Weak response measurements of photosensitive media

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A high-sensitivity interferometric method to measure parameters of volume diffraction gratings written in photosensitive media is proposed. The method is based on measuring the transient energy transfer among phase-modulated coherent beams that results from their own diffraction with dynamic gratings in the medium. It is theoretically shown that there is a simple mathematical dependence between the amplitudes of the output beams intensity modulation and the grating parameters. The method was applied to measure the photoresponse of polymer films containing bacteriorhodopsin.

Keywords: Photosensitive media; phase-modulated beams; measurement of volume grating amplitude

Se propone un método interferométrico de gran sensibilidad para medir parámetros de rejillas de difracción volumétricas grabadas en un medio fotosensitivo. El método está basado en la medición de la transferencia de energía transitoria entre haces coherentes modulados en fase, que resultan de la difracción con las rejillas dinámicas inmersas en el medio. Se demuestra teóricamente que existe una relación matemática sencilla entre las amplitudes moduladas de los haces de salida y los parámetros de la rejilla. Este método fue aplicado para medir la fotorrespuesta de películas poliméricas que contenían bacteriorodopsina.

Descriptores: Medias fotosensitivas; haces modulados en fase; mediciones de amplitud de rejillas de volumen

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1. Introduction

The parameters of volume holographic gratings written in photosensitive media are usually measured using a low-intensity probing beam. Besides its complexity, such procedure does not have high accuracy since the diffracted beam at the grating output is very small for low grating amplitudes. The diffraction efficiency η for the refractive index and absorption coefficient gratings is given by [1, 2]

$$\eta = \exp\left(-\frac{2\bar{\alpha}d}{\cos\theta}\right)\sin^2\left(\frac{\pi n_1 d}{\lambda\cos\theta}\right) \qquad (1)$$

$$\text{or} \qquad \eta = \exp\left(-\frac{2\bar{\alpha}d}{\cos\theta}\right)\sinh^2\left(\frac{\alpha_1 d}{2\cos\theta}\right), \qquad (2)$$

where n_1 and α_1 are the amplitude variation of the refractive index and the absorption coefficient respectively, d is the medium thickness, θ is the Bragg angle, λ is the wavelength of light in vacuum, and $\bar{\alpha}$ is the mean absorption coefficient. These formulae allow the calculation of the grating amplitudes using a measured diffraction efficiency of the medium.

A large number of novel photosensitive media, like polymer films, containing different types of bacteriorhodopsin (BR) [3, 4], dye [5], azobenzene [6], charge-transfer polymers [7], etc. have many advantages over high-efficiency photorefractive crystals. However they have low photoresponse due to small values of $n_1 d/\lambda$ and $\alpha_1 d$, which characterize the media diffraction efficiency. The diffraction efficiency of such media is approximately equal to $\exp(-2\bar{\alpha}d/\cos\theta) \times (\pi n_1 d/\lambda_a \cos\theta)^2$ or $\exp(-2\bar{\alpha}d/\cos\theta) \times (\alpha_1 d/2\cos\theta)^2$, depending on the grating type. For example, in the case of a pure refractive grating, if $n_1 = 10^{-5}$, $d = 50$ μm and $\lambda = 1$ μm, the diffraction efficiency is around 10^{-6}. The measurement of this value using the probing beam is very difficult and inaccurate. Another method to measure the media photoresponse, especially for media with weak optical non-linearity, is hereby proposed. This method has very high sensitivity because it is based on interferometric measurements. The main idea behind the method is in an analysis of a transient energy transfer among phase-modulated coherent light beams that result from their own diffraction with dynamic gratings in the photosensitive media. The output beam intensity modulation at the first and second harmonic of the light beams mutual phase modulation allows the of the calculation variations of the media refractive index and absorption coefficient. The method gives the possibility to measure these parameters independently and in real time. In addition, the method makes it possible to investigate media with nonlocal photoresponse, *i.e.* when interference pattern and dynamic grating maxima are spatially mismatched. The method was applied to the experimental investigation of weakly nonlinear polymer films containing BR.

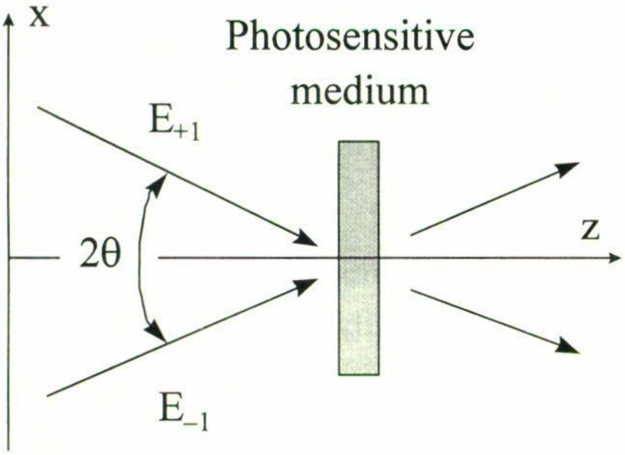

FIGURE 1. Geometry of two-wave mixing.

2. Theoretical description of the method

Equations describing the correlation between output beam intensity modulation and holographic parameters of the photosensitive media with weak optical non-linearity were obtained using the Coupled Wave Theory [2]. Let two y-polarized electromagnetic waves $E_{\pm 1}(x,z) = E_{\pm 1}(z)\exp(\pm ik\sin\theta x - ik\cos\theta z)$ go into the photorefractive medium (Fig. 1). $E_{\pm 1}(z)$ are the wave's amplitudes, $k = 2\pi/\lambda$ is the wave propagation constant and θ is the Bragg angle. The medium input surface is at $z = 0$, with a medium thickness of d. The interference pattern obtained from $E_{\pm 1}$ results in a grating for the refractive index and the absorption coefficient

$$\varepsilon(z) = \bar{\varepsilon} + \varepsilon_1(z)\cos(Kx + \varphi_\epsilon),$$
$$\alpha(z) = \bar{\alpha} + \alpha_1(z)\cos(Kx + \varphi_\alpha), \tag{3}$$

where ε_1 and α_1 are the grating amplitudes, $\bar{\varepsilon}$ is the averaged medium permittivity, $K = 2\pi/\Lambda$, Λ is the grating period, φ_ε and φ_α are the mismatches between the interference pattern and the gratings. In case of weak optical non-linearity the beams phases are approximately constant along the medium thickness. Substituting the sum of the electromagnetic waves and the grating amplitudes into the wave equation [2], and then grouping the terms with exponential factors, a solution for the wave equation may be obtained:

$$2i\cos\theta\frac{dE_{\pm 1}(z)}{dz} = -\bar{\alpha}E_{\pm 1}(z) - (ikn_1(z)\exp(i\varphi_\varepsilon) + \alpha_1(z)\exp(i\varphi_\alpha))E_{\mp 1}(z), \tag{4}$$

where $n_1 = \varepsilon_1/2\sqrt{\bar{\varepsilon}}$ is the grating's refractive index amplitude. Second order derivative terms of the electromagnetic wave amplitudes were neglected because the electric field amplitude changes very slowly. Terms with exponential factors like $\exp(\pm 3ik\sin\theta x)$ were also neglected because they do not satisfy the Bragg condition. The amplitude of the electric fields $E_{+1}(z)$ and $E_{-1}(z)$ can be written as

$$E_{\pm 1}(z) = \sqrt{I_{\pm 1}(z)}\exp(i\varphi_{\pm 1}), \tag{5}$$

where $I_{+1}(z)$ and $I_{-1}(z)$ are the beam intensities, φ_{+1} and φ_{-1} are the beam phases. Inserting (5) into (4) results in equations for the intensities and phases:

$$\cos\theta\frac{dI_{\pm 1}(z)}{dz} + iI_{\pm 1}(z)\frac{d\varphi_{\pm 1}(z)}{dz} = -\bar{\alpha}I_{\pm 1}(z) + \{-ikn_1(z)\exp[i(\mp\varphi + \varphi_\varepsilon)] + \alpha_1(z)\exp[i(\mp\varphi + \varphi_\alpha)]\} \times \sqrt{I_{+1}(z)I_{-1}(z)}, \tag{6}$$

where $\varphi = \varphi_{+1} - \varphi_{-1}$. Equation (6) can be separated in two equations, one for the real part and the other for the imaginary part. The real part gives an equation for beam intensities:

$$\cos\theta\frac{dI_{\pm 1}(z)}{dz} = -\bar{\alpha}I_{\pm 1}(z) + [\mp kn_1(z)\sin(\varphi + \varphi_\varepsilon) + \alpha_1(z)\cos(\varphi + \varphi_\alpha)]\sqrt{I_{+1}(z)I_{-1}(z)}. \tag{7}$$

If n_1 and α_1 are small, an approximate solution of Eq. (7) is obtained by expanding this in series in terms of a small parameter proportional to $(\pm kn_1\sin\psi_\varepsilon + \alpha_1\cos\psi_\alpha)z$, where $\psi_{\varepsilon,\alpha} = \varphi + \varphi_{\varepsilon,\alpha}$:

$$I_{\pm 1}(z) = I^0_{\pm 1}(z) + I^1_{\pm 1}(z) + \ldots \tag{8}$$

For the zeroth approximation (absence of the grating)

$$I^0_{\pm 1}(z) = I_{\pm 10}\exp\left(-\frac{\bar{\alpha}z}{\cos\theta}\right), \tag{9}$$

where $I_{\pm 10}$ is the input beam intensity. Small increases in the light intensities $I^1_{\pm 1}(z)$ are given by the first order correction, assuming a weak grating,

$$\cos\theta\frac{dI^1_{\pm 1}(z)}{dz} = -\bar{\alpha}I^1_{\pm 1}(z) + \frac{1}{2}m[\mp kn_1(z)\sin(\psi_\varepsilon) + \alpha_1(z)\cos(\psi_\alpha)]I_0(z), \tag{10}$$

where $I_0(z)$ is the total light intensity, m is the interference pattern contrast. For the case of beams with sinusoidal phase modulation, the mutual phase φ of the light beams is equal to $a\sin\Omega t$, where a is the amplitude of the phase modulation, and Ω is the angular frequency. This modulation is necessary to measure the media photoresponse. It should be noticed that in the general case the intensities $I^1_{\pm 1}(z)$ depend on the phase modulation frequency Ω, however at high frequencies ($\Omega \gg 1/\tau$, where τ is the photoresponse relaxation time) this dependence is very small and may be ignored. Expanding $\sin(\varphi_{\alpha,\varepsilon} + a\sin\Omega t)$ and $\cos(\varphi_{\alpha,\varepsilon} + a\sin\Omega t)$ in a series of Bessel functions and considering only first and second harmonic terms of the external phase modulation frequency, equations for the intensity modulation amplitude of the light beams are obtained,

$$\cos\theta\frac{dI^{\Omega}_{\pm 1}(z)}{dz} = -\bar{\alpha}I^{\Omega}_{\pm 1}(z) + (\mp kn_1(z)\cos\varphi_\varepsilon + \alpha_1(z)\sin\varphi_\alpha)J_1(a)mI_0(z),$$

$$\cos\theta\frac{dI^{2\Omega}_{\pm 1}(z)}{dz} = -\bar{\alpha}I^{2\Omega}_{\pm 1}(z) + (\mp kn_1(z)\sin\varphi_\varepsilon + \alpha_1(z)\cos\varphi_\alpha)J_2(a)mI_0(z), \quad (11)$$

where $J_{1,2}$ are Bessel functions of the first and second orders. Using (9) and considering that $n_1(z) = n_1(0)\exp(-\bar{\alpha}z/\cos\theta$ and $\alpha_1(z) = \alpha_1(0)\exp(-\bar{\alpha}z/\cos\theta)$, expressions for $I^{\Omega}_{\pm 1}(d)$ and $I^{2\Omega}_{\pm 1}(d)$ are obtained after simple transformations:

$$I^{\Omega}_{\pm 1}(d) = \frac{\cos\theta}{\bar{\alpha}}[\mp kn_1(0)\cos\varphi_\varepsilon + \alpha_1(0)\sin\varphi_\alpha] \times mI_0(0)J_1(a)\exp\left(-\frac{\bar{\alpha}d}{\cos\theta}\right)\left(1-\exp\frac{\bar{\alpha}d}{\cos\theta}\right),$$

$$I^{\Omega}_{\pm 1}(d) = \frac{\cos\theta}{\bar{\alpha}}[\pm kn_1(z)\sin\varphi_\varepsilon + \alpha_1(z)\cos\varphi_\alpha] \times mI_0(0)J_1(a)\exp\left(-\frac{\bar{\alpha}d}{\cos\theta}\right)\left(1-\exp\frac{\bar{\alpha}d}{\cos\theta}\right). \quad (12)$$

Relative amplitudes of the intensity modulation of the medium output beams at the first and second harmonic of the external phase modulation ($\delta I^{\Omega}_{\pm 1}(d) = I^{\Omega}_{\pm 1}(d)/I_0(d)$ and $\delta I^{2\Omega}_{\pm 1}(d) = I^{2\Omega}_{\pm 1}(d)/I_0(d)$) have the form

$$\delta I^{\Omega}_{\pm 1}(d) = \frac{\cos\theta}{\alpha}[\mp kn_1(0)\cos\varphi_\varepsilon + \alpha_1(0)\sin\varphi_\alpha] \times J_1(a)\left[1-\exp\left(-\frac{\bar{\alpha}d}{\cos\theta}\right)\right],$$

$$\delta I^{2\Omega}_{\pm 1}(d) = \frac{\cos\theta}{\alpha}[\pm kn_1(0)\sin\varphi_\varepsilon + \alpha_1(0)\cos\varphi_\alpha] \times J_2(a)\left[1-\exp\left(-\frac{\bar{\alpha}d}{\cos\theta}\right)\right]. \quad (13)$$

Finally, using a difference amplitudes at the first and second harmonic separately, results in equations for a phase mismatch between interference pattern and gratings, and the amplitudes of the grating's refractive index and absorption coefficient at the input medium plane,

$$\tan\varphi_\varepsilon = \frac{\delta I^{2\Omega}_{+1}(d) - \delta I^{2\Omega}_{-1}(d)}{\delta I^{\Omega}_{-1}(d) - \delta I^{\Omega}_{+1}(d)} \times \frac{J_1(a)}{J_2(a)}, \quad (14)$$

$$\tan\varphi_\alpha = \frac{\delta I^{\Omega}_{+1}(d) + \delta I^{\Omega}_{-1}(d)}{\delta I^{2\Omega}_{-1}(d) + \delta I^{2\Omega}_{+1}(d)} \times \frac{J_2(a)}{J_1(a)}, \quad (15)$$

$$n_1(0) = \frac{\bar{\alpha}\lambda[\delta I^{\Omega}_{-1}(d) - \delta I^{\Omega}_{+1}(d)]}{4\pi\cos\theta J_1(a)\left[1-\exp\left(-\frac{\bar{\alpha}d}{\cos\theta}\right)\right]\cos\varphi_\varepsilon} = \frac{\bar{\alpha}\lambda[\delta I^{2\Omega}_{+1}(d) - \delta I^{2\Omega}_{-1}(d)]}{4\pi\cos\theta J_2(a)\left[1-\exp\left(-\frac{\bar{\alpha}d}{\cos\theta}\right)\right]\sin\varphi_\varepsilon}, \quad (16)$$

$$\alpha_1(0) = \frac{\bar{\alpha}[\delta I^{\Omega}_{-1}(d) - \delta I^{\Omega}_{+1}(d)]}{2\cos\theta J_1(a)\left[1-\exp\left(-\frac{\bar{\alpha}d}{\cos\theta}\right)\right]\sin\varphi_\alpha} = \frac{\bar{\alpha}[\delta I^{2\Omega}_{+1}(d) + \delta I^{2\Omega}_{-1}(d)]}{2\cos\theta J_2(a)\left[1-\exp\left(-\frac{\bar{\alpha}d}{\cos\theta}\right)\right]\cos\varphi_\alpha}. \quad (17)$$

For the experimental determination of these parameters it is necessary to use the optimal value of the phase modulation amplitude. If the medium under study is not saturated, the amplitude of the output beams intensity modulation is proportional to $J_0(a)J_1(a)$ at the first harmonic of the phase modulation, and to $J_0(a)J_2(a)$ at the second one. The maxima of these functions are respectively reached at $a = 1.1$ rad., and at $a = 1.5$ rad. In this case the interference pattern contrast and, correspondingly, the grating amplitudes are decreased by $J_0(a)$. If the tested medium is under absorption saturation, it is necessary to maintain the contrast of the interference pattern by applying the phase modulation at smaller amplitude (a < 0.5 rad.).

Theoretically the maximum sensitivity of the method is limited by the light quantum noise. Pertinent calculation shows that, if input laser power is 1 mW, the medium thickness is 100 μm, the absorption coefficient is 10 mm^{-1} and photodetector frequency band is 100 Hz, the quantum limit gives grating refractive index and absorption coefficient amplitudes minima around 10^{-16} and 10^{-11} cm^{-1} respectively. Practically the method's sensitivity is limited by laser and photodetector noises and mechanical perturbations. Thus the method's sensitivity for refractive index variation is set at 10^{-7}, and to 10^{-4} cm^{-1} for absorption coefficient variation.

3. Experimental determination of the refractive grating amplitude for a polymer film containing bacteriorhodopsin

Using the method of phase modulated beams, the photoresponse of polymer films containing wild-type bacteriorhodopsin (BR) was measured [8] . BR is a biological photosensitive material having reversible changes of absorption and refractive index, which are observed during the photocycle of BR molecules. This material was embedded in the polymer film formed on a glass plate surface. The sample had a thickness of 100 μm, an absorption coefficient of 12 mm^{-1} and a photoresponse relaxation time of 1 s.

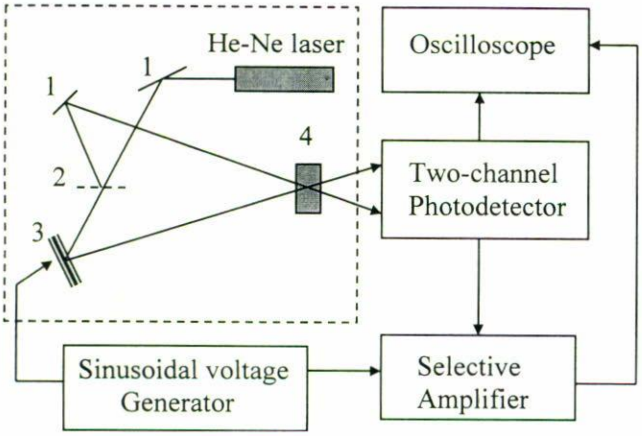

FIGURE 2. Schematic diagram of the experimental arrangement. 1-mirrors, 2-beam-splitting plate, 3-mirror bonded to piezoelectric modulator, 4-polymer film containing bacteriorhodopsin.

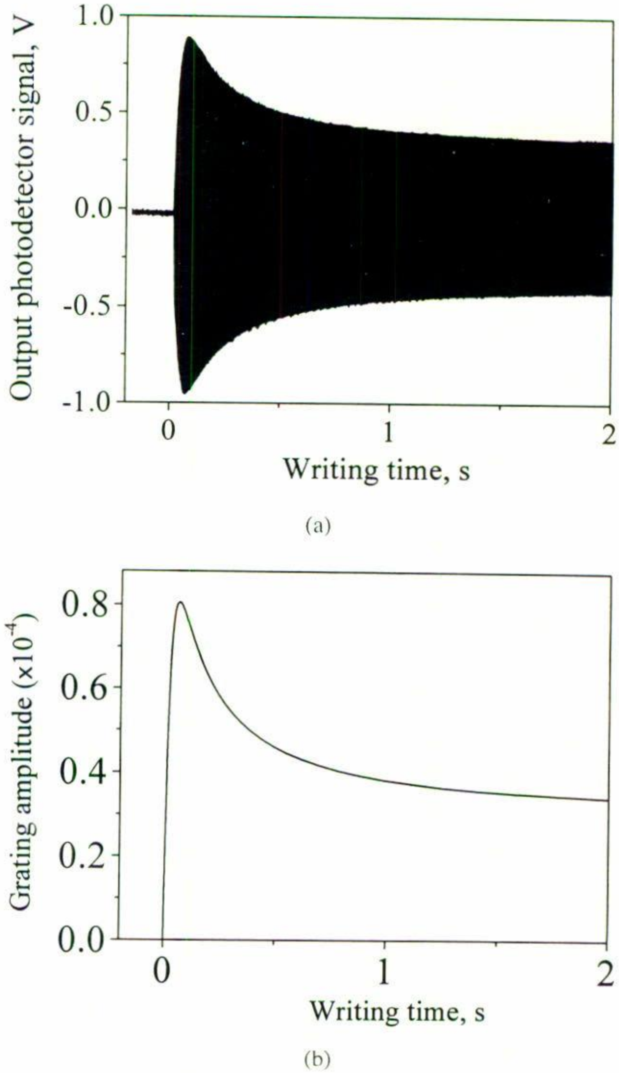

FIGURE 3. Output photodetector signal (a) and grating refractive index amplitude (b) correspond to start of the grating formation in polymer film containing bacteriorhodopsin.

The experimental setup of the two-beam interferometer utilizing the phase-modulated beams method for the experimental study of weak photoresponse is shown in Fig. 2. To record a photorefractive grating in a medium containing BR, coherent light from a He-Ne laser at 633 nm was divided by a semitransparent plate into two beams $I_{+1} = I_{-1} = I_0/2$, which were sent by mirrors 1 and 3 to a BR-containing polymer film. Mirror 3 was attached to an electromagnetic modulator, which was connected to a sinusoidal voltage generator operating at Ω. This modulator produced the light beams mutual phase modulation with a frequency band from 20 Hz up to 20 kHz, and allowed to measure the photoresponse with characteristic times less than 5×10^{-3}s. The use a high-frequency electro-optic phase modulator instead of an electromagnetic one allows the measurement of very fast photoresponses (up to 10^{-10}s). The frequency band of the selective amplifier needs to be at least twice as wide as the reciprocal characteristic time of the registered process. To decrease the laser noise influence, a two-channel differential photodetector and a selective amplifier were used. The photodetector output voltages at the first and second harmonic of the phase modulation gives the possibility to calculate the grating amplitudes.

Figure 3 a shows a typical oscilloscope trace of the selective amplifier output signal for the laser intensity $I_0 = 2$ mW/mm^2 at the first harmonic of the phase modulation frequency. It is known that BR has pure local photoresponse and therefore this signal describes only the grating refractive index amplitude. The calculated temporal dependence of the grating amplitude during recording is shown in Fig. 3 b. Such behavior in the grating recording is explained by the absorption saturation of BR molecules [9], which leads to spatial profile curving of the grating and as a result to grating fundamental spatial harmonic decrease. The maximum and the steady-state grating amplitudes of the BR sample is 8×10^{-5} and 4×10^{-5} respectively. These values are comparable with the refractive index variation of high-efficiency photorefractive crystals. Thus the weak optical non-linearity of the polymer films containing BR is due to its small thickness and high absorption coefficient.

4. Measurement of the transient energy transfer and the phase mismatch between the interference pattern and the grating during the writing process

The proposed method of phase modulated beams permits to measure weak effects occurring during the grating formation. One of them is a transient energy transfer and a transient phase mismatch between the interference pattern and the refractive grating during its formation in the photosensitive medium caused by unequal intensities of the writing beams. This effect is observed as a consequence of an energy exchange re-

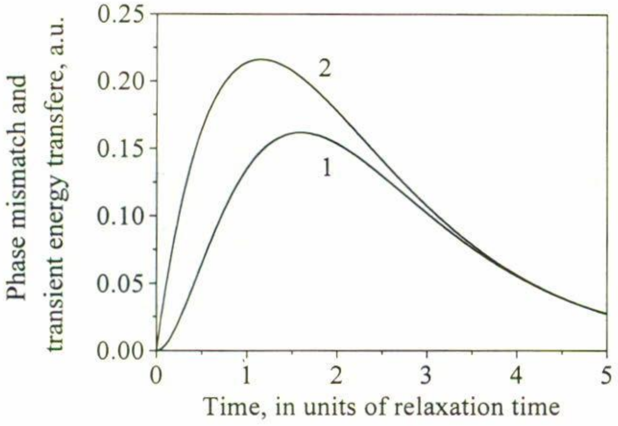

FIGURE 4. Theoretical temporal dependence of the transient energy transfer (curve 1) and the phase mismatch (curve 2) during the refractive grating formation in the photorefractive medium, with weak local response under unequal intensities of the writing beams.

sulting from a phase transfer between writing beams, resulting in the grating tilt. It is worth noting that this effect is observed in reversible photosensitive media with local response [10]. Theoretically the transient energy transfer is observed if the second order correction to the beams intensities is taken into account [Eq. (8)]. The method permits to evaluate this effect even if the media photoresponse is very weak.

Simultaneous solution of a nonlinear system for the permittivity and the wave equations under unequal intensities of writing beams, shows that the temporal dependence of the transient energy transfer and the phase mismatch between refractive grating and the interference pattern in media with weak photoresponse, is proportional to $\exp(-t/\tau)\{1-t/[\tau(1-\exp(-t/\tau)]\}$ and to $\exp(-t/\tau)\{(t/\tau)-[1-\exp(-t/\tau)]\}$ respectively [10]. Plots of these functions are depicted in Fig. 4. It is easy to see that during the grating formation the phase mismatch and the transient energy transfer increases with grating amplitude growth, then reaches a maxim and decreases towards zero. It corresponds to a tilting of the grating to a steady-state position defined by the writing beams ratio. Every time the transient phase mismatch is changed from zero at the input medium surface plane, it changes to a maximum at the output plane [10].

The phase modulation of the writing beams gives the possibility to measure the transient energy transfer during grating formation, and to obtain the phase mismatch between the interference pattern and the grating. Pertinent calculations show that the output beam intensity modulation amplitude at the second harmonic under compensation of the absorption grating signal is proportional to the transient energy transfer, and can be used for necessary measurements. Proper use of the differential photodetector and the selective amplifier allowed the study of a small increment to the constant level of the output beams intensities. This increment gives additional information for evaluation of the amplitude of the transient energy transfer and the phase mismatch. Figure 5 shows the amplitude of the selective amplifier output signal corresponding to the first and second harmonic of the output beams intensities modulation, and the averaged phase mismatch calculated using Eq. (14). These measurements were obtained in absence of absorption saturation (total writing beams intensity was 0.1 mW/mm^2). Such experimental condition was necessary to achieve the output signal maximum and to decrease the effects related to saturation. The amplitude of the phase modulation was chosen around 1.5 rad. The measured maximum of the transient energy transfer was 0.15% of the output beam intensity. This corresponds to the averaged phase mismatch maximum of 3.8 degrees. It should be noticed that the output detector signal at the second harmonic had higher noise levels than for the first one. This effect is explained by the high sensitivity of the interferometer to acoustic perturbations at the second harmonic of the beams mutual phase modulation.

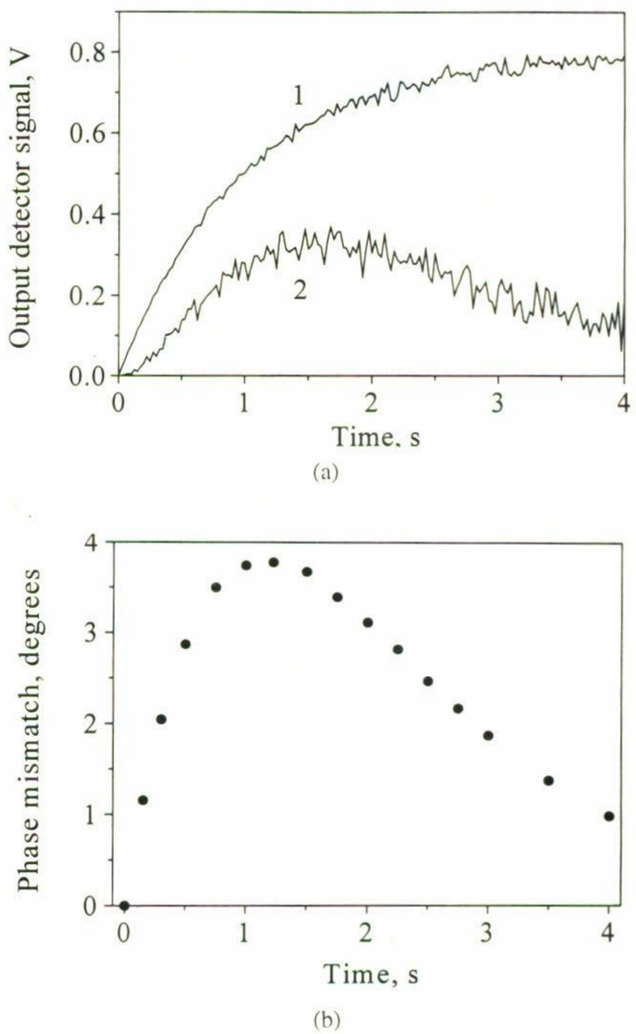

FIGURE 5. Amplitudes of the output detector signal at first (curve 1) and second (curve 2) harmonic (a) and phase mismatch (b).

5. Conclusions

In summary, a high-sensitive interferometer method was presented for the photoresponse measurement of photosensitive media. The method is characterized by the simplicity of its experimental realization as compared with the use of a probing beam. It also shows higher accuracy due to the interferometric measurement and selective amplification of the data signals, and has the possibility of determining the angle mismatch between the interference pattern and the grating. The sensitivity of the method permits measurement of weak effects that are connected with the grating formation process. This is of extreme importance to fully understand the intricacies of the two-wave mixing process in the media being tested. All these advantages make it possible to apply this method in the investigation of non-linear optical properties of a broad class of novel photosensitive media.

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