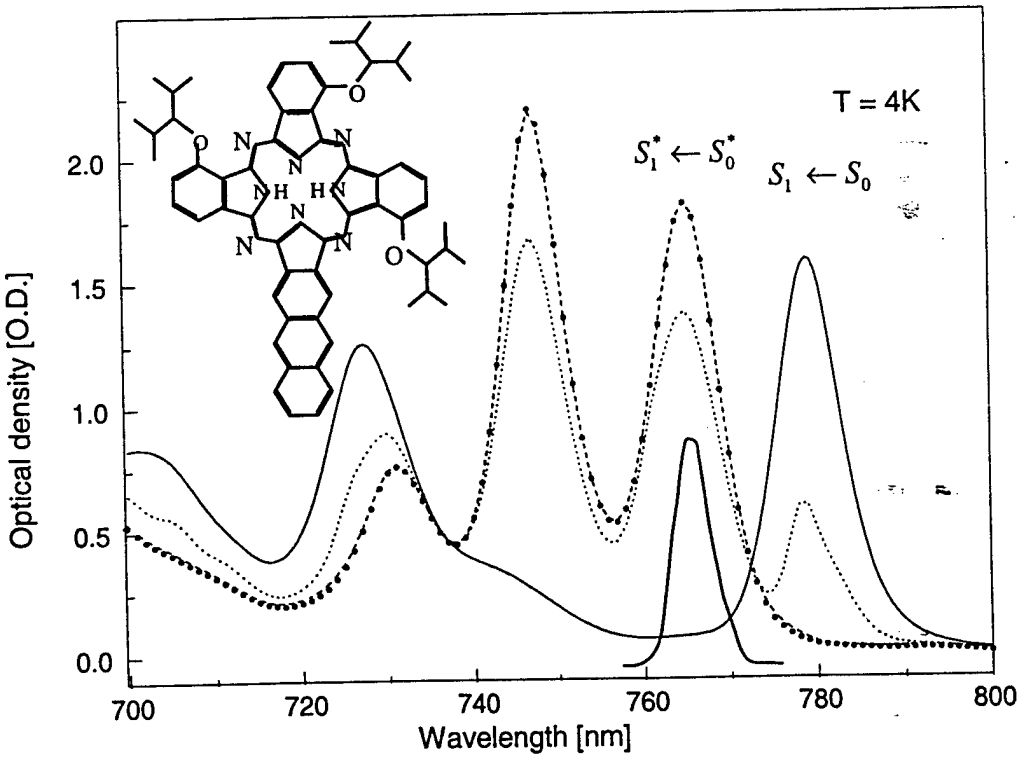

Fig.1. Absorption spectrum of the sample at low temperature (T= 4 K); Solid curve – stable tautomer form before illumination; Dashed-dotted curve – unstable tautomer form produced by illumination with spectrally broad light at 780nm; Dotted curve - after exposure to 250 pulses with 150 μJ cm^{-2} intensity at 764 nm wavelength; Solid curve shows the power spectrum of the pulses.

performed recording of single-shot holograms at this shorter wavelength, where the efficiency was more than one order of magnitude higher. These new results were published in *Optics Letters* and also reported at several international meetings (CLEO, Physics of Quantum Electronics, etc.).

A report on single-shot recording of ultrafast time-space holograms was given at AFOSR Contactors Meeting at SRI International in Menlo Park, CA, May 25-26, 2000.

In this way, the principal objective of this project has been achieved. There are, of course, many ways to further improve the experiment. One thing to consider in the future is improving our understanding how this material actually works. In particular, we have already performed some detailed spectroscopic studies of this new compound under femtosecond illumination, and measured important parameters such as hole recording quantum efficiency, Debye-Waller factor, temperature stability and oscillator strength. It turned out that it is important to avoid spectral overlap between the two tautomer forms. This depends both on the value of the intrinsic spectral shift between the two forms, as well as on the inhomogeneous bandwidth of the transitions. To avoid spectral overlap, the spectral shift has to be at least twice the inhomogeneous bandwidth. Earlier we used polyvinylbutyral polymer, which has a relatively large inhomogeneous width, and where two bands strongly overlap After discovering that this overlap is a serious problem, we switched to different kind of polymer polyethylene, which ha s a two times smaller inhomogeneous width. With this type of samples our experiments were successful. There are, however, several drawbacks in using polyethylene. First, this polymer doesn't dissolve practically in any solvents, which makes sample preparation extremely difficult. Secondly, polyethylene inherently contains tiny micro crystals, which scatter light and making hologram detection more difficult. To reduce the scattering, the sample needs to be hot pressed to a very thin film (< 0.1 mm). This, in turn, makes sample mechanically fragile and prone to cracking, especially after a few cooling cycles between room temperature and liquid helium. These problems need to be addressed in the future.

2.3.3. Photon Budget for Single-Shot Ultrafast Recording

An important technical information, which came out of this project, and which was not published separately in literature, consists in summarizing practical requirements for recording of SHB holograms in the femtosecond regime. Table 1. below presents numerical values for a range of

key experimental parameters. With this information in hand, one can evaluate practicality of future experiments. In particular, it turns out that requirement for energy density per one illuminating pulse is quite high – over 100 μJ per cm^2. Furthermore, at least in the present configuration, the sample cannot dissipate more heat than about 10mW per cm2, without causing the sample temperature to increase well over 4K.

Table 1.

Experimental parameters for optimal single pulse recording	
Pulse duration	150 fs
Wavelength	765 nm
Energy per photon	$2.6\ 10^{-19}$ J
Illuminated area	1 cm^2
Average pulse energy density	150 μJ cm^{-2}
Photons per pulse	$6\ 10^{14}$ photons/cm^2
Maximum average power	10 mW/cm^2 (limited by dissipation of heat)
SHB sample parameters	
Dye concentration	10^{-3} mol/liter
Molecules per volume cm^{-3}	$6\ 10^{17}$ cm^{-3}
Film thickness	0.1 mm
Molecules per area cm^{-2}	$6\ 10^{15}$ cm^{-2}
Transition oscillator strength	0.14
Probability of excitation	50-100%

Further projects should concentrate on detailed understanding of how spectral hole burning process works with intense ultrashort pulses. A useful approach will be here combining experimental studies with computer modeling of inhomogeneously broadened two-level medium interacting with ultrashort pulses of arbitrary intensity. Some of this kind of work is underway under different current projects. An important improvement could be achieved, if the transition dipole moments of all molecules could be aligned in one direction.

2.3.4. Spectral Hole Burning by Simultaneous Absorption of Two Photons

Most recently we have started a new direction of experiments on investigation of ultrafast recording by simultaneous absorption of two photons. The principal advantage of using two-photon resonance instead of conventional one-photon resonance consists in a possibility of true 3D volume recording and also in a possibility of recording high-order correlation proeporties of an optical field. In our recently published experiments we showed, for the first time, persistent

spectral hole burning by simultaneous absorption of two 1138-nm photons in an inhomogeneously broadened $S_1 \leftarrow S_0$ (Q_y) transition of unstable photo-tautomer of chlorin in polymer film at low temperature. Spectrally-selective hole burning is achieved due to high (10%) quantum efficiency of photo-transformation in that system and high peak intensity (tens of GW/cm^2) of near-infrared femtosecond pulses used for excitation. Two-photon absorption cross-section is measured for both stable and unstable photo-tautomers to be $\sigma_2 = (0.5 \pm 0.2) \times 10^{-50}$ cm^4 s $photon^{-1}$. This work is continued in the next grant entitled "Two-Photon Coherence and Ultrafast Optical Storage in Organic Molecules".

2.3.5. Patents and Commercialization Activities

We have started a new collaboration with Dr. Charlie Spangler from Montana State University Chemistry Department on application of organic chromospheres with large two-photon absorption cross-section. The principal idea is to use these materials for detection of mode-locked infra-red laser pulses, and potentially also as a memory material for optical storage. We have filed a patent "Mode-Locked Laser Infrared Detection Card and Method" (US Patent application 09/834,727). This technology is a licensed to a laser materials company in Bozeman, Scientific Materials Co.

2.3.6.Conclusion

We have accomplished the principal goal of this project: we have performed an experiment which shows that capturing a holographic image of a unique ultrafast event on subpicosecond time scale in a single recording exposure is indeed practically possible. This was achieved, on one hand, by investigating spectral hole burning properties of a range of organic materials, and on the other hand, by carefully optimizing the experimental conditions. After several unsuccessful attempts, finally success was achieved by modifying a previously known spectral hole burning materials in way, which increased the recording efficacy by more than one order of magnitude. This allowed us to record image holograms with 150-fs duration pulses without need to accumulate the SHB effect from many exposures. Results of this research show that it is possible to perform optical recording of data in frequency-domain on ultrafast time scale. These results can be used also as a new diagnostic tool for femtosecond dynamics in various ultrafast optical interactions.

3. Publications

3.1. Articles in refereed journals

1. "Terahertz, bit-rate parallel multiplication by photon echo in low-temperature dye-doped polymer film," O.Ollikainen, C.Nilsson, A.Rebane, Optics Commun. 147 (4/6), 429 (1998).
2. "Ultrafast time-and-space-domain holography in dye-doped polymers," A.Rebane, Chimia 52, 112-117 (1998).
3. "Nondestructive read-out of two-color photon-gated spectral hole burning holograms," D.Reiss, A.Rebane, U.P.Wild, Mol.Cryst. Liq. Cryst, v.314, 161-166 (1998).
4. "Demonstration of ultrafast logic gate by interference of coherent transients," W.Ross, M.Drobizhev, C.Sigel, A.Rebane, Laser Physics, v.6, n.5, p.1102-1108 (1999).
5. "Ultrafast processing with photon echoes," A.Rebane, M.Drobizhev, C.Sigel, W.Ross, J.Gallus, J. of Luminescence, 83-84, 325-333 (1999).
6. "Photo-tautomer of Br-porphyrin: a new frequency-selective material for ultrafast time-space holographic storage," M.Drobizhev, C.Sigel and A.Rebane, J. of Luminescence 86, 391-397 (2000).
7. "Interference between Femtosecond Pulses Observed via Time-Resolved Spontaneous Fluorescence," A.Rebane, M.Drobizhev and C.Sigel, Phys.Chem. Lett. 322, 287-292 (2000).
8. **"Single femtosecond exposure recording of image hologram by spectral hole burning in unstable tautomer of phthalocyanine derivative," A.Rebane, M.Drobizhev, and C.Sigel, Opt. Lett. 25, 1633-1635 (2000).**
9. "Persistent spectral hole burning by simultaneous two-photon absorption ," M. Drobizhev, A. Karotki and A. Rebane, Chem. Phys. Lett 334, 76-82 (2001).

3.2. Conference proceedings and presentations

1. A.Rebane, "Organic frequency-selective materials for ultrafast optical storage and processing," SPIE Proceedings, v.3468, 270-278 (1998).
2. A.Rebane, "Organic materials for ultra-fast holographic storage and processing," Optical Science and Laser Technology Conference, Bozeman, August 17-18, 1998.
3. A.Rebane and D.Reiss, "Nondestructive and transient read-out of photon-gated hole-burning holograms," CLEO, San Francisco, May 1- 4 1998.
4. O.Ollikainen, J.Gallus, U.Wild, A.Rebane, "New coherent optical technique for single-shot homogeneous spectrum measurement," CLEO, San Francisco, May 1- 4 1998.
5. O.Ollikainen, J.Gallus, C.Nilsson, D.Erni, A.Rebane, "Terahertz bit-rate optical processing by femtosecond two-pulse photon echo," CLEO, San Francisco, May 1- 4 1998.

6. Workshop on Spectral Hole Burning, Bozeman, March 8-11, 1998.

7. (INVITED) A.Rebane, SPIE 43rd Annual Meeting, Conference on Advanced Optical Memories and Interfaces to Computer Storage, 19-24 July 1998, San Diego.

8. (INVITED) A.Rebane, Topical Meeting of the International Commission for Optics "Optics for Information Infrastructure", Tianjin, China, 3-6 August 1998;

9. Workshop on Spectral Hole Burning, Bozeman, March 1999.

10. (INVITED) A.Rebane, 12th International Conference on Dynamical Processes in Excited States of Solids, Humacao, Puerto Rico, May 23 - 27, 1999.

11. A.Rebane, "Efficient frequency- selective materials for ultrafast time- and frequency domain data storage and processing", 6th International Meeting on Hole Burning and Related Spectroscopies: Science and Applications, Hourtin, France, September 18-23, 1999.

12. A.Rebane, A.Tchouassi-Djiki, R.Babbitt, "Recording of ultrafast image holograms by photo-induced frequency-doubling in glass", IEEE Laser and Electro-Optics Society (LEOS) 12th Annual Meeting, 8-11 November 1999.

13. A.Rebane, "Femtosecond holography and pulse interactions in inhomogeneously broadened media," 30th Winter Colloquium on the Physics of Quantum Electronics, Snowbird, Utah, 9-12 January 2000.

14. A.Rebane, M.Drobizhev and Ch. Sigel, "Single-shot recording of ultrafast time-space holograms," CLEO 2000, San Francisco.

15. A.Rebane, M.Drobizhev and Ch. Sigel, "Photon-gated holographic hole burning and read-out in Si-naphthalocyanine-doped polymer film," M.Drobizhev, C.Sigel, and A.Rebane, CLEO 2000 Proceedings, p.488 (2000).

16. A.Rebane, "Interference between femtosecond pulses observed via time-resolved spontaneous fluorescence," Workshop on Applications of Spectral Hole Burning, Big Sky, July 9 -12, 2000.

17. A.Rebane, "Femtosecond holography and pulse interactions in inhomogeneously broadened media," 30th Winter Colloquium on the Physics of Quantum Electronics, Snowbird, Utah, 9-12 January 2000;

18. A.Karotki, M.Drobizhev, A.Rebane, "Femtosecond frequency-domain storage by two-photon absorption," CLEO 2001 Proceedings (2001).