Optically stimulated electron–hole resonance in photorefractive CdTe with a low frequency ac field

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We report the results of an experimental and theoretical study of electron–hole competition in CdTe:Ge photorefractive crystal with an incoherent auxiliary illumination and alternating low-frequency field. Resonant two-wave mixing (TWM) gain enhancement is studied that has been found depending on the wavelength and intensity of the incoherent illumination. We show that a low-frequency ac field can be used for an effective TWM gain enhancement under conditions appropriate to the electron–hole resonance. A time oscillation of the photorefractive gain concerned with the ac field is studied experimentally, and self-generation of time subharmonics is reported. © 2007 Optical Society of America

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1. INTRODUCTION

Photorefractive semiconductors (CdTe, GaAs, InP, and others) are considered the best candidates for many applications due to high sensitivity in the near infrared and fast response. The distinctive feature of the photorefractive effect in semiconductor crystals is the bipolar charge transport, which involves electrons and holes in the space-charge formation. The bipolar charge transport results in many new photorefractive phenomena, which are not inherent in the crystals with monopolar conductivity. In particular, it has been shown that temperature or optical control of the ratio between electron and hole ionization rates enables strong two-wave mixing (TWM) gain enhancement, when a dc field is applied to the crystal [1–4]. This effect known as electron–hole resonance occurs when the photorefractive grating recording meets two requirements [3]. First, one type of carrier, electrons or holes, is dominant for the recording light photoexcitation; second, the total excitation of electrons has to be exactly balanced by the hole excitation. These requirements can be met when unbalance of the recording light excitation is compensated by the thermal generation [2,3] or photogeneration when the crystal is illuminated by the auxiliary light at a different wavelength [4]. The electron–hole resonance allows that the photorefractive grating shifted by $\pi/2$ with respect to the interference pattern is enhanced giving rise to the maximum TWM gain, when the dc field is applied to the crystal.

In some cases an impediment of the dc-field approach is space-charge effects that result in the build up of a high electric field under negative electrode and reduce the field in the crystal volume [5–7]. These phenomena caused by a nonohmic electrode-crystal junction have been observed with indium–tin–oxide (ITO), silver-paint, and copper–indium–gold/CdTe contacts [6]. The use of this screening effect for two-dimensional spatial light modulation and one-dimensional optically controlled switching array based on a CdTe crystal has been proposed [6]. Although the clear evidence of the importance of the contact behavior for dc-field enhancement of the TWM gain in this crystal has not been published. The ac-field technique requires the ac-field period to be much shorter than the photorefractive grating formation time, which for high speed photorefractive CdTe imposes some difficult requirements on the ac frequency [5]. A TWM gain of more than $10\,\text{cm}^{-1}$ was reported with a $23\,\text{kV/cm}$ square-shaped 230 kHz ac electric field. Unfortunately, so high an ac frequency makes this method unpractical in many cases. Several authors have reported that the ac-field frequency dependence of the gain reveals a relatively low-frequency region (10–100 Hz) in which gain has a pronounced maximum approaching the value measured with a high-frequency field [8–10]. The authors of [9] explained this experimental result by considering a two-band two-level model of the photorefractive effect.

In this paper effects of auxiliary incoherent illumination of the crystal at different wavelengths on the electron–hole competition are studied in a CdTe:Ge sample. We demonstrate experimentally a TWM gain enhancement by auxiliary illumination in the presence of a low-frequency ac field. We explain the experimental results by a one-level two-band model, which includes electron–hole effects, supplemented by the model of two-band adsorption, in which one band is responsible for free electrons and other for free holes photogeneration. The study of the TWM gain time modulation reveals that the frequency spectrum of the modulation contains harmonics and subharmonics of the ac-field frequency. The conditions that allow the observation of the temporal subharmonics are discussed.

2. EXPERIMENT

In our experiment we measured the effect of auxiliary incoherent illumination on the TWM gain in a CdTe:Ge crystal. A schematic of the experimental setup is shown in Fig. 1. Two unexpanded coherent beams from an He–Ne laser ($\lambda = 1150$ nm) enter the crystal through the front face. These beams record the photorefractive grating that causes the energy transfer between the beams. The total recording light intensity is 12 mW/cm^2. An auxiliary incoherent light from the monochromator illuminates the crystal through the top side. The monochromator allows the wavelength scanning in the range of 600–1400 nm and provides the illumination with the maximum intensity of approximately 4 mW/cm^2 and spectrum width of 40 nm. The relatively broad spectrum width is selected to increase the intensity of illumination. The CdTe:Ge crystal used in our experiments was grown by the Bridgman technique in the Chernovtsy State University (Ukraine) by Zakharuk and Rarenko. The dimensions of the sample are 4 mm × 5 mm × 10 mm along the crystallographic directions $[11\bar{2}]$, [111], and $[1\bar{1}0]$, respectively.

Figure 2 shows the dependence of TWM gain on the auxiliary illumination wavelength for different intensities of the auxiliary light. For the gain measurement, a 500 Hz bipolar square-shaped electric field with amplitude $E = 5.6$ kV/cm^2 is applied along the [111] crystal axis using silver-paint electrodes, which are deposited on lateral sides (4 mm × 10 mm) of the sample. Two coherent beams incident upon the $(1\bar{1}0)$ face of the crystal produce interference fringes with approximately 30 μm spacing. As discussed below in detail, in our experiments the period of ac field is longer than the crystal response time that results in an oscillation of the amplified beam intensity following the switching of the applied field polarity. In measurements of the photorefractive gain we record experimentally the time dependence of the signal beam intensity using a digital oscilloscope and then calculate its temporal mean value I_S. The TWM gain is calculated as $g = I_S / I_{S0}$, where I_{S0} is the intensity of the transmitted signal wave in the absence of the pump beam. The ratio $\beta = I_{P0}/I_{S0}$ of input intensities of the pump beam I_{P0} to the signal beam I_{S0} is 145. Our experiments show that auxiliary illumination at a wavelength shorter than 700 nm and longer than 1300 nm does not noticeably affect the gain. Thus the gain at these wavelengths can be used as a reference value to estimate the gain enhancement by auxiliary light.

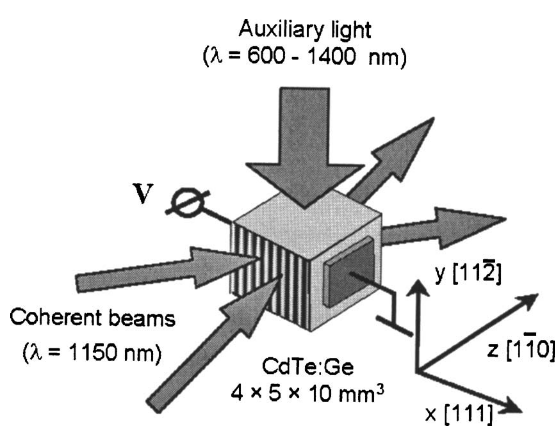

Fig. 1. Experimental setup.

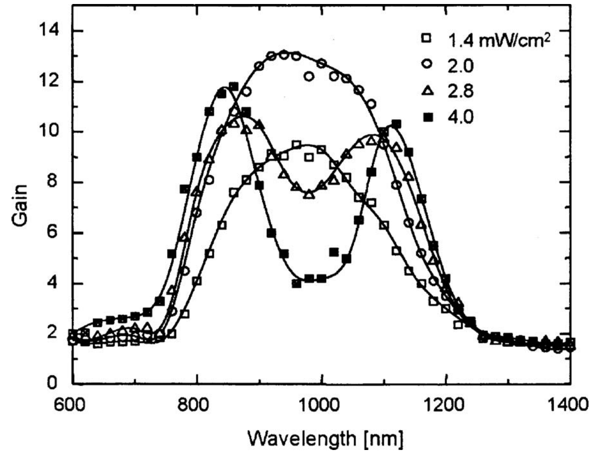

Fig. 2. Gain dependence on auxiliary light wavelength for different intensities of auxiliary illumination.

When the intensity of auxiliary illumination is low ($I_A < 2$ mW/cm^2), the spectral dependence of the gain is a one-peak curve with maximum enhancement of the gain at $\lambda_A \approx 1000$ nm as shown in Fig. 2. The gain at this wavelength decreases when the intensity of auxiliary illumination is $I_A > 2$ mW/cm^2, which yields two-peak curves at higher intensities. The peak separation increases with the intensity I_A, while their widths diminish. Considering a relatively broad spectrum of auxiliary light, one can conclude that real widths of the gain peaks are narrower than they appear in Fig. 2.

3. THEORY AND DISCUSSION

Our experimental results show that auxiliary homogeneous illumination with adequately selected wavelength noticeably enhances the photorefractive gain. The experiments were conducted by applying a 500 Hz ac field. The ac-field method requires the period of the field to be much shorter than the grating formation time. For conditions of our experiment, we calculated the crystal response time of 0.7 ms that requires an ac-field frequency well above 1.5 kHz. Thus an important requirement of ac-field enhancement was not fulfilled that, in particular, resulted in strong time modulation of the signal beam that will be discussed below. To the best of our knowledge, the theoretical model that describes the formation of the space-charge field in a photorefractive crystal with an ac field of arbitrary frequency was developed only for the materials with only one type of carrier [11]. This model cannot be used to explain our experimental results. The theory developed for the ac field, which considered the electron–hole competition, cannot be used in our case since it predicts that any uniform illumination additional to the coherent recording light decreases the space-charge field amplitude due to contrast decrease keeping optimum $\pi/2$-phase shift between the photorefractive grating and interference pattern [12]. Here we show that our experimental results can be explained using the two-band one-level model of the photorefractive crystal with a dc electric field, which includes both electron and hole optical generation. We will show that switching of the field polarity does not affect the space-charge field when the condition of the electron–hole resonance is fulfilled. In this case the dc-field model should yield an exact solution. Moreover comparison of the theoretical and experimental results allows considering the dc-field model as a satisfactory approximation in experimental conditions at any intensity and wavelength of auxiliary illumination. To support this approach we have conducted the experiments with a dc field and auxiliary illumination that yielded the shape of TWM gain dependence on the auxiliary illumination wavelength similar to the curves presented in Fig. 2. The maximum gain measured with the dc field was approximately five times smaller as compared with the results of the ac-field measurements. We attribute this gain diminution to the external field screening associated with the contact-induced phenomenon similar to what has been observed in [6]. In our modeling we consider the low-frequency ac field as the dc field with switching polarity. The polarity switching eliminates or considerably decreases the screening effects.

To calculate the photorefractive gain we suppose that the crystal is illuminated simultaneously by the recording light with sinusoidal intensity, $I_R(x)=I_{R0}(1+m\sin Kx)$ at the 1150 nm wavelength, and by the auxiliary light with homogeneous intensity I_A at variable wavelength. For the case of $|m|\ll 1$, the solution of the photorefractive equations for the steady-state space-charge field is given by [2,3]

$$E_{SC}=im\frac{G_{Rn}-G_{Rp}}{G_n\left(\frac{1}{E_q}+\frac{1}{E_{Dn}-iE_0}\right)+G_p\left(\frac{1}{E_q}+\frac{1}{E_{Dp}+iE_0}\right)},\tag{1}$$

with

$$E_q=\frac{e}{\varepsilon K}\frac{N_nN_p}{N_n+N_p},\quad E_{Dn}=E_{Mn}+E_D,\quad E_{Dp}=E_{Mp}+E_D,$$

$$E_D=K\frac{k_BT}{e},\quad E_{Mn}=\frac{\gamma_nN_p}{\mu_nK},\quad E_{Mp}=\frac{\gamma_pN_n}{\mu_pK}.\tag{2}$$

Here n and p stand for electron and holes, N is the concentration of traps containing electrons or holes, μ is the mobility, γ is the recombination coefficient, G is the total rate of free electron or hole photogeneration by the recording and auxiliary light, which also can include thermal excitation, G_R is the space average generation of the free carriers by recording light. Equation (1) is the simplified version of the expression for the space-charge field obtained in [2,3], which is valid when a sufficiently strong external field is applied so that $E_0\gg E_{Mn,p}$.

The spectroscopic study of CdTe:Ge has revealed four absorption bands at ~920, 1020, 1130, and 1320 nm [13–15]. It was found that 920 and 1020 nm bands are responsible for the generation of free electrons, while holes are dominant photocarriers when the crystal is illuminated at a longer wavelength. For our qualitative analysis we approximate these results by two absorption Gaussian bands given by

$$\alpha_n(\lambda)=\alpha_{n0}\exp[-(\lambda-\lambda_n)^2/\Delta\lambda_n^2],$$

$$\alpha_p(\lambda)=\alpha_{p0}\exp[-(\lambda-\lambda_p)^2/\Delta\lambda_p^2],\tag{3}$$

where $\alpha_n(\lambda)$ and $\alpha_p(\lambda)$ account for the emission of free electrons and holes, respectively. The total absorption, which results in free charge photogeneration, is $\alpha(\lambda)=\alpha_n(\lambda)+\alpha_p(\lambda)$. Two absorption bands have the same spectral widths $\Delta\lambda_n=\Delta\lambda_p=200$ nm, different central wavelengths, $\lambda_n=1000$ nm and $\lambda_p=1250$ nm, and the same maximum absorption $\alpha_{0n}=\alpha_{0p}=1\ \mathrm{cm}^{-1}$. According to our model the light with a wavelength of $\lambda=1125$ nm generates the same number of electrons and holes. This wavelength is marked by the dashed line in Fig. 3. The holes slightly dominate when the crystal is illuminated by the recording coherent light at $\lambda_R=1150$ nm, which we use in our experiments. The electron and hole photogeneration rates can be presented as

$$G_{n,p}=\frac{1}{2\pi\hbar c}[\alpha_{n,p}(\lambda_A)\lambda_AI_A+\alpha_{n,p}(\lambda_R)\lambda_RI_{R0}],\tag{4}$$

where the thermal excitation is neglected; λ_A and λ_R are the wavelengths of the auxiliary and recording light, respectively.

Figure 4 shows the TWM gain, which was calculated using Eqs. (1), (3), and (4) as

$$g=\frac{1+\beta}{1+\beta\exp(-\Gamma L)},$$

with

$$\Gamma=\frac{4\pi n^3r_{41}\,\mathrm{Im}(E_{SC})}{\sqrt{3}\lambda_Rm},\tag{5}$$

where β is the input beam ratio, $\beta=145$, L is the crystal thickness, n is the refractive index, r_{41} is the electro-optic coefficient, $\mathrm{Im}(E_{SC})$ is the imaginary part of the space-charge field complex amplitude. For the gain calculation, the crystal parameters were taken so as to correspond

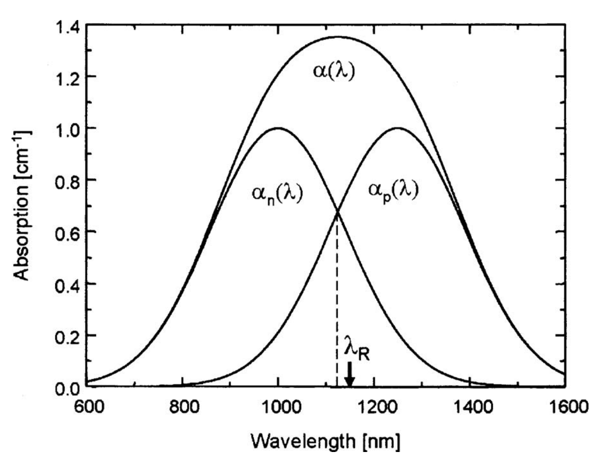

Fig. 3. Spectral dependence of the total absorption $\alpha(\lambda)$. Light absorption in the bands $\alpha_n(\lambda)$ and $\alpha_p(\lambda)$ results in electrons and holes generation, respectively.

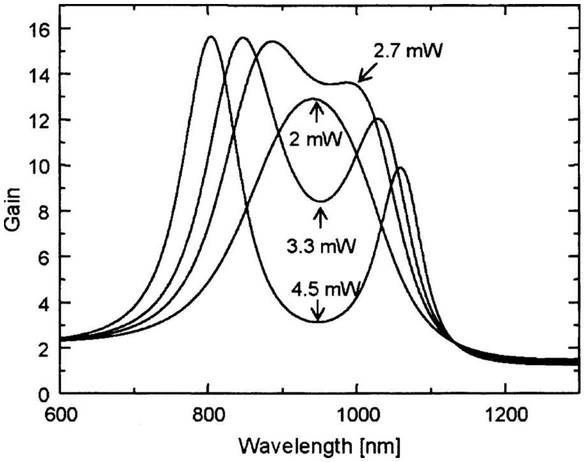

Fig. 4. Calculated gain dependences on auxiliary light wavelength for different intensities of auxiliary illumination.

roughly to values quoted in the literature for CdTe:Ge [13,14] and for conditions of our experiment, namely, $N_n = N_p = 10^{15}\,\text{cm}^3$, $\mu_n = 500\,\text{cm}^2\,\text{V}^{-1}\,\text{s}^{-1}$, $\mu_p = 0.2\mu_n$, $r_{41} = 4.5\,\text{pm/V}$, $\gamma_n = \gamma_p = 10^{-8}\,\text{cm}^3\,\text{s}^{-1}$, $E_0 = 5.6\,\text{kV/cm}$, $I_{R0} = 12\,\text{mW/cm}^2$, and $L = 10\,\text{mm}$. The results of the gain calculation shown in Fig. 4 are in good agreement with the experimental data presented in Fig. 2. Note that the calculation was done for the dc external field, while the experiment was conducted with a 500 Hz ac field.

In our experiments with the low-frequency ac field the intensity of the amplified beam has noticeable oscillation. This occurs as a result of difference between the phases of the gratings recorded under positive and negative external fields. A switching of the field polarity leads to grating rerecording, and consequently results in the time modulation of the photorefractive gain. Figure 5 shows a phase shift between the interference pattern and the photorefractive grating, which was calculated using the same parameters as for the gain calculation presented in Fig. 4. The curve calculated for positive and negative fields has the common points, when the intensity of auxiliary light is sufficiently high. At these points the grating phase does not depend on the applied field polarity and therefore the switching of the field at the first approximation does not perturb the process of space-charge field formation. Comparing Figs. 4 and 5 one can see that the gain has maxima at the wavelengths, which provide the same $\pi/2$-shift for the grating recorded under positive and negative fields. At these wavelengths the condition of the electron–hole resonance is met, the total generation of electrons is balanced by the holes generation, $G_n = G_p$. When the thermal generation of free carriers is negligible, from Eq. (4) the ratio between the intensities of the auxiliary and recording light, which meets the requirement for electro–hole resonance, is given by

$$R = \frac{I_A}{I_R} = \frac{\alpha_n(\lambda_R) - \alpha_p(\lambda_R)}{\alpha_p(\lambda_A) - \alpha_n(\lambda_A)}\frac{\lambda_A}{\lambda_R}. \qquad (6)$$

Curve 1 in Fig. 6 shows the dependence of R on the wavelength of auxiliary light λ_A calculated using Eq. (6) with $\lambda_R = 1150\,\text{nm}$, which allows us to explain the shape of the curves in Figs. 2 and 4. Indeed the effect of the gain enhancement due to electron–hole competition is resonant in intensity with a too high intensity, the enhancement is reduced. This explains the dip in the wavelength curves and also the increase in the distance between the peaks at larger auxiliary light intensity. The photoconductivity is lower when the wavelength is moved away from the optimum wavelength $\lambda_A = 975\,\text{nm}$, thus a higher intensity is required to reach the resonance. In compliance with our model, the auxiliary light cannot enhance the gain, when $\lambda_A > 1125\,\text{nm}$ and so $R < 0$. Holes are dominant photocarriers at these wavelengths, the same as for recording light, which makes the electron–hole resonance stimulation by auxiliary light impossible. Note that this is valid only when the thermal generation of free carriers is negligible. Curve 2 in Fig. 6 is calculated assuming the thermal generation of electrons with a rate of $2\times10^{16}\,\text{cm}^{-3}\,\text{s}^{-1}$ and the recording light intensity $I_R = 12\,\text{mW}$. Obviously, this curve predicts the enhancement effect in a wider wavelength range. We suppose that ther-

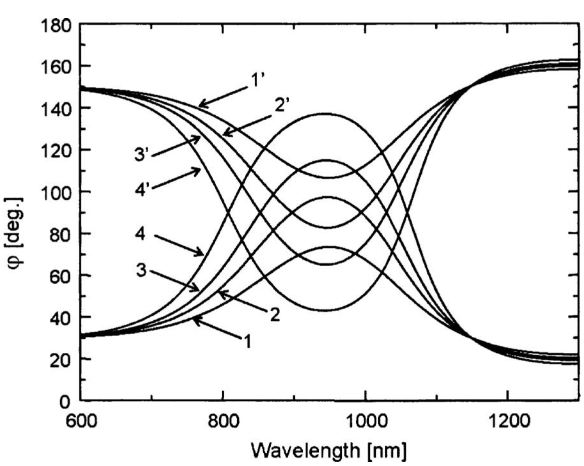

Fig. 5. Theoretical dependencies of the photorefractive grating phase on the wavelength of auxiliary illumination. 1 and 1′, $I(A) = 4\,\text{mW/cm}^2$; 2 and 2′, $I(A) = 5.5$; 3 and 3′, $I(A) = 6.5$; 4 and 4′, $I(A) = 9.0$. Curves 1, 2, 3, and 4 are calculated for positive applied dc field $E_0 = 5.6\,\text{kV/cm}$; curves 1′, 2′, 3′, and 4′ are calculated for negative field $E_0 = -5.6\,\text{kV/cm}$.

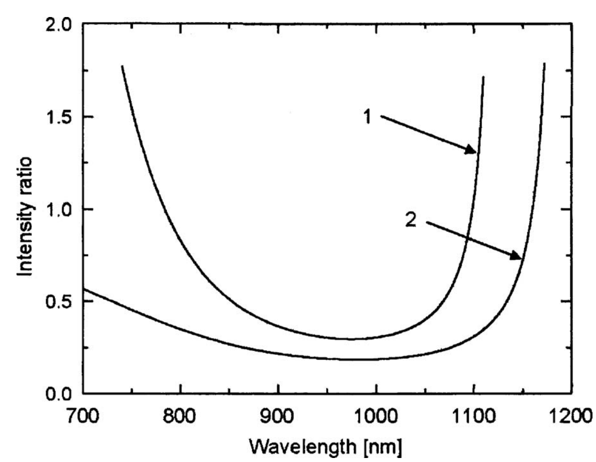

Fig. 6. Intensity ratio between the auxiliary and recording light intensities that yields the electron–hole resonance as a function of auxiliary light wavelength. Curve 1 is calculated without taking into account the thermal generation of free carriers using Eq. (6), while curve 2 is calculated assuming the thermal generation of electrons with a rate of $2\times10^{16}\,\text{cm}^{-3}\,\text{s}^{-1}$.

mal generation is responsible for some minor discrepancy between calculated and experimental curves. Unfortunately, for experimental investigation of the combined effect of thermal generation and auxiliary light one needs a tunable light source for auxiliary illumination with narrower spectrum than was at our disposal.

4. SUBHARMONICS OF THE GAIN OSCILLATION

We found experimentally that amplitude and frequency of the amplified beam oscillation depend on the intensity and wavelength of auxiliary illumination. Typical time dependences of the intensity are presented in Fig. 7. Here we show the signal beam intensity deviation from the mean level for different intensities of the auxiliary illumination together with the applied voltage. For convenience of curve comparison, the amplitudes of the intensity deviations were normalized. In this experiment the square-wave applied field has the frequency $f_0=500$ and amplitude $E_0=5.6\,\text{kV/cm}$ calculated as the applied voltage divided by the distance between the electrodes. At the spectrum of oscillation (Fig. 8) we found at least four harmonics and three subharmonics of fundamental frequency f_0. Taking this result into account, the time dependence of the gain can be given by

$$g=g_0\left[1+\sum_{k=1}^{8} c_{k/2}\sin\left(\frac{2\pi k f_0 t}{2}+\varphi_{k/2}\right)\right], \qquad (7)$$

where g_0 is the temporal average gain, $c_{k/2}$ and $\varphi_{k/2}$ are the modulation coefficient and phase of respective harmonic or subharmonic. Note that the experimental gain presented in Fig. 2 is calculated also as the temporal mean value, which corresponds to g_0. The measured gain g_0 for different auxiliary light intensities and $\lambda_A=1000\,\text{nm}$ is shown in Fig. 9 along with modulation coefficients for the first and second harmonic, c_1 and c_2. The modulation coefficients were calculated as the ratios between the amplitudes of the gain modulations at corresponding frequencies and the mean gain g_0. The mean gain has maximum, when $I_A \cong 3.2\,\text{mW/cm}^2$. At this intensity the first harmonic has a value close to zero, while the second harmonic reaches its maximum. As our theoretical analysis has shown, the electron–hole resonance takes place at this intensity of auxiliary light, and the grating has the same $\pi/2$-phase shift when the positive as the well as the negative fields are applied to the crystal. The zero value of the fundamental harmonic observed experimentally at this intensity means that the gain dynamic is the same at the positive and negative half-cycle of the applied field, which indirectly confirms the correctness of our analysis.

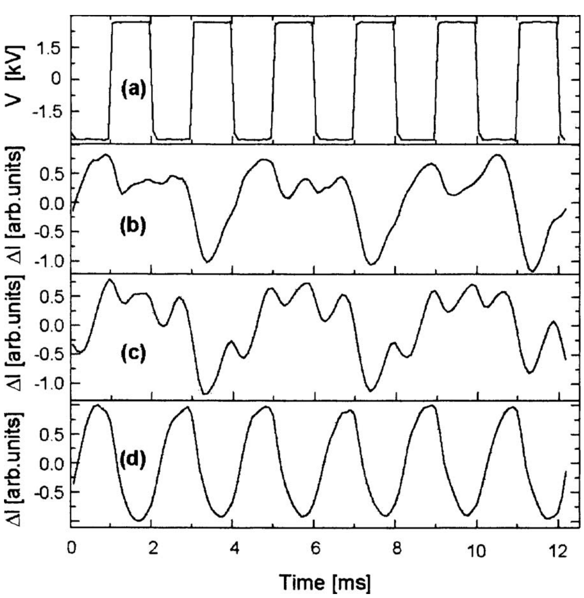

Fig. 7. Time dependences of the applied voltage and temporal modulation of the amplified beam intensity recorded with different intensities of the auxiliary light. (a) Applied voltage; (b) modulation of intensity, $I_A=2.6\,\text{mW/cm}^2$; (c) $I_A=3.2\,\text{mW/cm}^2$; (d) $I_A=4.0\,\text{mW/cm}^2$.

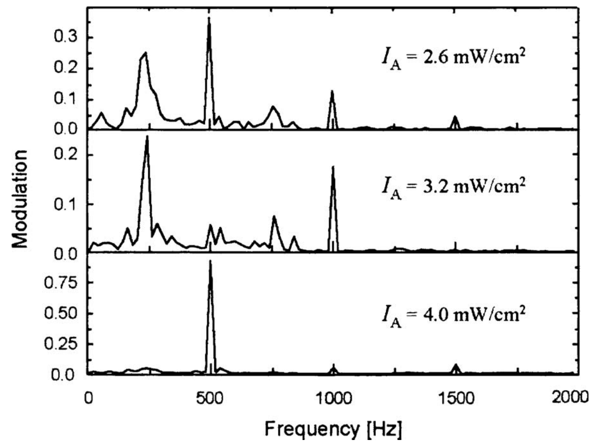

Fig. 8. Time-frequency spectra of the TWM gain modulation for different intensities of the auxiliary light I_A.

The gain modulation spectra are shown in Fig. 8, where the zero-component, which corresponds to g_0, is suppressed to emphasize harmonics and subharmonics. The subharmonics $f_0/2$ and $3f_0/2$ are strong, when the intensity $I_A<3.2\,\text{mW/cm}^2$. As shown by theoretical analysis, at these intensities the holes generated by the recording light dominate in photocurrent. When auxiliary illumination is stronger and dominant carriers in photocurrent are electrons generated by the uniform incoherent light,

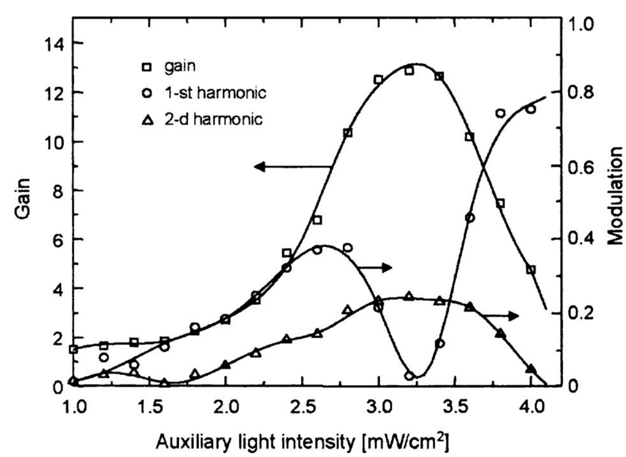

Fig. 9. Temporal average gain g_0 and modulation coefficients of the first and second harmonics as a function of auxiliary light wavelength.

the gain modulation is almost sinusoidal with the fundamental frequency f_0. To the best of our knowledge, there is no adequate theoretical description of the appearance of time subharmonics of the photorefractive gain. We can only speculate that the physical mechanism of this phenomenon should be a time domain analog to what has been proposed for the space subharmonic generation [12,16,17].

5. CONCLUSIONS

We have shown that incoherent illumination is an effective and easy implemented method for electron–hole resonance control in CdTe, which allows strong enhancement of the photorefractive response in the presence of a low-frequency ac field. Our experiments show more than a tenfold increase of the two-wave mixing (TWM) gain as a result of the auxiliary illumination. The enhancement has been characterized as a function of the wavelength and intensity of the auxiliary light. We have proposed the simple model of two-band absorption that explains qualitatively the rather complicated spectrum and intensity dependences of the TWM gain on auxiliary light. Probably, the same simple model should be used for qualitative analysis of spectral dependences of the photorefractive effect in other semiconductors with bipolar photoconductivity. We have revealed experimentally that the low frequency ac field, which has a period longer than the response time of the crystal, results in the photorefractive gain modulation with complicated time-frequency spectrum. This spectrum contains harmonics and subharmonics of the ac-field frequency. The amplitudes of the modulation spectrum components depend on the balance between the rates of electrons and holes photogeneration. The fundamental harmonic of the modulation disappear when the electron–hole resonance takes place. We believe that a further study of time harmonics and subharmonics generation should result in deeper insides into the mechanism of the photorefractive nonlinearity of semiconductors.

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