Optics and Spectroscopy, Vol. 83, No. 6, 1997, pp. 926–929. Translated from Optika i Spektroskopiya, Vol. 83, No. 6, 1997, pp. 1001–1004.

HOLOGRAPHY

Simultaneous Spatial and Frequency Transformations of Laser Radiation Diffracted from a $\chi^{(2)}$-Hologram in the Bulk of a Glass

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Received October 17, 1996

Abstract—Recording of a grating of second-order nonlinear susceptibility ($\chi^{(2)}$-hologram) in the bulk of a glass capable of providing simultaneously the diffraction of laser radiation and its frequency doubling is proposed and realized. The period and the inclination angle of the $\chi^{(2)}$-grating recorded are calculated. Expressions for the irradiation angle of the grating recorded and the diffraction angle are obtained.

INTRODUCTION

In the last few years, great attention has been paid to the search for efficient materials for optical memory and optical processing of information. The use of photosensitive materials, for example, photorefractive crystals capable of varying their linear optical properties under the action of light, is the standard method for optical data storage [1–3]. A linear hologram created by modulation of the components of the linear susceptibility $\chi^{(1)}$ (refractive index and/or absorption coefficient) is formed in these materials in the recording process. In this case, the optical wavelength does not vary in the process of information readout.

At the same time, the phenomenon of photoinduced second harmonic generation (PSHG) has been known since 1987 [4]. In this process, information is recorded when a glass is irradiated by collinear light beams of the laser fundamental harmonic and its second harmonic simultaneously [5]. In this case, modulation of nonlinear optical properties of a glass takes place: a grating of the second-order nonlinear susceptibility modulated along the direction of propagation of light beams ($\chi^{(2)}$-grating) is created. To read out the information recorded, a glass is irradiated only by the laser fundamental harmonic. In this case, second harmonic generation at the $\chi^{(2)}$-grating recorded is observed. Recording and reading are performed at room temperature. Information recorded is not erased after numerous readouts and is conserved in a glass for a long time (several months) at room temperature.

After recording, coherent erasing of a $\chi^{(2)}$-grating recorded earlier is possible [6]. Such erasing is realized when glass is irradiated by the laser fundamental harmonic and its second harmonic simultaneously, but the relative phase shift between the first and the second harmonics differs by π from that used in recording.

Since the process of recording and readout involve light waves with significantly different wavelengths, a high signal-to-noise ratio can be obtained even for a low diffraction efficiency (of order of 10^{-8}) through the use of spectral selection. Recording of $\chi^{(2)}$-holograms with such an efficiency is possible in glasses of certain compositions upon irradiation for a time of order of 10^{-8} s using 30 ps pulses of energy 0.5 mJ [7, 8].

Thus, the PSHG phenomenon possesses a number of advantages over traditional processes of information recording. These advantages made it possible to discuss the possibility of the utilization of this phenomenon for a long-term optical memory with the use of frequency multiplexing [9] and the development of a neuron matrix in an optical neural network [10].

This work was aimed at studying the possibility of producing a $\chi^{(2)}$-hologram in a glass that would allow spatial transformation of a light beam to be performed concurrently with radiation frequency doubling.

RESULTS AND DISCUSSION

At present, several different microscopic models describing the PSHG phenomenon are available (see, for example, [11, 12]). Since glasses are isotropic materials in which the macroscopic second-order nonlinear susceptibility is zero, all the models proposed are associated, in one way or another, with the assumption of the appearance of a regular structure in a glass upon optical recording. Ordering in these models occurs either due to directed charge separation resulting from the coherent photovoltaic effect or due to orientation of dipoles in the optically rectified field.

In spite of the fact that the physical mechanism of recording is not conclusively clear, most researchers consider that the amplitude of a $\chi^{(2)}$-grating recorded is

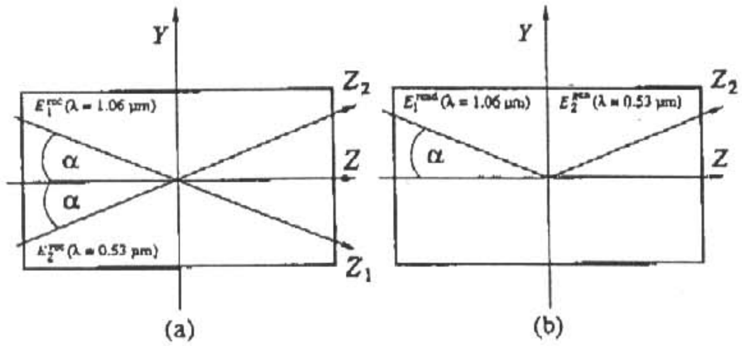

Fig. 1. Diagram of interaction of light beams in the formation of the $\chi^{(2)}$-grating: (a) recording, (b) readout. Z_1 and Z_2 are the directions of propagation of the beams of the first (E_1^{rec}) and the second (E_2^{rec}) harmonics in recording, E_1^{read} is the incident beam with a wavelength of 1064 nm, E_2^{gen} is the diffracted beam with a wavelength of 532 nm.

determined by the steady-state term of the cube of the total light field used for recording

$$\chi^{(2)} \sim E_1^{rec} E_1^{rec} E_2^{rec*}. \quad (1)$$

Therefore, in the general case of simultaneous action of the field of the fundamental harmonic of laser radiation

$$E_1^{rec} = \mathscr{E}_1^{rec} \exp(i\mathbf{k}_1 z_1 - i\omega t) = \mathscr{E}_1^{rec} \exp(i\mathbf{k}_1(z\cos(\alpha) - y\sin(\alpha)) - i\omega t) \quad (2)$$

and the field of the second harmonic coherent with it

$$E_2^{rec} = \mathscr{E}_2^{rec} \exp(i\mathbf{k}_2 z_2 - 2i\omega t) = \mathscr{E}_2^{rec} \exp(i\mathbf{k}_2(z\cos(\alpha) + y\sin(\alpha)) - 2i\omega t) \quad (3)$$

(where $\mathscr{E}_1$ and $\mathscr{E}_2$ are the slowly varying amplitudes, $\mathbf{k}_1$ and $\mathbf{k}_2$ are the wave vectors, z_1 and z_2 are the directions of propagation, ω and 2ω are the frequencies of the fundamental and the second harmonics, 2α is the angle between the beams of the fundamental and the second harmonics), a $\chi^{(2)}$-grating, i.e., spatially modulated second-order nonlinearity, is recorded in a glass (Fig. 1)

$$\chi^{(2)}(y, z) \sim \mathscr{E}_1^{rec} \mathscr{E}_1^{rec} \mathscr{E}_2^{rec*} \times \exp(iz\cos(\alpha)(2\mathbf{k}_1 - \mathbf{k}_2) - iy\sin(\alpha)(2\mathbf{k}_1 + \mathbf{k}_2)). \quad (4)$$

If $\alpha = 0$, we obtain a familiar expression for the amplitude of a $\chi^{(2)}$-grating modulated only in one direction, along the Z-axis

$$\chi^{(2)}(z) \sim \mathscr{E}_1^{rec} \mathscr{E}_1^{rec} \mathscr{E}_2^{rec*} \exp(iz(2\mathbf{k}_1 - \mathbf{k}_2)). \quad (5)$$

It was this case of collinear recording of a $\chi^{(2)}$-grating that was the subject of studies of preceding works in this field.

In the case of noncollinear recording ($\alpha \neq 0$), one can obtain from expression (4) that $\chi^{(2)}$-grating can be characterized by a grating vector $\mathbf{k}_g$ with a length determined by the expression

$$\mathbf{k}_g^2 = 4\mathbf{k}_1^2 + \mathbf{k}_2^2 - 4\mathbf{k}_1\mathbf{k}_2\cos(2\alpha), \quad (6)$$

and the direction determined by the angle γ (relative to the Z-axis)

$$\tan(\gamma) = \tan(\alpha)(n_{2\omega} + n_\omega)/(n_{2\omega} - n_\omega), \quad (7)$$

where n_ω and $n_{2\omega}$ are the refractive indices of a glass at frequencies ω and 2ω.

It follows from expressions (6) and (7) that the vector $\mathbf{k}_g$ of the grating recorded can be determined with the help of the vector diagram depicted in Fig. 2. It is also clear from this figure that irradiation of a $\chi^{(2)}$-grating recorded by radiation with a wave vector $\mathbf{k}_1$ will result in generation of a wave with doubled frequency propagating in the direction of the vector $\mathbf{k}_2$. In this case, the period L_Z of the $\chi^{(2)}$-grating along the Z-axis determined by expression

$$L_Z = \lambda_2/(\cos(\alpha)(n_{2\omega} - n_\omega)) \quad (8)$$

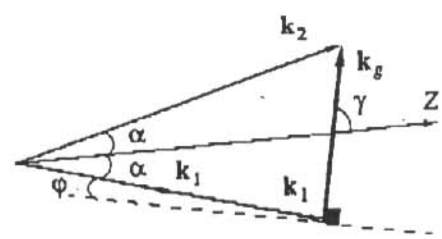

Fig. 2. Vector diagram of diffraction from the $\chi^{(2)}$-grating. $\mathbf{k}_1$ is the wave vector of incident radiation with a wavelength of 1064 nm, $\mathbf{k}_2$ is the wave vector of diffracted radiation with a wavelength of 532 nm, $\mathbf{k}_g$ is the wave vector of the $\chi^{(2)}$-grating.

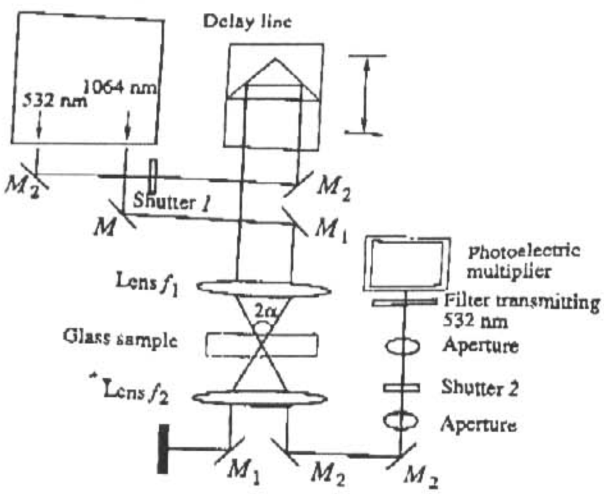

Fig. 3. Schematic of the experimental setup. M_1 are mirrors reflecting 1064 nm; M_2 are mirrors reflecting 532 nm; the focal lengths of the collecting lenses f_1 and f_2 are 50 mm. Parameters of the delay line were selected in such a way as to provide temporal coincidence of light pulses in the glass sample. The angle between the beams in the sample is 12°. After recording, the sample was irradiated by the first harmonic alone, and green radiation propagating in the direction of second harmonic radiation used for recording was detected by a Hamamatsu 853 01 photoelectric multiplier. In the process of recording, shutter 1 was opened and shutter 2 was closed; in the process of readout, the states were converse.

allows quasi-phase-matched second harmonic generation to be obtained, while the period along the *Y*-axis

$$L_Y = \lambda_2/(\sin(\alpha)(n_{2\omega}+n_\omega)), \quad (9)$$

(where λ_1 and λ_2 are the wavelengths of the first and the second harmonics, respectively) determines the beam deviation in the *ZY*-plane.

Expression (9) can be rewritten in the form

$$\sin(\alpha) = \lambda_2/(2L_Y(n_{2\omega}+n_\omega)/2), \quad (1$$

which changes to the familiar Bragg condition in t case of light diffraction from an ordinary grating wit out wavelength variation

$$\sin(\alpha) = \lambda/(2Ln), \quad (1$$

where λ is the wavelength of light, L is the gratir period, and n is the refractive index.

Therefore, simultaneous irradiation of a glass by th first and the second harmonics of the form (2) and (results [in the case when a grating is formed accordin to expression (1)] in the formation of a $\chi^{(2)}$-gratin determined by expressions (6) and (7) in the glass. T read out the information recorded this grating should b irradiated by a plane wave with a wavelength λ_1 at th angle φ to the grating plane (Fig. 2)

$$\varphi = \alpha + \gamma - 90, \quad (12$$

where the angle γ is determined by expression (7) an the angle α is determined by the modified Bragg condi tion (10). In this case, light will diffract with a deflec tion through an angle 2α and concurrent doubling o the incident wave frequency.

Recording of $\chi^{(2)}$-gratings in a glass was studied exper imentally using a setup shown in Fig. 3. A Coheren Antares 76s picosecond Nd : YAG laser was used wit a pulse repetition rate of 76 MHz, with an intracavit frequency doubler with 90° rotation of the plane o polarization of the second harmonic to obtain coheren polarizations. The pulse lengths at the wavelengths o 1064 and 532 nm were 100 and 60 ps, respectively; th mean powers were 20 W and 400 mW, respectively. Ar optical delay line was used to make light pulses coinci- dent in time. A sample of multicomponent lead–silicate

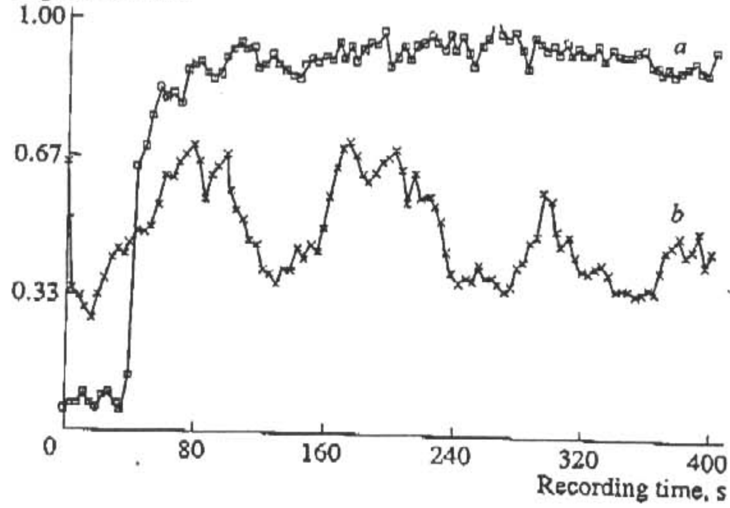

Fig. 4. Results of the measurements of the diffraction efficiency of the $\chi^{(2)}$-grating as a function of time. (a) recording; (b) repeated recording–erasing of the $\chi^{(2)}$-hologram.

glass (19 mol % of PbO and 68 mol % of SiO_2) 3 mm thick was used to record $\chi^{(2)}$-gratings. Second harmonic radiation upon passing the delay line was focused into the sample by a 50-mm lens. The optical system was aligned in such a way as to provide maximum spatial overlap of infrared and green pulses in the sample. The angle between the beams in the sample (2α) was 12°. After recording, the sample was irradiated by the first harmonic alone, and green radiation propagating in the direction of second harmonic radiation used for recording was detected by a Hamamatsu 853 01 photomultiplier connected to an SR400 counter [13].

Record–readout cycles were repeated with a period of 4 s. In the recording process, the first shutter was opened and the second was closed, their states being converse in the readout process. Such a technique allowed the diffraction efficiency to be obtained as a function of the recording time.

The results of the measurements of the diffraction efficiency D as a function of the recording time are shown in Fig. 4. Curve *a* in Fig. 4 represents a typical dependence with a rapid growth of the diffracted signal up to a certain saturation level. Curve *b* shows the time dependence of the diffraction efficiency as the angular position of the sample is varied. Nonmonotonic behavior of this dependence is likely to be determined by a variation in the relative location of light beam waists (with a transverse size of order of 15 μm) in the focal plane of the lens as well as by the instability of spatial characteristics of the laser. This results in recording of one grating, then its erasing and recording of another grating. The maximum diffraction efficiency was 10^{-8}, which allowed a signal-to-noise ratio of order of 300 to be obtained.

Recording was made with an angle $\alpha = 6°$, which allowed the parameters of the $\chi^{(2)}$-grating produced to be calculated by using expressions (8) and (9): $L_Z = 30$ μm, $L_Y = 1.4$ μm.

CONCLUSIONS

Thus, recording of a hologram of the second-order nonlinear susceptibility by noncollinear light beams in a glass is proposed and realized in this work. The $\chi^{(2)}$-grating recorded is shown to be capable of simultaneous deflection and frequency doubling of the incident light. The possibility of erasing the $\chi^{(2)}$-grating recorded by varying space–time parameters of radiation is demonstrated. The maximum diffraction efficiency of order of 10^{-8} and the deflection angle of 12° are obtained.

The period and the inclination angle of the $\chi^{(2)}$-grating recorded are calculated, expressions for the irradiation angle of a grating recorded and the diffraction angle are derived.

The results obtained demonstrate the possibility of recording $\chi^{(2)}$-holograms capable of reconstructing images recorded concurrently with frequency doubling of the incident light in the bulk of a glass. This extends the possibilities for the development of devices for optical memory and information processing in comparison with the use of ordinary $\chi^{(1)}$-holograms.

ACKNOWLEDGMENTS

The authors are grateful to Yu.N. Denisyuk for helpful discussions.

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Translated by A. Mozharovskii