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All-optical gel memory

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We have studied the behavior of organic molecules (Rhodamine 640) that are encaged in a matrix synthesized by the sol-gel process. By applying a strong optical-polarized electric field, we can control the alignment of the molecules within the sample and locally create a birefringent effect. Using this sample in a Kerr experiment, we obtained a transmission ratio of 6%. Since the birefringent effect lasts at least several days, this type of sample could be used in an optical process for storage of information. Moreover, we show that such information can be deleted and rewritten repetitively.

This Letter describes a Kerr-effect experiment in a new nonlinear medium: a solid matrix synthesized by the sol-gel process and doped with an organic molecule. The solid sample that contains the organic dye molecules will be referred to as a doped xerogel.

Recently several authors[1-8] have reported optical properties of transparent silica and alumina porous media prepared at low temperature by the sol-gel technique and doped with various organic dyes. In this Letter, we describe oxide matrices that have been prepared from a modified sol-gel technique. The hydrolysis of metal alkoxides diluted with alcohol is carried out without catalyst or additive by using the moisture in the ambient atmosphere.[9] This method produces large pieces of dense gel doped with organic molecules that are within the silica–zirconia system. The following experimental procedure is used: Raw materials $Zr(OC_4H_9)_4$ and $Si(OC_2H_5)_4$ in the molar ratio 1:4 are mixed in a dry atmosphere with ethanol as the solvent and stirred for several minutes. The volume ratio of ethanol to alkoxides is kept at 1:1. The solution is doped with Rhodamine 640 perchlorate at a concentration of 2×10^{-4} mol/L. Complete gelation occurs within one week at room temperature. After aging for a month in the ambient atmosphere, optically clear, dense xerogels (density 1.7 g/cm^3) of glassy aspect are obtained, which do not require polishing prior to optical measurements. The volume of the gel samples corresponds to about 10% of the initial volume of the solution. No detectable leakage of dye is observed after immersing the samples in alcohol, which shows that the dye molecules are completely trapped in a surrounding rigid cage.

The average size of the pores is thought to be slightly greater than the steric hindrance of the organic molecule used (Rhodamine 640). As we have previously shown,[10,11] it can be presumed that movement of the organic dye molecules is impeded and

that their relaxation time should be longer than that within usual liquid solvents in which the viscosity governs such relaxation.

The xerogel sample was used as the active medium of a Kerr cell and was thus located between crossed polarizers. An optical-polarized femtosecond pump pulse was focused upon a point of the solid matrix to induce a birefringent effect. We read the transmission variation with a cw He–Ne laser focused on the same point with a polarization of 45° with respect to the pump polarization. The analyzer, orthogonal to the probe-beam polarization, prevented any light from being issued from the cell when the doped xerogel sample had not been exposed to the pump beam (Fig. 1). The transmission value T, which is a ratio of the output I and input I_0 energies ($T = I/I_0$), of such a Kerr cell is

$$ T = \sin^2\left(\frac{\pi e \Delta n}{\lambda}\right), $$
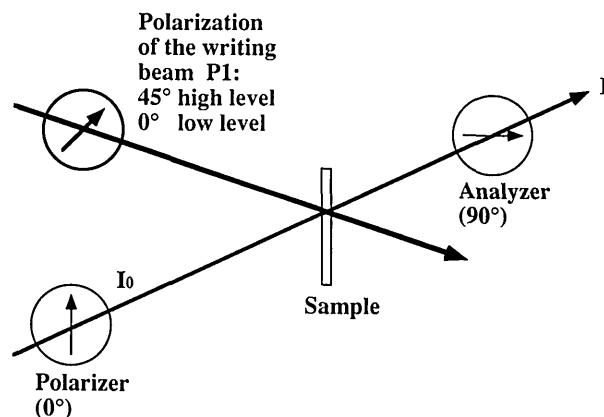
where e is the active medium thickness, λ is the

Fig. 1. Experimental setup. I_0, input energy; I, output energy. Sample has thickness e, birefringence $\Delta n = n_{ex} - n_{or}$.
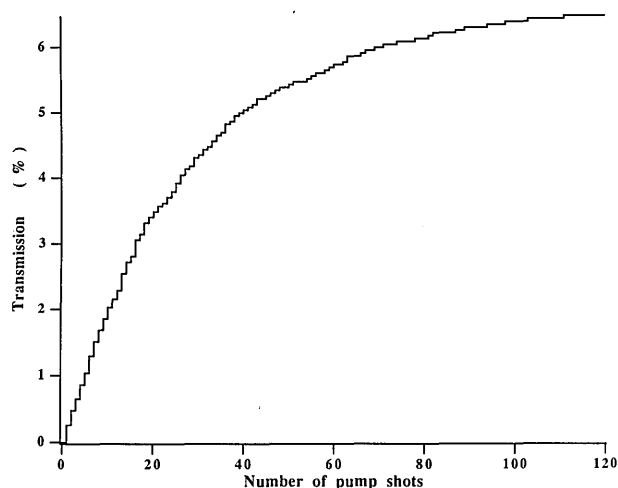
Fig. 2. Transmission coefficient of the Kerr cell as a function of the number of pump shots applied to the sample. Equilibrium is reached after a few hundred shots.

wavelength of the probe beam, and birefringence Δn is the difference between the indices of refraction for the two neutral axes of the birefringent medium. $\Delta n = n_{ex} - n_{or}$, where n_{ex} and n_{or} are the refraction indices of the extraordinary and ordinary axes. This maximum value is obtained with the crossed polarizers when the probe beam is at an angle of 45° to the two neutral axes. In an isotropic medium, $\Delta n = 0$, which leads to $T = 0$. Our medium is initially isotropic owing to a random distribution of the organic molecule orientations. The transmission of the cell is then $T_0 = 0$ and does not depend on the polarization of the probe beam or orientation of the xerogel matrix that we adopt.

The pump beam is produced by a colliding-pulse passively mode-locked dye laser operating at 620 nm and amplified by a four-stage dye amplifier that is pumped by a frequency-doubled Q-switched Nd:YAG laser.[12] . We use 100-fs pulses with an energy of approximately 1 μJ per pulse, which gives a peak power of 10 MW. The repetition rate is 50 Hz. The probe beam is generated with a cw He–Ne laser of 4-mW intensity. The beams are focused to a diameter of approximately 50 μm.

After a single shot of the pump beam, the transmission value is no longer equal to zero; the point of the Kerr cell is now active. This single-shot-induced transmission, T_1, is approximately 0.25% in our experimental conditions.

If the pump beam is again applied upon the same point of the sample, the transmission value keeps rising until it reaches a maximum value T_∞, after which subsequent shots from the pump beam do not significantly affect the transmission coefficient. Equilibrium is reached after a few hundred shots (Fig. 2). The value of T_∞ is approximately 6%.

The induced transmission value is, to a large extent, permanent. It decreases slightly during the first few hours but remains at 70% of the maximum value after a couple of days (Fig. 3).

Having lit a point, we can now rotate the pump-beam polarization so that it becomes parallel or orthogonal to the probe polarization. After a number

of pump-beam shots comparable with that used to light up the point, the point can be turned off again. The transmission value decreases from T_∞ to a transmission T_0' smaller than 0.5%.

If we decrease the pump-pulse energy to 0.25 μJ, no effect is observed even after millions of shots. Notice that the optimal energy (1 μJ) used to induce the birefringent effect is one order of magnitude lower than the threshold energy for nonlinear effects such as self-phase modulation or self-focusing in the xerogel.

Alternately rotating the pump polarization between 45° and directions parallel or perpendicular to the probe-beam polarization causes the transmission value to oscillate between T_∞ and T_0' (Fig. 4). We can thus command the state of the output: active or inactive, or high or low level in terms of logic electronics. This point can be rewritten indefinitely, and we have experimentally repeated more than ten cycles without noticing any changes in the behavior of the material. We should note here that the effect is nonresonant. Thus we see no reason for a degradation of the behavior with time.

As we have written a bit of information that does not need to be refreshed, we can address another point of the sample with the pump beam (Fig. 5). This can be repeated indefinitely, being limited only by the sample size. There is no interaction between
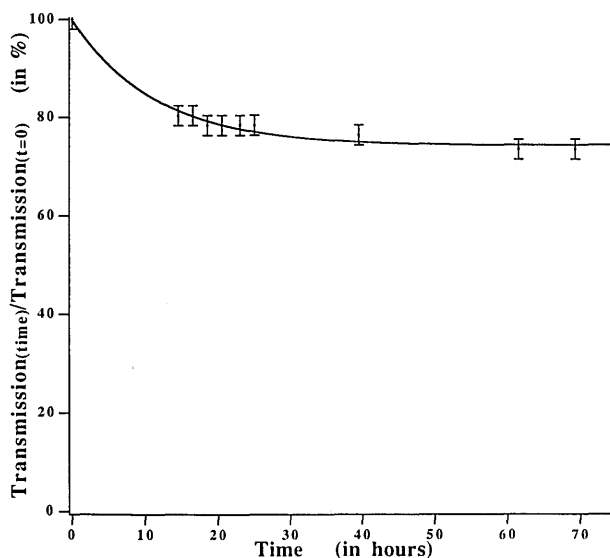

Fig. 3. Memorization effect. The induced transmission coefficient decreases slightly but remains at more that 70% of the initial value, even after a couple of days.
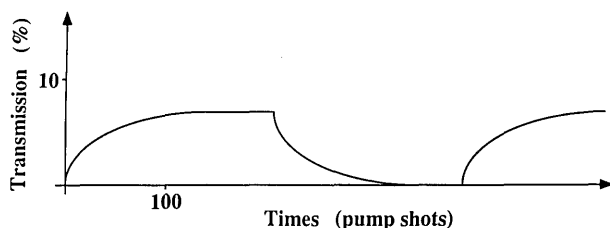

Fig. 4. Behavior of the transmission coefficient as the pump polarization is alternately rotated between 45° and 0° or 90° to the probe-beam polarization.
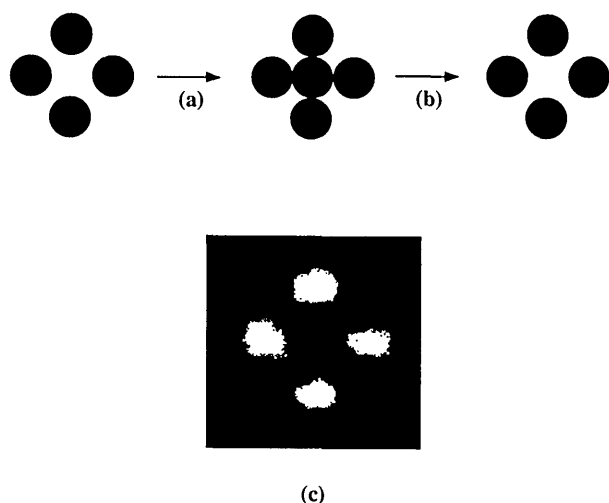
(c)

Fig. 5. Schematic representation of five points of our all-optical matrix. We can light up a point (a) or turn it off (b). Photo (c) gives an idea of our resolution; two opposite points are 200 μm apart.

one point and another; the resolution of our medium is supposed to be molecular. The doped xerogel can be of arbitrary size since no electrical connections are needed as with electro-optical devices. A 1000×1000 point matrix addressed with a 25-μm-diameter pump pulse would only be 2.5 cm $\times$ 2.5 cm. Such a matrix would represent a 10^6-bit memory. If the probe beam used is spatially large enough, this matrix can be read simultaneously, in parallel logic.

Physically, we interpret these results as indicating the possibility of commanding the alignment of the organic molecules in the xerogel matrix. The organic dopant molecules are sufficiently held by the host matrix as to be immobilized with respect to usual forces (e.g., thermal agitation), but under a powerful electric field they are compelled to rotate. The induced dipolar moments of the organic molecules align themselves in the direction of the electric field, which leads to local anisotropy of the medium that induces a local birefringence Δn. As our sample is 0.7 mm thick and we measure our transmission values at 632.8 nm, we estimate our induced birefringence Δn to be approximately 7×10^{-5}.

In the initial state at a local point of the doped xerogel, the organic molecules are isotropically dispersed in the solid medium, which leads to a transmission value $T_0 = 0$. The first inscription of the material leads to a preferential alignment of the molecular dipole in the direction of the electric field of the pump pulse and at 45° to the field of the probe. When the probe electric field encounters a birefringent medium, it is elliptically polarized as it comes out of the solid medium, and this leads to a transmission value of a few percent. The next command sequence, which brings the value back to zero, does not bring the sample back to its former state but only

rotates the neutral axes of the birefringent medium to align one of the axes with the probe beam polarization. Hence the probe beam polarization no longer encounters the medium as birefringent, and the overall transmission of the Kerr cell returns to zero. We have checked this behavior simply by rotating the crossed polarizer by 45°, which causes the transmission to return to its T_∞ value. Further command sequences cause the system to alternate between the latter two states T_∞ and T_0'. As we control the number of pump shots used and particularly the shot polarization, not necessarily at 0°, 45°, or 90°, but at any orientation between 0° and 90°, we can impose a continuous infinity of stable states in the doped xerogel matrix, which leads to any stable transmission between T_0' and T_∞.

To summarize, we demonstrate the possibility offered by the sol-gel process of encaging organic molecules with sufficient strength to prevent any change of orientation of these molecules unless a powerful optical electric field is applied to align their induced dipole moments. We can thus locally control the direction (0–2π) and value (0–7 $\times$ 10^{-5}) of the birefringence Δn. This leads us to propose a simple and cheap way of obtaining a multistable, addressable memory matrix of arbitrary size whose resolution is limited only by problems inherent to parallel treatment of information in optics.

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