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EFFECT OF LIGHT-INDUCED SCATTERING ON DIFFRACTION EFFICIENCY IN PHOTOREFRACTIVE CRYSTALS*

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The coupled wave equations and their analytic solutions in photorefractive crystals are presented for the case of two-beam writing, while the light-induced scattering is considered. Using this result, the diffraction efficiency for each writing beam is discussed in doped KNSBN crystals with unidirectional fan, which is explained theoretically and verified experimentally.

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I. INTRODUCTION

Light-induced scattering is the result of the energy exchange among the input beam and the beams scattered randomly by the defects in crystals. Due to the interactions among the scattered beams and the input beam, complex refractive phase gratings are formed, which strengthen the scattered beams and weaken the input beam, and the strengthened beams will be scattered again by other defects in the crystals. And this process will repeat again and again. Because this energy transfer is not isotropic, the scattered beams can be strengthened only in a specified angular range.[1,2] As for the spatial distribution of the scattered beams, there are two kinds of light-induced scattering in photorefractive crystals: one is of unidirectional fan,[3,4] spread along the orientation of optical axis of the crystal (e.g. $BaTiO_3$) or counter to this orientation (e.g. KNSBN). Another is of bidirectional fan, which distributes symmetrically along the $+c$ and $-c$ directions of the crystals (e.g. $LiNbO_3$). We have observed that the light-induced scattering has different influences on the recording of holographic gratings for these two kinds of photorefractive crystals. Due to the asymmetric fan of the first kind of crystals, the scattering has different effects on the two-wave mixing and on the diffraction efficiency of each writing beam. However, the symmetric fan of the second kind of crystals does not affect the symmetry of energy transfer in two-wave coupling

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and the diffraction efficiency of each beam. Only the measured values are slightly less than the theoretical ones.[5] So far, the investigation on the diffraction efficiencies of the two writing beams in photorefractive two-wave mixing has not involved the effect of the light-induced scattering. Therefore, it is considered that the diffraction efficiencies of two writing beams are identical.[6]

In this paper, the coupled wave equations in photorefractive crystals are derived for the case of two-beam writing when the light-induced scattering is taken into account. Using these equations, the diffraction efficiency is discussed for each writing beam in doped KNSBN crystals. It is concluded that the diffraction efficiencies of the two writing beams are unequal in photorefractive crystals with unidirectional fan. The result is also verified experimentally.

II. THE COUPLED WAVE EQUATIONS INVOLVING THE EFFECT OF LIGHT-INDUCED SCATTERING

The schematic diagram of the two-wave mixing involving light-induced scattering is given in Fig.1 for a uniaxial crystal.

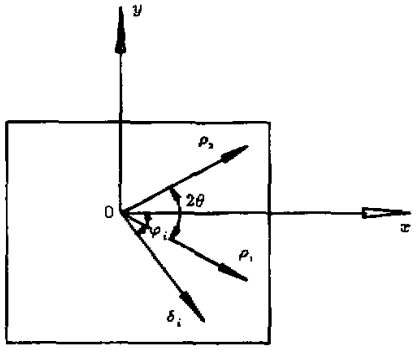

Fig. 1. Schematic diagram of two-beam mixing involving light-induced scattering.

Two extraordinarily-polarized beams, R_1 and R_2, are incident on the crystal. The wave vectors in the crystal, $\boldsymbol{\rho}_1$ and $\boldsymbol{\rho}_2$, make equal angles of θ with the x axis in xoy plane. The polarization direction vector is $\boldsymbol{r}_0=(0, 0, 1)$. Considering the scattering of the extraordinarily-polarized beam alone, we see that the wave vector $\boldsymbol{\delta}_i$ of the scattered light $S_i(x)$ is at an angle ϕ_i with the x-axis in xoy plane and the polarization direction vector $\boldsymbol{r}_0=(0, 0, 1)$ is normal to the c-axis.

The optical fields can be expressed as

$$\boldsymbol{E}_1 = R_1(x)\exp[-j(\boldsymbol{\rho}_1 \cdot \boldsymbol{r})]\boldsymbol{r}_0 , \tag{1}$$

$$\boldsymbol{E}_2 = R_2(x)\exp[-j(\boldsymbol{\rho}_2 \cdot \boldsymbol{r})]\boldsymbol{r}_0 , \tag{2}$$

$$\boldsymbol{E}_i = S_i(x)\exp[-j(\boldsymbol{\delta}_i \cdot \boldsymbol{r})]\boldsymbol{r}_0 \ , \tag{3}$$

For the extraordinary beam the refractive index is n_e. The relations among the wave vectors are given by $\rho_1 = \rho_2 = \delta_i = kn_e$. We assume that the wave amplitude is a function of x only. The total electric field in the overlapped region in the crystal is

$$\boldsymbol{E} = \boldsymbol{E}_1 + \boldsymbol{E}_2 + \sum_{i=1} \boldsymbol{E}_i \ . \tag{4}$$

The light intensity is expressed as

$$I = \boldsymbol{E} \cdot \boldsymbol{E}^* \ .$$

When the interaction among the scattered beams is neglected, we have

$$I = I_0\left[1 + \frac{2\sqrt{I_1 I_2}}{I_0}\cos(\boldsymbol{k}_1 \cdot \boldsymbol{r}) + \sum_{i=1}\frac{2\sqrt{I_1 I_i}}{I_0}\cos(\boldsymbol{k}_{2i} \cdot \boldsymbol{r}) + \sum_{i=1}\frac{2\sqrt{I_2 I_i}}{I_0}\cos(\boldsymbol{k}_{3i} \cdot \boldsymbol{r})\right] \ , \tag{5}$$

where $I_0 = I_1 + I_2 + \sum_{i=1} I_i$, $I_1 = \boldsymbol{E}_1 \cdot \boldsymbol{E}_1^*$, $I_2 = \boldsymbol{E}_2 \cdot \boldsymbol{E}_2^*$, $I_i = \boldsymbol{E}_i \cdot \boldsymbol{E}_i^*$.

$$\boldsymbol{k}_1 = \boldsymbol{\rho}_1 - \boldsymbol{\rho}_2 \ , \quad \boldsymbol{k}_{2i} = \boldsymbol{\rho}_1 - \boldsymbol{\delta}_i \ , \quad \boldsymbol{k}_{3i} = \boldsymbol{\rho}_2 - \boldsymbol{\delta}_i \ . \tag{6}$$

The change of refractive index resulting from the change of intensity distribution caused by the photorefractive effect in the crystal is given by

$$\Delta n = n_1\left\{\frac{2\sqrt{I_1 I_2}}{I_0}\cos(k_1 \cdot r + \phi) + \sum_{i=1}\frac{2\sqrt{I_1 I_i}}{I_0}\cos(k_{2i} \cdot r + \phi) + \sum_{i=1}\frac{2\sqrt{I_2 I_i}}{I_0}\cos(k_{3i} r + \phi)\right\} \ , \tag{7}$$

where n_1 is a proportionality constant, ϕ the phase shift between the refractive index phase grating and the interference pattern. The corresponding change of the relative dielectric tensor is then

$$\delta\varepsilon = \delta\varepsilon_1\cos(\boldsymbol{k}_1 \cdot \boldsymbol{r} + \phi) + \sum_{i=1}\delta\varepsilon_{2i}\cos(\boldsymbol{k}_{2i} \cdot \boldsymbol{r} + \phi) + \sum_{i=1}\delta\varepsilon_{3i}\cos(\boldsymbol{k}_{3i}\boldsymbol{r} + \phi) \ , \tag{8}$$

where $\delta\varepsilon_1 = \dfrac{4n_e n_1 k(I_1 I_2)^{1/2}}{I_0}$, $\delta\varepsilon_{2i} = \dfrac{4n_e n_1 k(I_1 I_i)^{1/2}}{I_0}$, $\delta\varepsilon_{3i} = \dfrac{4n_e n_1 k(I_2 I_i)^{1/2}}{I_0}$. The vector wave equation in the anisotropic photorefractive crystals is given by

$$\nabla^2\boldsymbol{E} - \nabla(\nabla \cdot \boldsymbol{E}) + \tilde{k}\boldsymbol{E} = 0 \ , \tag{9}$$

where the complex wave vector $\tilde{k}$ is written in the form

$$\tilde{k} = \left(\frac{\omega}{c}\right)^2 \mu_r\tilde{\varepsilon} + \mathrm{i}\omega\mu\sigma = k^2\mu_r\tilde{\varepsilon} + \mathrm{i}\omega\mu\sigma \ , \tag{10}$$

$$\tilde{\varepsilon} = \varepsilon_0 + \delta\varepsilon \ . \tag{11}$$

For non-magnetic materials, the relative permeability μ_r=1, the conductivity σ is assumed to be unmodulated. Substituting relations (4), (8), (10) and (11) into the wave equation (9), the coupled wave equations involving the light-induced scattering can be obtained

$$\frac{\mathrm{d}R_1}{\mathrm{d}x} - \frac{\mu c\sigma}{2n_e\cos\theta}R_1 = \frac{k\sin\phi}{4n_e\cos\theta}\left(r_0\delta\varepsilon_1 r_0 R_2 - \sum_{i=1} r_0\delta\varepsilon_{2i} r_0 S_i\right), \tag{12}$$

$$\frac{\mathrm{d}R_2}{\mathrm{d}x} - \frac{\mu c\sigma}{2n_e\cos\theta}R_2 = \frac{k\sin\phi}{4n_e\cos\theta}\left(r_0\delta\varepsilon_1 r_0 R_1 + \sum_{i=1} r_0\delta\varepsilon_{3i} r_0 S_i\right), \tag{13}$$

$$\frac{\mathrm{d}S_i}{\mathrm{d}x} - \frac{\mu c\sigma}{2n_e\cos\phi_i}S_i = \frac{k\sin\phi}{4n_e\cos\phi_i}\left(r_0\delta\varepsilon_{2i} r_0 R_1 + \sum_{i=1} r_0\delta\varepsilon_{3i} r_0 R_2\right), \tag{14}$$

using the phase match condition (6) and slow-varying amplitude approximation.

Substituting $I_1 = |R_1|^2$, $I_2 = |R_2|^2$, $I_i = |S_i|^2$ into Eqs.(12)-(14), we obtain:

$$\frac{\mathrm{d}I_1}{\mathrm{d}x} + \alpha_1 I_1 = \frac{\Gamma_1 I_1}{I_0}[I_2 - \sum_{i=1} I_i], \tag{15}$$

$$\frac{\mathrm{d}I_2}{\mathrm{d}x} + \alpha_1 I_2 = \frac{\Gamma_1 I_2}{I_0}[I_1 + \sum_{i=1} I_i], \tag{16}$$

$$\frac{\mathrm{d}I_i}{\mathrm{d}x} + \alpha_2 I_i = \frac{\Gamma_{2i} I_i}{I_0}[I_1 + I_2], \tag{17}$$

where $\Gamma_1 = \dfrac{2kn_e\sin\phi}{\cos\theta}$, $\Gamma_{2i} = \dfrac{2kn_e\sin\phi}{\cos\phi_i}$,

$\alpha_1 = \dfrac{\mu c\sigma}{n_e\cos\theta}$, $\alpha_2 = \dfrac{\mu c\sigma}{n_e\cos\phi_i}$.

Except for a few special cases, Eqs.(15)-(17) are insolvable analytically, and one has to rely on approximate solutions. Since there is a maximum of scattered beams distributed over the angular range, so we consider only the maximum scattered beam I_j in the direction of ϕ_j. After neglecting the absorption in crystals, a set of coupled wave equations can be obtained:

$$\frac{\mathrm{d}I_1}{\mathrm{d}x} = \Gamma_1\frac{I_1(I_2 - I_j)}{I_0}, \tag{18}$$

$$\frac{\mathrm{d}I_2}{\mathrm{d}x} = \Gamma_1\frac{I_2(I_1 + I_j)}{I_0}, \tag{19}$$

$$\frac{\mathrm{d}I_j}{\mathrm{d}x} = \Gamma_{2j}\frac{I_j(I_1 + I_2)}{I_0}. \tag{20}$$

The analytic solutions of Eqs.(18)-(20) are derived as

$$I_1 = \frac{I_0 m_0\exp(\Gamma_1 x)}{1+\beta_1\exp(\Gamma_1 x)}\cdot\left(\frac{1+\beta_2}{1+\beta_2\exp(\Gamma_{2j}x)}\right)^{\frac{\Gamma_1}{\Gamma_{2j}}}, \tag{21}$$

$$I_2 = \frac{I_0}{1+\beta_1\exp(\Gamma_1 x)}, \tag{22}$$

$$I_j = \frac{I_0\beta_2 \exp(\Gamma_{2j}x)}{1+\beta_2 \exp(\Gamma_{2j}x)} , \tag{23}$$

where $\beta_1 = \frac{I_{10}+I_{j0}}{I_{20}}$, $\beta_2 = \frac{I_{j0}}{I_{10}+I_{20}}$, $m_0 = \frac{I_{10}}{I_{20}}$, and I_{j0} is the initial scattered beam caused by defects and impurities in the crystal. I_{10} and I_{20} are the initial input beam intensities. I_{j0} is generally very small, and so is β_2. If the crystal is perfect, the input beam cannot be scattered in the crystal. Thus, $I_{j0} = 0$, $\beta_2 \to 0$, $\beta_1 = m_0$, and Eqs.(21)–(23) become

$$I_1 = \frac{I_0 m_0 \exp(\Gamma_1 x)}{1+m_0 \exp(\Gamma_1 x)} , \tag{24}$$

$$I_2 = \frac{I_0}{1+m_0 \exp(\Gamma_1 x)} , \tag{25}$$

$$I_j = 0 . \tag{26}$$

These are just the expressions of the intensities for two-wave mixing without the effect of the light-induced scattering.

III. DIFFRACTION EFFICIENCIES OF BOTH WRITING BEAMS IN THE DOPED KNSBN CRYSTAL

The energy transfer in two-wave coupling results from the constructive or destructive interference between the diffracted and transmitted beams after the two input beams have formed the volume index phase gratings.[7] The energy transfer direction between the two beams coincides with the direction of the fan for KNSBN crystals. Considering the strongest scattered beam I_j only, three sets of refractive index gratings are created in the crystal, which is evidenced from the relation (7). Thus the diffraction efficiencies can be defined as

$$\eta_1 = \frac{I_{12d}}{I_{10}} = \frac{(R_{12d})^2}{(R_{10})^2} , \qquad \eta_1' = \frac{I_{1jd}}{I_{10}} = \frac{(R_{1jd})^2}{(R_{10})^2} ,$$

$$\eta_2 = \frac{I_{21d}}{I_{20}} = \frac{(R_{21d})^2}{(R_{20})^2} , \qquad \eta_2' = \frac{I_{2jd}}{I_{20}} = \frac{(R_{2jd})^2}{(R_{20})^2} ,$$

where I_{12d} is the diffracted light of I_{10} to the direction of beam I_2, I_{1jd} the diffracted light of I_{10} to the direction of beam I_j, I_{21d} the diffracted light of I_{20} to the direction of beam I_{10}, and I_{2jd} is the diffracted light of I_{20} to the direction of beam I_j. Ignoring the diffraction of the scattered beam I_{j0}, the transmitted I_1 interferes constructively with the diffracted I_2, the transmitted I_2 interferes destructively with the diffracted I_1, and the diffracted light of I_1 and I_2 interferes constructively in the direction of the maximum scattered light I_j. Hence we have

$$[\sqrt{\eta_2}R_{20} + \sqrt{1-\eta_1-\eta_1'}R_{10}]^2 = R_1^2 , \tag{27}$$

$$[\sqrt{\eta_1}R_{10} + \sqrt{1-\eta_2-\eta_2'}R_{20}]^2 = R_2^2 , \tag{28}$$

$$|\sqrt{\eta_1'}R_{10} + \sqrt{\eta_2'}R_{20}|^2 = S_j^2 \ . \tag{29}$$

When $\sqrt{\eta_1}R_{10} \geq \sqrt{1-\eta_2-\eta_2'}R_{20}$, using $I_{10} = |R_{10}|^2$, $I_{20} = |R_{20}|^2$, $I_1 = |R_1|^2$, $I_2 = |R_2|^2$, $I_j = |S_j|^2$, we obtain

$$\eta_2 = \left(\frac{1-\eta_2'}{1-\eta_1'}\right)\cdot\eta_1 \ . \tag{30}$$

Substituting Eq.(30) into Eq.(28) yields

$$\eta_1 = \frac{2AB + C \pm \sqrt{4ABC + C^2}}{2B^2} \ , \tag{31}$$

where $A = I_{20} - I_2 - I_{2jd}$, $B = I_{10}\dfrac{(I_{10} - I_j)}{I_{10} - I_{1jd}}$, $C = 4I_2I_{10}$. It can be seen from Fig.1 that the included angle between the beams I_2 and I_j is much larger than that between I_1 and I_j. So the coupling between beams I_2 and I_j is much weaker than that between I_1 and I_j.[2,4] This fact can be explained as due to the energy loss of I_2 being smaller than that of I_1 when I_1 and I_2 form stable fans separately. Therefore the diffracted light of I_2 to I_j can be ignored, i.e. I_{2id}=0. The relation (31) becomes

$$\eta_1 = \frac{m_0[\exp(\Gamma_1 x/2) - 1]^2}{(1+\beta_1)[1+\beta_1\exp(\Gamma_1 x)]}\cdot\left\{1 - \frac{\Gamma_1\beta_2\exp(\Gamma_1 x/2)[\exp(\Gamma_{2j}x) - 1]}{\Gamma_{2j}[\exp(\Gamma_1 x/2) - 1]}\right\} \ , \tag{32}$$

and

$$\eta_2 = \frac{m_0[\exp(\Gamma_1 x/2) - 1]^2}{(1+\beta_1)[1+\beta_1\exp(\Gamma_1 x)]}\cdot\left\{1 + \frac{m_0+1}{m_0}\cdot\beta_2\exp(\Gamma_{2j}x) - \frac{\Gamma_1\beta_2\exp(\Gamma_1 x/2)[\exp(\Gamma_{2j}x) - 1]}{\Gamma_{2j}[\exp(\Gamma_1 x/2) - 1]}\right\} . \tag{33}$$

If the scattered beams in all directions are taken into account, the diffraction efficiencies for both writing beams can be rewritten approximately as

$$\eta_1 = \frac{m_0[\exp(\Gamma_1 x/2) - 1]^2}{(1+\beta_1)[1+\beta_1\exp(\Gamma_1 x)]}\cdot\left\{1 - \frac{\Gamma_1\beta_2\exp(\Gamma_1 x/2)}{[\exp(\Gamma_1 x/2) - 1]}\cdot\sum_{i=1}\frac{\beta_{2i}[\exp(\Gamma_{2j}x) - 1]}{\Gamma_{2i}}\right\} \ , \tag{34}$$

$$\eta_2 = \frac{m_0[\exp(\Gamma_1 x/2) - 1]^2}{(1+\beta_1)[1+\beta_1\exp(\Gamma_1 x)]}\cdot\left\{1 + \frac{m_0+1}{m_0}\cdot\sum_{i=1}[\beta_{2i}\exp(\Gamma_{2i}x)] - \frac{\Gamma_1\exp(\Gamma_1 x/2)}{[\exp(\Gamma_1 x/2) - 1]}\cdot\sum_{i=1}\frac{\beta_{2i}[\exp(\Gamma_{2i}x) - 1]}{\Gamma_{2i}}\right\} . \tag{35}$$

From Eqs.(32)–(35) we can conclude that $\eta_2 > \eta_1$, i.e. the diffraction efficiency of I_2 (which makes an acute angle with the optical axis) is larger than that of I_1 (which makes an obtuse angle with the optical axis); so the diffraction efficiency will vary as a function of the ratio of input beam intensities to their incident angles. If the effect of scattered beams is neglected, we have $\eta_1 = \eta_2$, i.e. the diffraction efficiencies of both writing beams are identical.

IV. EXPERIMENTAL RESULTS

The experimental arrangement for measuring the diffraction efficiencies of the two input beams is shown schematically in Fig.2. The photorefractive crystal used is a Ce-doped

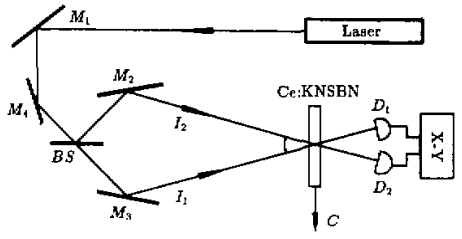

Fig. 2. Experimental arrangement for measuring the diffraction efficiencies of the two input laser beams.

KNSBN crystal with a thickness of 0.4 cm. The source is a He-Ne laser with a wavelength λ=632.8 nm. A half-wave plate is used to change the polarization of the two input beams and ensure that the beams incident on the crystal are e-polarized. Figures 3 and 4 show the diffraction efficiencies of the input beams versus the input intensity ratio at a certain incident angle and those versus the incident angle with a certain input beam ratio, respectively.

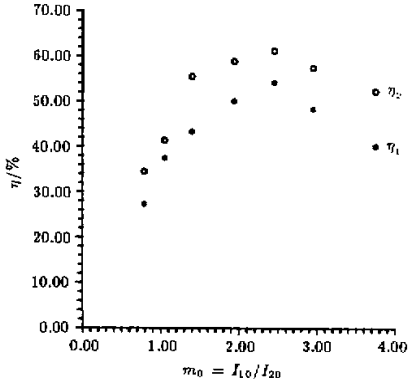

Fig. 3. Diffraction efficiency versus the input intensity ratio m at fixed angle 2θ=40°.

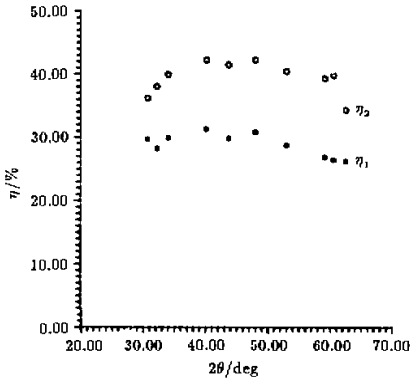

Fig. 4. Diffraction efficiency versus the input angle 2θ at fixed intensity ratio m=1.

It is seen from Figs.3 and 4 that the diffraction efficiencies of the two beams are obviously different for the Ce-doped KNSBN crystal. According to the above-mentioned theoretical analyses, the scattered beams will create two sets of photorefractive phase gratings for the two input beams in the crystal with a unidirectional fan, if the interaction among the scattered beams is ignored. The coupling between the beam I_2 at an acute angle with the optical axis and the scattered beams is weaker. But the coupling between the beam I_1 at

an obtuse angle with the optical axis is stronger. Masking I_2, the measured intensity of I_{12d} which is diffracted from I_1 is smaller than that in the ideal case in which the scattering is ignored. Hence the diffraction efficiency of I_1 is smaller than that in the ideal case. Moreover, a part of the scattered beams of I_2 may be in the direction of I_1 due to the unidirectivity of the fan. Masking I_1, the measured value of I_{21d} which is diffracted from I_2 is larger than the value obtained from diffraction on the grating formed by I_1 and I_2.

It is the two factors mentioned above that produce different diffraction efficiencies measured for the two writing beams, whose projections of wave vectors are along and counter to the orientation of the optical axis.

The diffraction efficiencies of the two beams versus the input intensity ratio are also measured for the doped $LiNbO_3$ crystal. It is observed that the diffraction efficiencies of the two input beams are almost the same.

V. CONCLUSION

The two-wave coupled equations involving light-induced scattering in photorefractive crystals are presented. We have analysed and explained the difference of the diffraction efficiencies for both of the writing beams in doped KNSBN crystals and conclude that:

1) For the crystals with unidirectional fan (e.g. KNSBN), the diffraction efficiency of the input beam, whose wave vector lies at an obtuse angle with the $+c$-axis, is larger than that of the other input beam, whose wave-vector lies at an acute angle with the $+c$-axis.

2) For the crystals with symmetric fan, the diffraction efficiencies for both of the input beams are identical.

This conclusion is favorable for the applications of photorefractive crystals in optical information processing, associate memory, etc.

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