Phase conjugation by four-wave mixing in a stratifying solution

S. A. Viznyuk, P. P. Pashinin, A. M. Prokhorov, S. F. Rastopov, and A. T. Sukhodol'skii

Institute of General Physics, Academy of Sciences of the USSR, Moscow

(Submitted 17 November 1989)
Pis'ma Zh. Eksp. Teor. Fiz. **51**, No. 2, 86–90 (25 January 1990)

Results are presented for the first experiments in phase conjugation with degenerate four-wave mixing of cw laser radiation in a stratifying solution. It is shown that solutions with a low critical stratification point may serve as the basis of fundamentally new recording media with an effective nonlinearity parameter $dn/dI \gtrsim 10^{-3}$ cm^2/W.

Of the mechanisms of four-wave mixing employed in phase conjugation with the use of cw lasers, the principal ones are thermal nonlinearities in organic liquids[1,2] and the nonlinearity resulting from the dependence of the optical properties of liquid crystals on the intensity of the incident radiation.[3] Investigations have also been carried out on phase conjugation with cw pumping in thin layers of solid solutions of dyes.[4] The main mechanism for the nonlinear interaction in this case is absorption saturation.

Previously, the effect of separation of the components of a stratifying solution in a nonuniform temperature field with a laser-induced phase transition[5] has been used for recording a diffraction grating in an aqueous solution of butyl cellosolve, a medium that has a low critical stratification point.[6] Modulating the refraction index of a binary solution by separation of its components is an interesting and fundamentally new mechanism of optical nonlinearity that can be used for phase conjugation and the dynamic recording of information.

In this investigation we have, for the first time, achieved phase conjugation in a stratifying solution. A time of ~ 1 h was obtained for the stability of the phase dynamic structure recorded in the medium. We compare the nonlinearity parameter dn/dT of the stratifying solution we studied with those of the principal nonlinear media that have been used so far.

A diagram of the experimental apparatus is shown in Fig. 1. The experiment was performed with the cw radiation of an LGN–404 argon laser 1, operating at the wavelength 0.5145 μm. The initial beam was split by a beam-splitting cube 2 into two beams of equal intensity. The pump beam I_p passing through mirror 3 was directed by mirror 4 into the layer of the stratifying solution 5 placed between two glass substrates 2.7 mm thick. The beam reflected by mirror 3 was directed by mirror 6 counter to I_p, forming the readout beam I_r. The beam passing through cube 2 was directed into the layer of the stratifying solution 5 at an angle $\varphi = 9.1 \times 10^{-3}$ rad to beam I_p and formed and the writing beam I_s. Mirror 7, which reflects 64% of the radiation, served to bring out the conjugated beam I_d, whose total intensity was measured by photodiode 8 and recorded by the recorder 9. The intensity distribution in the conjugated

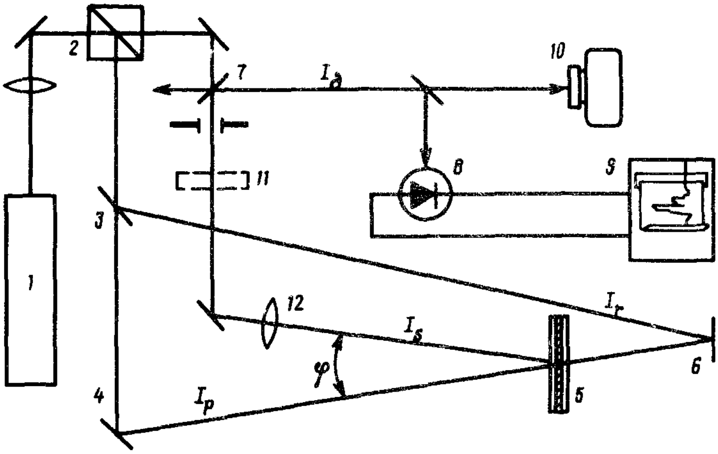

FIG. 1. Diagram of the apparatus.

beam was recorded by a photographic apparatus 10, which was located 2 m from mirror 7 in order to eliminate the scattered background.

The values of the power of I_p, I_s, and I_r were, respectively 60 mW, 60 mW, and 120 mW. The pump beam diameter in the mixing region was ≈ 1.6 mm (Fig. 2a). The intensities of I_p, of the write beam, I_s, and of the readout beam, I_r, in the mixing region were, respectively 3 W/cm^2, 3 W/cm^2, and 6 W/cm^2. From this we find the modulation amplitude of the intensity in the interference pattern from beams I_p and I_s: $I_g = 2\sqrt{I_p I_s} = 6$ W/cm^2. The period of the interference pattern for a nondistorted write beam was 56 μm.

The stratifying solution was a solution of butyl cellosolve in water in the proportion 2:3 by volume. The dye rhodamine G was introduced into the solution. The initial absorptivity of the solution was $\alpha = 3200 \pm 150$ cm^{-1}. Because of light-induced dissociation of the dye, the absorptivity decreased on the average to $\alpha = 2660$ cm^{-1}. The thickness of the layer of solution between the substrates was set at $h = 16 \pm 2$ μm.

To demonstrate phase conjugation in the write beam, we placed a distorting phase transparency 11 into it (a curved glass plate). The write beam, diverging after passing through the distorting plate, was focused by lens 12 so that the crossover after the focus was located between the lens and the mixing region.

Figure 2 shows photographs of the write beam after it has been reflected in the plane of the layer by an ordinary mirror and by the conjugating mirror (the stratifying solution). Figure 2a shows photographs of the intensity distributions of beams I_p and I_s, spread out at a certain distance in the plane of the layer 5. The rest of the pictures were taken by the photographic apparatus set up as shown in Fig. 1. All the pictures are on the same scale.

The results in Fig. 2 show that as a result of the reflection by the four-wave mixing in the stratifying solution and the restoration on passing back through the

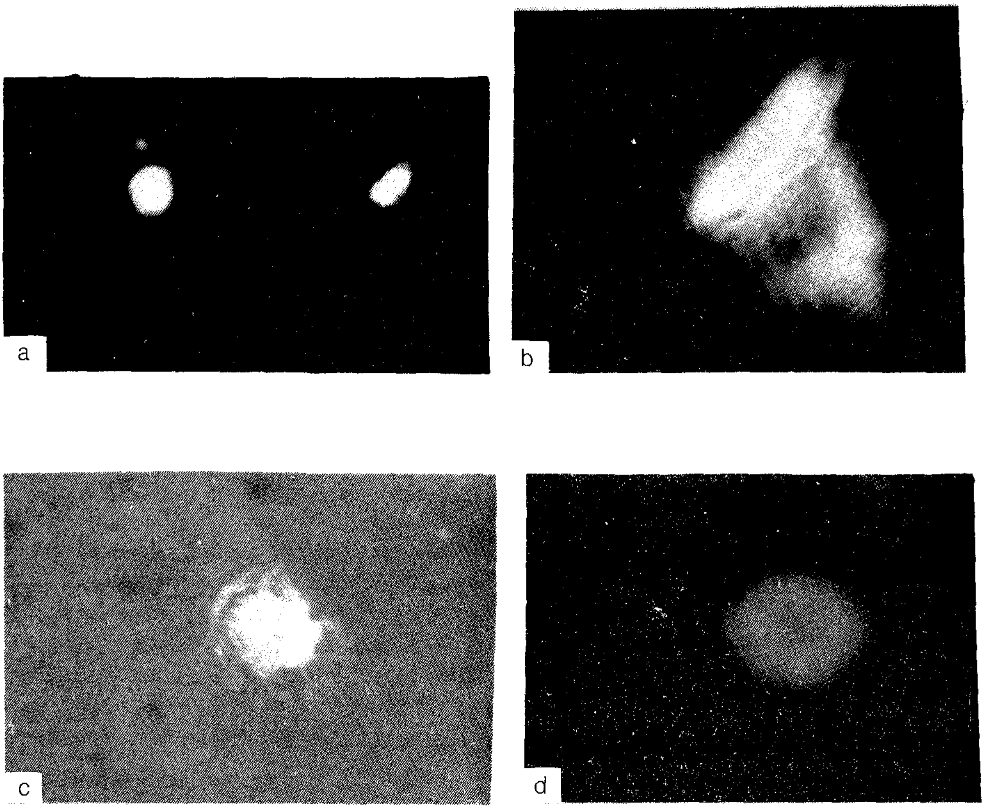

FIG. 2. a) Pump beam and writing beam (after distortion) in the mixing region; b) intensity distribution in the write beam after its reflection by an ordinary mirror located at the position of the stratifying solution and a second pass through the distorting plate; c) restored beam; d) restored beam when the write beam is not distorted.

distorting plate, the large-scale nonuniformities (divergences) of the initial write beam are canceled. Distortions of the restored beam are due to the finite spatial resolution of the heterogeneous spatial structure that arises in the region of the beam mixing. By taking out the dependence of the diffraction efficiency on the period of the grating written in the solution one can estimate the minimum resolution size as $\approx 10\,\mu$m (the minimum period of a diffraction pattern that can be recorded).

The time dependence of the power of the conjugated beam from the time of turning on the laser is shown in Fig. 3. It can be seen that the structure induced in the medium is relatively stable (it does not decay during at least 30 min of continuous irradiation). Variations in the intensity of the conjugated beam are due evidently to an instability in the pump laser and mechanical vibrations of the apparatus. The maximum power of a conjugated wave obtained in the experiment with the stated parameters of the apparatus was 0.26 mW.

Let us estimate the nonlinearity parameter dn/dI of the stratifying solution and of

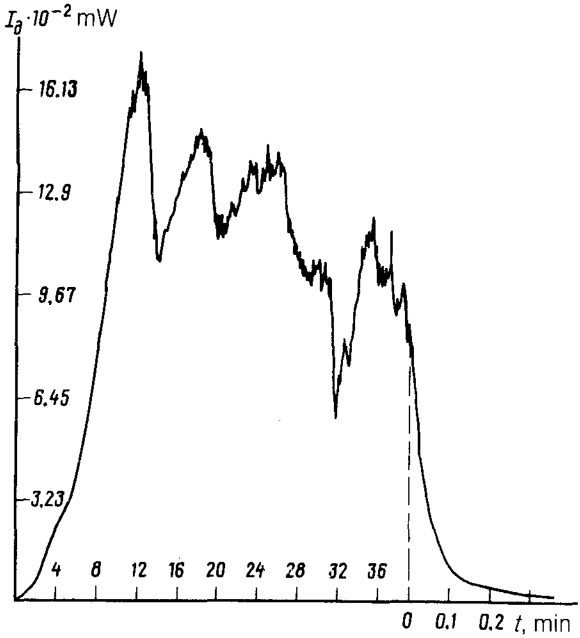

FIG. 3. Time dependence of the power of the conjugated beam starting from the time the laser was turned on. At the instant of time indicated the write beam was turned off. At the same time, the rate of advance of the recorder paper was increased from 2.5 mm/min to 100 mm/min.

the nonlinear media used in Refs. 1, 3, and 4. If we use an approximation linear in $d\epsilon/dL\ I$ and if the condition

$$|\nabla I||\mathbf{k}| \ll \frac{\epsilon}{c^2}\omega^2 I ,$$

From (1) follows an expression for the effective nonlinearity parameter:

$$\frac{\partial \mathbf{A}_d}{\partial z} = -i\frac{\omega}{c}\frac{dn}{dI}\mathbf{A}_r I_g e^{\alpha(z-h)} ,$$

where ϵ is the dielectric constant, $\mathbf{k}$ is the wave vector of the readout wave, $\mathbf{E}_r = A_r \exp(i\omega t - i\mathbf{kr})$ + complex conjugate (here ϵ, and $\mathbf{k}$ are complex if there is absorption), c is the velocity of light, ω is the frequency, n is the index of refraction, and z is the coordinate in the direction of propagation of the diffracted wave. The intensity of the diffracted wave is then given by the equation

$$I_d = \left[\frac{\omega}{\alpha c}(1-e^{-\alpha h})\frac{dn}{dI}I_g\right]^2 I_r e^{-\alpha h} \tag{1}$$

From (1) follows an expression for the effective nonlinearity parameter:

$$\frac{dn}{dI} = \frac{\alpha\lambda}{2\pi I_g}\frac{e^{\alpha h/2}}{1-e^{-\alpha h}}\sqrt{\frac{I_d}{I_r}} \tag{2}$$

where λ is the wavelength of the pump light. It should be mentioned that when the response of the medium is not a local function of the intensity, expression (2) for the effective nonlinearity parameter is only an approximate formula.

An estimate of the nonlinearity parameter dn/dI of the nonlinear media used previously[1,3,4] [using the parameters cited in those references in expression (2)], gives a value of 7.3×10^{-4} cm²/W for the liquid crystal,[3] 6×10^{-5} cm²/W for the absorbing liquid,[1] and 2.4×10^{-6} cm²/W for the dye solid solution.[4] In our case it follows from (2) that $dn/dI\approx1.4\times10^{-3}$ cm²/W; i.e., if we assume that the indices of refraction of the butyl cellosolve and water after the separation do not differ by more than in the third decimal place (on the order of 1.38 and 1.35),[5] we can conclude that even at $I_g\sim30$ W/cm², I_d should saturate (regardless of I_g or the grating period). We note that the value obtained for dn/dI exceeds considerably the value estimated in Ref. 8.

These experiments show that binary solutions with a low critical stratification point can be used as the basis for fundamentally new recording media with an effective nonlinearity parameter $dn/dI\gtrsim10^{-3}$ cm²/W. The pump power that can be used with them is an order of magnitude lower than that used for phase conjugation in other systems (~10 W/cm², compared to ~100 W/cm² in Refs. 1 and 3). The degree of restoration for conjugation in a stratifying solution is limited for the same reasons as in any kind of phase conjugation that uses the heating action of the radiation in a medium, since the amplitude of the induced thermal grating falls off as Λ^2 with decreasing characteristic period Λ of the interference pattern[3] (an exact solution of the thermal problem is given in Ref. 7). Moreover, the resolution of the stratifying solution as a recording medium is obviously limited also by the size of the particles of the disperse phase, the spatial distribution of which forms the image.

[1] A. A. Betin *et al.*, Kvantovaya Elektron. (Moscow) **12**, 1890 (1985) [Sov. J. Quantum Electron. **15**, 1248 (1985)].
[2] V. I. Bespalov *et al.*, IEEE J. Quantum Electron. **QE-25**, 360 (1989).
[3] L. Richard, J. Maurin, and J. P. Huignard, Opt. Commun. **57**, 365 (1986).
[4] G. Ravindra Kumar *et al.*, Opt. Commun. (1989), to be published.
[5] S. A. Viznyuk *et al.*, Pis'ma Zh. Eksp. Teor. Fiz. **45**, 559 (1987) [JETP Lett. **45**, 713 (1987)].
[6] S. A. Viznyuk and A. T. Sukhodol'skii, Short Communications in Physics [in Russian], No. 10, 14 (1988).
[7] S. A. Viznyuk *et al.*, Zh. Tekh. Fiz. (in press).
[8] N. K. Berger *et al.*, Short Communications in Physics [in Russian], No. 10, 11 (1988).

Translated by J. R. Anderson