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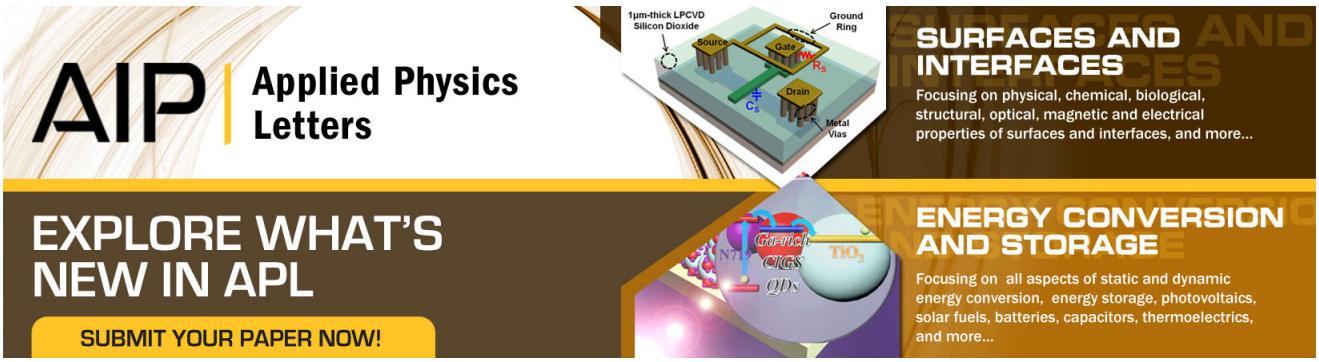

Exciton mediated self-organization in glass driven by ultrashort light pulses

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We propose an exciton-polariton-mediated self-organization effect in transparent SiO_2 glass under intense femtosecond light irradiation. Interference and dipole-dipole interaction of polaritons causes formation of gratings of dielectric polarization. Due to an ultrafast exciton self-localization into a quasicrystal structure, the polariton gratings remain frozen in glass and a permanent three-dimensional image of exciton-polariton gas is created. We show that coherent effects in propagation of exciton-polaritons can serve as a tool for nanostructuring and fabrication of 5-dimensional optical memories in glass, opening new horizons for polaritronics. © *2012 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4742899]

Progress in high power ultrashort pulse lasers has opened new frontiers in physics of light-matter interaction ranging from microsurgery to non-reciprocal writing.[1–3] One of the unsolved puzzles in this field is the observation of self-assembled periodic structures in variety of optical materials subjected to intense laser irradiation.[4] A decade ago, highly ordered subwavelength structures with features smaller than 20 nm were discovered in the ultrafast laser irradiated volume of SiO_2 glass,[5] which were found to be responsible[6] for the induced optical anisotropy reported earlier.[7,8] Despite several attempts to explain the peculiar self-organization process, the formation of these nanostructures still remains debatable.[5,9] The effect of nanograting formation has already found numerous applications for polarization control devices[10–12] and microfluidics.[13] More recently, the subwavelength nanostructures were also proposed for rewritable polarization multiplexed optical memory,[14] where the information encoding would be realized by means of two birefringence parameters, i.e. the slow axis orientation (4th dimension) and strength of retardance (5th dimension), in addition to three spatial coordinates. However, the implementation of the independent manipulation in 4th and 5th dimensions has not been demonstrated. In this letter, we demonstrate the crucial role of coherent light-exciton waves in ultrafast laser writing. We explain the observed self-assembling process by the interference of coherent exciton-polariton modes induced in a wide band-gap amorphous material and demonstrate harnessing of this effect for independent multidimensional access for optical data recording.

In our experiments, we used two femtosecond pulse sources. The first laser source was a fibre laser system (IMRA America FCPA μJewel D-400) with a variable repetition rate from 200 kHz to 1 MHz, pulse duration of 400 fs 1045 nm. The implementation of 5-dimensional memory was performed on a regeneratively amplified mode-locked system based on a Yb:KGW crystal operating at 1030 nm and emitting pulses of 300 fs at 200 kHz repetition rate. Pulse energy during the experiment was set to 1.3 μJ.

The irradiated sample was lapped with aluminium oxide and then polished with Syton. Sample was imaged with backscattering scanning electron microscope without subsequent chemical etching, which is normally used imaging such structures. The images revealed periodic nanostructures due to the material density variation created during laser irradiation (Fig. 1). An evident abrupt change of the density suggests that the mechanism leading to the periodic nanostructuring is highly nonlinear. Backscattered electron analysis reveals that the nanogratings consist of a periodic distribution of oxygen-deficient regions (SiO_{2-x}).[5] The main observed features of the nanogratings: (i) the period in the propagation direction increases with the distance from the front of the laser induced structure, starting from a value close to the laser radiation wavelength; (ii) the transverse grating appears in the direction of the polarization vector and has a period less than the wavelength of light (Fig. 1).

The earlier attempts to explain the ultrafast laser induced nanogratings inside transparent materials were made by two different models based on a laser generated electron plasma.[5,9] One, adopted from the conventional theory of laser induced surface ripples formation, relies on the interference of bulk electron plasma waves with the incident light,[5] while the other attempts to explain the subwavelength structure formation by nanoplasmas.[9] However, none of these mechanisms can explain all peculiar features of the ultrafast laser induced nanogratings. We note that in the conditions of our experiments the silica glass remains below the critical

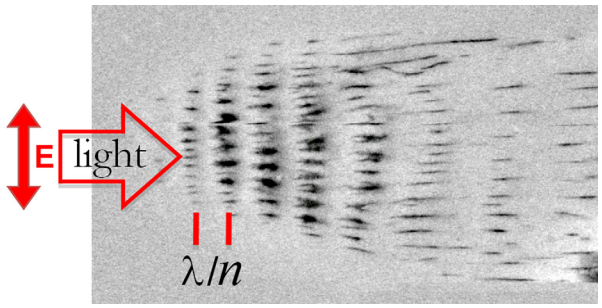

FIG. 1. Self-assembled nanograting induced by femtosecond laser irradiation ($\lambda = 1045$ nm) inside fused silica. SEM image with backscattered electrons is taken of not chemically etched sample. Two periodicities (along the light propagation and electric field directions) can be clearly seen. The arrows indicate light propagation and electric field **E** polarization directions, n = 1.45 is the refractive index of fused silica.

plasma concentration,[16] so that one can hardly expect significant plasmon features. The plasma frequency cannot be defined as it depends on the concentration of free carriers, which is nearly zero in the absence of light and strongly varies as a function of time when the sample is illuminated with a light pulse. It is also unclear how plasmons may be responsible for the formation of a grating in the propagation direction.

We rule out the plasmon scenario, and show here that the nanograting is formed due to interference, attractive interaction and self-trapping of exciton-polaritons.[17] Exciton-polaritons are mixed light-matter quasiparticles responsible for fascinating non-linear optical effects in semiconductors including polariton lasing, Bose-Einstein condensation and are promising for applications in so-called *polaritronics*.[18] Exciton-polaritons in amorphous dielectrics have not been studied experimentally until now, while, theoretically, nothing prevents their formation and propagation in glasses. Propagation of exciton-polaritons can be understood as a chain of virtual absorption-emission acts, and it does not imply motion of individual excitons, in general. This is why exciton-polaritons may spread even in strongly localised systems, e.g., arrays of quantum dots.

It is well-known that the absorption spectrum of SiO_2 is characterised by a strong exciton peak at about $\hbar\omega_0 = 10.4\,\text{eV}$.[19,20] The binding energy and oscillator strength of excitons in SiO_2 are expected to exceed by orders of magnitude those in GaAs, where excitonic effects are studied in detail. In striking similarity to our observation of nanostructures in fused silica, formation of a polarization grating in the direction of propagation of light due to interference of exciton-polariton modes in GaAs was predicted more than 11 years ago.[21] The period of this grating was found to gradually increase as a function of the distance from the front edge of the sample due to the dependence of the splitting between two interfering exciton-polariton modes on the group velocity of the exciton-polaritons. In our case, the two dispersion branches of exciton-polaritons (Fig. 2(a)) may be excited simultaneously by multiphoton absorption. The interference of propagating exciton-polaritons results in formation of the polarization grating shown in Fig. 2(b).

Within the polariton model, we simulated the nanograting using a linear semiclassical approach. The dielectric polarization in the direction of propagation is calculated as

$$\mathbf{P}_{exc}(z,t) = \frac{1}{2\pi}\int_{-\infty}^{\infty} dz' g(z-z') \int_{-\infty}^{\infty} d\omega[\chi_e(\omega)\mathbf{E}(z',\omega)e^{-i\omega t}], \tag{1}$$

where $\mathbf{E}(z, \omega)$ are solutions of Maxwell's equations in the frequency domain, $g(z) = I^N(z)$ where $I(z)$ is the spatial shape of the incident pulse, N is a number of photons participating in the multiphoton absorption process (typically, 7 Ti:Sapphire photons are needed to excite one exciton in silica glass). The dielectric susceptibility is given by,

$$\chi_e = \frac{\varepsilon_b \omega_{LT}}{\omega_0 - \omega - i\gamma}, \tag{2}$$

ω_0 and ω_{LT} denote the exciton resonance frequency and the exciton longitudinal-transverse splitting respectively, γ is the exciton non-radiative broadening, and ε_b is the background dielectric constant. The scattering states can be found using the transfer matrix method, however in our case we are dealing with a single layer (i.e., a bulk structure), in which case the scattering state reduces to a plane wave $E(z,\omega) = e^{ik(\omega)z}$.

We underline that the inevitable exciton inhomogeneous broadening does not qualitatively change the effect of grating formation.[21] The splitting of exciton-polariton modes in SiO_2 is more than 100 meV, which exceeds the inhomogeneous broadening of the free exciton resonance. Also note that the first period of the polariton grating shown in Fig. 2(b) is very close to the wavelength of light. Then the period increases as a square root of the distance from the sample edge, approximately. These two features are in excellent agreement with the experiment which confirms the validity of the model.

To obtain the distribution of the exciton density in the direction of light polarization, we resolve a one-dimensional Gross-Pitaevskii equation describing propagation and attractive dipole-dipole interaction between coherent exciton-polaritons,

$$H = -\frac{\hbar^2}{2m}\frac{\partial^2}{\partial x^2} + \alpha|\psi|^2, \tag{3}$$

where m is the mass of an exciton and α is the negative (attraction) exciton-exciton interaction constant. The equation is solved variationally using the following trial function:

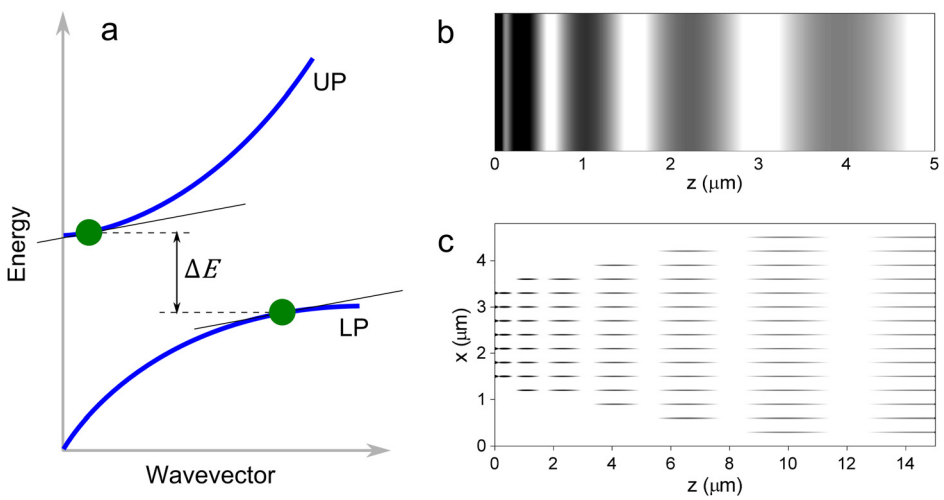

FIG. 2. Theoretical simulation of the formation of the nanograting. (a) A schematic of the exciton-polariton dispersion, showing a point on the upper polariton branch (UP) and a point on the lower polariton branch (LP) with the same group velocity, and their splitting in energy ΔE (not to scale). (c) shows the grating in x and in z, while (b) shows a zoom of the grating in the z-direction. In (c) the exciton mean free path is taken as $d = 300\,\text{nm}$ and the exciton mass equal to the free electron mass. Time, $t = 300$ fs relative to the arrival of the light pulse at $z = 0$. $\hbar\omega_{LT} = 0.5\,\text{meV}$, $\hbar\gamma = 1\,\text{meV}$, $n_e = 10^5\,\text{cm}^{-1}$, and $\varepsilon_b = 3$.

$$\psi = \sqrt{\frac{N}{a}}\left(\frac{2}{\pi}\right)^{\frac{1}{4}} e^{-x^2/a^2}, \quad (4)$$

where $N = nd$ is the number of excitons, d is the exciton mean free path and a defines the width of the Gaussian. We then may find the energy and minimise with respect to a using only two fitting parameters: interaction constant α and exciton free mean path d. The best fits were obtained with $\alpha = 2.25 \times 10^{-7}$ meV·cm and $d = 300$ nm. The calculation with only two fitting parameters (the interaction constant and exciton mean free path in the polarization direction) yields an excellent qualitative agreement with the experiment. We estimate that critical concentration for periodic exciton-polariton grating formation is about 10^{15} cm^{-3}. If exciton-polariton concentration is below this value the grating is not formed and induced modification does not exhibit anisotropy. This could explain phase transition from isotropic refractive index increase to birefringent modification reported by several groups.[22,23]

Self-organization of exciton-polaritons is qualitatively similar to the exciton pattern formation effect observed in cold exciton gases:[24] in both cases the instability in the exciton (exciton-polariton) gas appears due to dipole-dipole interactions between its constituents. The difference is in the sign of interaction, which is repulsive in Ref. 24 and attractive in our case. We note that exciton-polaritons form a dynamical grating which evolves in time. However, the lifetime of exciton-polaritons is limited to a fraction of a picosecond by their phonon assisted relaxation to indirect exciton states decoupled from light.[25] These indirect excitons are essentially immobile. They are easily self-trapped and live for several microseconds or even longer.[17] Excitons generated by subsequently arriving pulses of light are accumulated in significant concentrations in the specific sites of the sample corresponding to the peaks of the exciton-polariton density in the polariton grating that is generated in the same fashion by all pulses. Recombination of self-trapped excitons is accompanied by generation of molecular oxygen[26,27] due to the photosynthesis-like reaction,

$$SiO_2 + X \rightarrow Si + O_2, \quad (5)$$

where X denotes an exciton. Nanopores of silica filled by oxygen[27] are formed in the locations of high concentrations of self-trapped excitons. Formation of pores in fused silica was recently confirmed by another group investigating volume expansion induced by femtosecond laser irradiation.[28] Agglomerations of these nanopores form the grating seen in the SEM images (Fig. 1). They are responsible for the freezing of the polariton-induced polarisation grating in silica glasses.

The described nanogratings are responsible for strong birefringence, which can be described by two independent parameters: retardance and slow axis direction. The ability of independent control of these two parameters with exposure and polarization of irradiation was demonstrated by simultaneously recording of two data sets represented by portraits of I. Newton and J. C. Maxwell (Fig. 3) inside fused quartz. The 4th dimension of optical data storage, which is as an orientation of slow axis, could be recorded with a resolution of several degrees in the range from 0 to 180°. The best results were achieved when the experimental conditions were chosen to avoid retardance saturation over a certain number of pulses. As a result we were able to expand the retardance range over 100 nm and determine eight discrete levels, which were used to record data in the 5th dimension. The reading of the polarization information was implemented with a quantitative birefringence measurement system based on a liquid crystal polarization compensator and a conventional optical microscope. Both dimensions were read-out with a high degree of distinction and negligible cross-talk. The demonstrated method of information encoding by means of birefringence can be implemented not only in optical storage, but also as a counterfeiting marking technology. As a result we are able to validate the possibility of producing optical memory with five independent parameters by using monochromatic illumination for recording and reading—as opposed to the previously suggested plasmonic multidimensional optical storage.[15]

In conclusion, we show that coherent polariton fluid, exited by multiphoton absorption, can play an important role in a wide bandgap amorphous material. The coherence time of polaritons in the glass is extremely short, less than a fraction of picoseconds. However, an ultrafast exciton self-trapping mechanism imprints the permanent image of a polariton grating. The effect gives a deep insight into the exciton-polariton dynamics during the first picoseconds of their life. The rapid transformation of free exciton-polaritons to immobile self-trapped indirect excitons freezes the exciton-polariton polarization grating in glass. Recombination of the self-trapped excitons induces the synthesis of molecular oxygen and

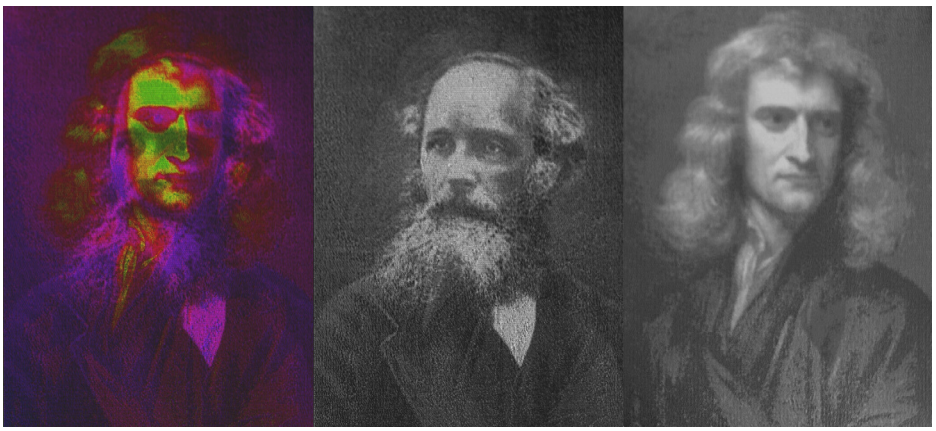

FIG. 3. Ultrafast optical recording via self-assembled nanograting induced birefringence in fused silica. Maxwell and Newton are recorded in one image (left, in pseudo colors), however, they can be easily decoupled as Maxwell is recorded in strength of retardance (center) and Newton in azimuth of the slow axis (right). Size is 1.5 × 2 mm.

creates nanopores in silica, which modify the refractive index and induce birefringence in the glass. In addition, we demonstrated five-dimensional optical memory based on the effect of polaritonic self-assembly. The reported effect of polaritonic self-assembly will open new opportunities for material structuring and control at subwavelength scales and shows the high potentiality of polaritronics.[18]

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