Influence of the Bragg angle on the optimal energy exchange in the course of two-wave mixing in a $Bi₁₂SiO₂₀$ piezoelectric crystal

V V Shepelevich, A A Firsov

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Abstract. The problem of maximising the relative intensity of the signal optical wave in the course of two-wave mixing was solved taking into account the self-diffraction in a $Bi₁₂SiO₂₀$ piezoelectric crystal. The maximisation was carried out simultaneously with respect to four parameters: the polarisation (ψ) and orientation (θ) angles, the crystal thickness d , and the Bragg angle φ . It was established that there are two sets of optimal parameters for a $Bi₁₂SiO₂₀$ crystal at the wavelength of 632.8 nm and for a typical acceptor concentration of 10^{22} m⁻³: $\varphi \approx 11^{\circ}$, $\theta \approx 39.1^{\circ}$, $\psi \approx 98.85^{\circ}$, $d \approx 7.11$ mm (for the first maximum) and $\varphi \approx 11^{\circ}$, $\theta \approx 320.9^{\circ}$, $\psi \approx 54.15^{\circ}$, $d \approx 7.11$ mm (for the second maximum). The dependences of these optimal parameters on the acceptor concentration in the range $10^{21} - 4 \times$ 10^{22} m⁻³ were found. The model proposed for two-wave mixing in the special case of a fixed Bragg angle and the constant visibility approximation improved the agreement between the theoretical results and the known experimental data.

1. Introduction

Cubic photorefractive crystals of the sillenite type $[Bi₁₂SiO₂₀]$ (BSO), $Bi_{12}GeO_{20}$ (BGO), $Bi_{12}TiO_{20}$ (BTO)] are promising media for various applications in dynamic holography $[1 - 3]$. In order to reduce the output signal distortions, photorefractive crystals are frequently used in the diffuse regime $[4 - 7]$. In view of the fact that in this regime the diffraction efficiency of a holographic grating is low (of the order of 1%), maximisation of the output characteristics of holograms by selecting the interaction (mixing) geometry, the polarisation of the optical waves, and the crystal thickness is topical. This problem was initially frequently considered ignoring the piezoelectric properties of crystals (see, for example, Refs [\[8, 9\]\)](#page-4-0). Later, an allowance for these properties was made (see, for example, Refs $[10 - 15]$).

The first theoretical results concerning optimisation of the exchange of the energy of optical waves in two-wave mixing in a BSO crystal as a result of a change in the polar-isation of the 'read' optical waves were obtained in Refs [\[10\]](#page-4-0) and [\[11\],](#page-4-0) and were then confirmed experimentall[y \[12\].](#page-4-0) It was also established in Ref. [\[12\]](#page-4-0) that the optimal polarisation

V V Shepelevich, A A Firsov Mozyr State Pedagogical Institute, Studencheskaya ul. 28, 247760 Mozyr, Belarus

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azimuths of optical waves are the same for diffraction and for two-wave mixing in the orientation-angle ranges $0 - 90^{\circ}$ and $270 - 360^\circ$, whereas in the range $90 - 270^\circ$ they differ by 90° . The orientation optimisation of the energy exchange of optical waves for constant polarisation was investigated earlier [\[13\].](#page-4-0) The dependences of the optimal orientation angle on the thickness of BSO and BTO crystals in two-wave mixing were found [\[14\]](#page-4-1) and it was shown that the [\[11\]](#page-4-0) direction in the sillenite crystals is generally not optimal.

Local maxima are known $[14, 16 - 18]$ to arise in the dependences of the diffraction efficiency and of the relative intensity of the object optical wave, maximised with respect to the polarisation angle, on the crystal thickness. This makes it possible to maximise the above characteristics not only with respect to the polarisation and orientation angles, but also with respect to the crystal thickness.The polarisation and orientation angles and the thickness of a BSO crystal at the wavelength of 514.5 nm and of a BTO crystal at the wavelength of 632.8 nm, at which the relative gain in two-wave mixing is maximised simultaneously with respect to these three parameters, were found [\[19\]](#page-4-1) in the approximation postulating small coupling constants and Bragg angles.

In addition, there is also a fourth parameter with respect to which it is possible to maximise the output characteristics of holograms—the Bragg angle. An increase in this angle does not always increase monotonically the diffraction efficiency and the gain in two-wave mixing (see, for example, Refs [\[2\]](#page-4-2) and [\[20\]\)](#page-4-3). However, so far as we are aware, maximisation of the output characteristics of holograms with respect to the Bragg angle combined with maximisation with respect to the orientation and polarisation angles as well as the crystal thickness has not been carried out.

Our study is devoted to maximisation of the relative intensity of the signal optical wave simultaneously with respect to four parameters: the polarisation and orientation angles, the crystal thickness, and the Bragg angle. This problem may be solved in the constant grating approximation: it is then assumed that the field amplitude of a holographic grating is constant (see, for example, Ref. [\[12\]\)](#page-4-0) or allowance is made for the redistribution of the energy of optical waves during the writing of a hologram (selfdiffraction [\[15, 21\]\)](#page-4-4). In the latter case, account is taken of the dependence of the interference pattern visibility and hence of the field amplitude of a holographic grating on the polarisation azimuth of the `read' light and on the crystal thickness (see, for example, Refs [\[22, 23\]\).](#page-4-5)

2. Theoretical foundations

The coupled-wave equations, describing the diffraction of light in transparent photorefractive gyrotropic cubic piezoelectric crystals [\[11\]](#page-4-0) for arbitrary values of the orientation angles, coupling constants, Bragg angle, and of the ratio of the intensities of `read' optical waves will be used to solve the formulated problem:

$$
R'_{\perp} = \varkappa_1 S_{\perp} + \varkappa_2 S_{\parallel} + \alpha_0 R_{\parallel} ,
$$

\n
$$
R'_{\parallel} = \varkappa_3 S_{\perp} + \varkappa_4 S_{\parallel} - \alpha_0 R_{\perp} ,
$$

\n
$$
S'_{\perp} = -\varkappa_1 R_{\perp} - \varkappa_3 R_{\parallel} + \alpha_0 S_{\parallel} ,
$$

\n
$$
S'_{\parallel} = -\varkappa_2 R_{\perp} - \varkappa_4 R_{\parallel} - \alpha_0 S_{\perp} ,
$$

\n(1)

where $\alpha_0 = \alpha/\cos\varphi$; α is the specific rotation of the polarisation of light in a crystal; φ is the Bragg angle within the crystal; R_{\perp} , R_{\parallel} , S_{\perp} , and S_{\parallel} are, respectively, the projections of the vectors \vec{R} and \vec{S} of the electric field strength of the optical beams onto directions perpendicular and parallel to the plane of incidence. The coupling constants x_1, x_2, x_3 , and x_4 , taking into account the piezoelectric effect, may be represented in the following form for the (110) crystal cut in the case of an untilted holographic grating:

$$
\varkappa_1 = \frac{1}{2\cos\varphi} \varkappa E \cos\theta \left[2r(1 - 3\sin^2\theta) + 2(B + C)\sin^2\theta \right]
$$

$$
- (B + A) \Big],
$$

$$
\varkappa_{2,3} = \frac{1}{2} \varkappa E \sin\theta \left[2r(1 - 3\cos^2\theta) + 2(B + C)\cos^2\theta \right]
$$

$$
- C \mp 2D \tan\varphi \Big],
$$

$$
\varkappa_4 = \frac{1}{2} \varkappa E \cos\varphi \cos\theta \Big\{ 2[3r - (B + C)]\sin^2\theta + B - A \Big\}
$$

$$
(2)
$$

+2(P + r) tan²
$$
\varphi
$$
 ,
where E is the amplitude of the electric field strength of the
space charge; $\varkappa = \pi n^3/2\lambda$; *n* is the refractive index of the crys-
tal; λ is the wavelength of the optical beams; θ is the
orientation angle between the holographic-grating vector **K**
and the [001] direction [measured clockwise looking in the

direction of propagation of the optical beams (Fig. 1)];

$$
A = \frac{2e}{M}\sin^2\theta\{[2p_1 + (p_2 + p_3)][4c_2 - 3c_1
$$

\n
$$
- c_3 - (c_1 - 5c_3 - 4c_2)\cos 2\theta] + 2[2p_4 + (p_2 + p_3)]
$$

\n
$$
\times [c_2 - c_3 - 2c_1 - (c_2 - c_3 + 2c_1)\cos 2\theta]\};
$$

\n
$$
B = \frac{2e}{M}\sin^2\theta\{4p_4(c_2 - c_3 - 2c_1) - 4p_4(c_2 - c_3 + 2c_1)\cos 2\theta
$$

\n
$$
- [2p_1 - (p_2 + p_3)](c_1 + 2c_2 + c_3)(1 + 3\cos 2\theta)\};
$$

\n
$$
C = \frac{2ep_4}{M}[-(7c_2 + 5c_1 + 2c_3)\cos 4\theta
$$

\n
$$
+ 4(c_2 - 3c_1 + 2c_3)\cos 2\theta + 3(c_2 - 2c_3 - 5c_1)];
$$

\n
$$
D = \frac{4e}{M}\sin\theta\cos^2\theta(p_2 - p_3)(2c_2 + c_3 + c_1)(3\sin^2\theta - 2);
$$

Figure 1. Geometry of the interaction of optical waves in the crystal.

$$
P = \frac{2e}{M}\sin^2\theta\{[(p_2 + p_3)(4c_2 + 5c_3 - c_1) -2(p_1 - 2p_4)(2c_1 + c_2 - c_3)]\cos 2\theta + (p_2 + p_3) \times (4c_2 - 3c_1 - c_3) - 2(p_1 - 2p_4)(2c_1 + c_3 - c_2)\};
$$

\n
$$
M = [(c_1 + c_2)(c_3 - c_1) + 2(c_2 + c_3)^2] \cos 4\theta + 4c_3(c_1 - c_2 - 2c_3)\cos 2\theta + (c_1 + c_2)(c_1 + 3c_3) + 8c_3(c_1 + c_3) - 2(c_2 + c_3)^2 ;
$$

 $p_{1}\equiv p_{11}^E\equiv p_{22}^E\equiv p_{33}^E,\, p_{2}\equiv p_{12}^E\equiv p_{23}^E\equiv p_{31}^E,\, p_{3}\equiv p_{13}^E\equiv p_{21}^E\equiv p_{41}^E\equiv p_{52}^E\equiv p_{53}^E\equiv p_{61}^E\equiv p_{72}^E\equiv p_{81}^E\equiv p_{82}^E\equiv p_{83}^E\equiv p_{93}^E\equiv p_{94}^E\equiv p_{95}^E\equiv p_{95}^E\equiv p_{96}^E\equiv p_{97}^E\equiv p_{97}^E$ p_{32}^E , and $p_4 \equiv p_{44}^E \equiv p_{55}^E \equiv p_{66}^E$ are the photoelastic coefficients; $r \equiv r_{41} \equiv r_{123}^{\text{u}} \equiv r_{132}^{\text{u}} \equiv r_{231}^{\text{u}} \equiv r_{213}^{\text{u}} \equiv r_{312}^{\text{u}} \equiv r_{321}^{\text{u}}$ is the electro-optical coefficient of the clamped crystal; $c_1 \equiv c_{11}^E \equiv c_{22}^E \equiv c_{33}^E, \ \ c_2 \equiv c_{12}^E \equiv c_{13}^E \equiv c_{23}^E \equiv c_{31}^E \equiv c_{21}^E \equiv c_{32}^E,$ and $c_3 \equiv c_{44}^E \equiv c_{55}^E \equiv c_{66}^E$ are the elastic moduli; $e \equiv e_{14} \equiv$ $e_{123} \equiv e_{132} \equiv e_{213} \equiv e_{231} \equiv e_{312} \equiv e_{321}$ is the piezoelectric coefficient [\[24\].](#page-4-5)

The following expression is used to calculate the amplitude of the electric field strength of a holographic grating formed in the diffuse regime, neglecting thermal excitation [\[25\]:](#page-4-5)

$$
E = 2 \frac{1 - (1 - V^2)^{1/2}}{V} \frac{E_d}{1 + E_d/E_q},
$$
\n(3)

where E_d is the diffuse field of the grating; E_q is the saturation field; V is the interference pattern visibility.

Suppose that the optical beams R and S incident on a crystal are linearly polarised. For simplicity, we shall ignore the reflection of light on the crystal boundaries. We make use of the following expression for the interference pattern visibility $V(z)$ (see, for example, Ref. [\[22\]\)](#page-4-5) varying along the crystal thickness:

$$
V(z) = \frac{2[R_{\perp}(z)S_{\perp}(z) + R_{\parallel}(z)S_{\parallel}(z)\cos 2\varphi]}{I_0}, \qquad (4)
$$

where I_0 is the total intensity of the optical waves; $R_1(z) = R(z)$ sin $\psi_R(z)$, $R_{\parallel}(z) = R(z) \cos \psi_R(z)$, $S_{\perp}(z) = -S(z) \sin \psi_S(z)$, and $S_{\parallel}(z) = S(z) \cos \psi_s(z)$ are, respectively, the projections of the electric field vectors [\[9\]](#page-4-0) of the optical beams R and S onto directions perpendicular and parallel to the plane of incidence; $R(z)$, $S(z)$ are the electric field amplitudes; $\psi_R(z)$ and $\psi_s(z)$ are the polarisation azimuths of the optical waves in the crystal; the z axis is directed at right angles to the plane of the (110) cut in the crystal (Fig. 1).

We shall consider the case where the incident optical beams have the same polarisation azimuths. It follows from expression (4) that, for a sufficiently large Bragg angle, the interference pattern visibility and hence the field amplitude of a holographic grating [expression (3)] depend on the polarisation azimuths of the optical waves, which vary with the crystal thickness. For each orientation of the holographic-grating vector there is a specific optimal initial polarisation azimuth of the incident optical waves for which the relative intensity γ of the signal optical wave is maximal [\[11\].](#page-4-0) Therefore, for different orientation angles θ , the grating field amplitudes (corresponding to the maximum value of γ) will be different at the entry of the waves into the crystal. Furthermore, the field amplitude will vary along the crystal thickness because of variation of the polarisation angle $\psi(z) \approx \psi_R(z) \approx \psi_S(z)$ owing to the optical activity of the crystal and to the transfer of polarisation [\[9,](#page-4-0) [26\].](#page-4-5)

Thus, the system of equations (1) together with expressions $(2) - (4)$ constitute a theoretical basis for the investigation of maximisation of the relative intensity of the signal optical wave both in the constant visibility approximation (Section 3) and in the case when the self-diffraction is taken into account (Section 4).

3. Constant visibility approximation. Comparison of the theoretical results with the experiment

If the crystal thickness is so small that self-diffraction hardly influences the writing of a holographic grating and the initial polarisation azimuth of the optical waves does not change sufficiently during the passage of light through a crystal ($\alpha d \ll \pi$, where d is the crystal thickness), one may use the constant (along the crystal thickness) visibility approximation, and retain the dependence of the visibility on the initial polarisation state of the optical waves.

The expression for the interference pattern visibility then assumes the following form:

$$
V = 2(I_{R0}I_{S0})^{1/2}(\sin^2 \psi + \cos^2 \psi \cos 2\varphi)I_0^{-1} ,
$$
 (5)

where I_{R0} and I_{S0} are the intensities of the incident optical waves; ψ is the initial polarisation azimuth of the optical waves within the crystal. The angles ψ and φ generally differ from the angles ψ_0 and φ_0 in air (Fig. 1), but (if necessary) a relationship between them may be established.

Allowance for the expression (5) in formula (3) and subsequent use of formula (3) in the solution of the system of equations (1) make it possible to improve the agreement between the theoretical and experimental results, compared with the approximation [\[12\]](#page-4-0) which ignores the dependence of the grating visibility on the polarisation azimuth of the `read' waves.

Fig. 2 gives the dependence of the maximum relative intensity of the object optical wave γ_{ψ}^{m} on the orientation angle θ . The experimental results obtained in Ref. [\[12\]](#page-4-0) are indicated by symbols. The maxima were selected for each specified θ by choosing the optimal initial polarisation azimuth ψ of the optical waves for a crystal thickness of 2.1 mm and a Bragg angle of 13° . It is seen from Fig. 2 that, when account is taken of the influence of the initial polarisation azimuth of the optical waves on the field amplitude of a holographic grating (continuous curve), there is a characteristic leftward shift of the maxima in the $\gamma_{\psi}^{\text{m}}(\theta)$

Figure 2. Experimental (symbols) and theoretical dependences, on the orientation angle θ , of the maximum relative intensity γ_{ψ}^{m} of the object optical wave taking into account (continuous curve) and ignoring [\[12\]](#page-4-0) (dashed curve) the influence of the initial polarisation azimuth of the wave on the field amplitude of a holographic grating for a Bragg angle in the crystal of 13°, a crystal thickness of 2.1 mm, and $I_{R0}/I_{S0}=3.5$.

relationship, compared with the theoretical c[urve](#page-4-0) [12], and this agrees well with the experimental results [12].

Thus, even in the constant visibility approximation, allowance for the dependence of the electric field amplitude of a holographic grating on the polarisation azimuth of the `read' waves improves appreciably the agreement between the theoretical results and the experiment.

4. Optimisation of energy exchange taking the self-diffraction into account

In a study of the diffraction of light in a crystal of arbitrary thickness and for large Bragg angles, account must be taken of the influence of the energy redistribution between the optical waves during the writing of a hologram on the electric field amplitude of the holographic grating. This can be done by solving numerically the system of coupled-wave equations (1) with a stepwise variation of the interference pattern visibility [expression (4)] as a result of a redistribution of the energy of the optical waves between elementary constant-visibility holographic gratings. We may note that expression (4) takes into account implicitly both the optical activity and the piezoelectric effect through the dependence of the projections of the electric field vectors of the optical beams \bf{R} and \bf{S} on the specific rotation α of the polarisation of light in the crystal and on the piezoelectric coefficient e_{14} .

The method used to take into account the energy redistribution during the writing of a holographic grating in numerical solution of the system of equations (1) was as follows. We carried out an imaginary subdivision of the crystal into thin parallel layers Δz thick. We then specified the parameters of two incident optical beams (for $z = 0$), including their intensity and polarisation. The interference pattern visibility $V(0)$ and hence the field amplitude of the space charge E [formula (3)] were determined for $z = 0$ from formula (4). We then substituted E in the expressions for the coupling constants. Next, having solved numerically the coupled-wave equations (1) for the crystal thickness Δz , we obtained the optical-amplitude components R_{\perp} , R_{\parallel} , S_{\perp} , and S_{\parallel} at the exit from the first layer. These optical amplitudes obtained were substituted in formula (4) and $V(\Delta z)$ was found.We then determined the electric field amplitude of the grating E and the coupling constants x_1, x_2, x_3 , and x_4 for $z = \Delta z$ and solved the system of equations (1) for the crystal thickness Δz . We thus obtained the optical amplitudes at the exit from the second layer, i.e. for the crystal thickness $2\Delta z$, and the procedure was continued until the required crystal thickness was reached. The precision of the numerical calculations was increased by reducing Δz .

As in Ref. [\[19\],](#page-4-1) in the present study we considered only the first local maximum in the $v(d)$ relationship.

The constant grating and small Bragg angle approximations [\[19\]](#page-4-6) make it possible to obtain an analytical dependence of the relative gain in two-wave mixing, maximised simultaneously with respect to the polarisation angle and the crystal thickness, on the orientation angle. The dependences of the optimal parameters (the polarisation angle and the crystal thickness) on the orientation angle were also found analytically. Such dependences were used to calculate numerically the orientation and polarisation angles as well as the crystal thickness corresponding to the maximal gain and a small fixed Bragg angle.

An approach similar to that in Ref. [\[19\]](#page-4-7) was used in our study, but all the calculations were performed numerically because self-diffraction was taken into account.

As in Ref. [\[19\]](#page-4-1), we established initially the maximum relative intensity γ^m of the signal optical wave, selected from all the values obtained by varying the azimuth of the linearly polarised `read' light, the orientation angle, and the crystal thickness for a fixed Bragg angle. We then plotted $\gamma^{\rm m}$ against the Bragg angle and determined the angle φ ^m corresponding to the maximum relative intensity (point A in Fig. 3a). Next, we obtained the Bragg-angle dependence of the optimal parameters—the polarisation (ψ^m) and orientation (θ^m) angles for a crystal thickness d^m for which γ^m was attained. The optimal Bragg angle φ ^m (φ ^m \approx 11°), obtained in this way, was used to find the polarisation (point B in

Figure 3. Dependences of the maximum relative intensity of the signal optical wave γ^m (a), of the optimal azimuth of the 'read' light wave γ^m (b), of the optimal orientation angle θ^m (c), and of the crystal thickness d^m (d) on the Bragg angle φ .

Fig. 3b) and orientation (point C in Fig. 3c) angles, as well as the crystal thickness (point D in Fig. 3d) for which the relative intensity of the signal optical wave had the absolute maximum value, i.e. when it was maximised simultaneously with respect to four parameters.

The method described above was used to establish that, in the maximisation of the relative intensity of the signal optical wave simultaneously with respect to four parameters (the polarisation and orientation angles, the crystal thickness, and the Bragg angle), there are two sets of these optimal parameters for which the relative intensity assumes virtually a single maximum value. Subsequently, we arbitrarily associated a relative intensity maximum with the first of the sets and referred to it as the first maximum. We then assigned a maximum to the second set and called it the second maximum.We shall now describe how we found the first of these two sets.

In the calculations we used the same constants of the BSO crystal as in Ref. [\[11\].](#page-4-0) The wavelength of the 'read' light λ was assumed to be 632.8 nm, the initial intensity ratio of the optical waves was $I_{R0}/I_{S0}=400$, and the acceptor-impurity concentration was $N_a = 10^{22} \text{ m}^{-3}$ [\[2\].](#page-4-2) It can be seen from Fig. 3 that the parameters corresponding to the absolute maximum of the relative intensity are as follows: $\varphi^m \approx 11^\circ$, $\theta^m \approx 39.1^\circ$, $\psi^m \approx 98.85^\circ$, and $d^m \approx 7.11$ mm. The second set of the optimal parameters corresponding to the second relative intensity maximum was determined in a similar way: $\varphi^m \approx 11^\circ$, $\theta^m \approx 320.9^\circ$, $\psi^m \approx 54.15^\circ$, and $d^m \approx 7.11$ mm. In finding the optimal parameters φ^m , θ^m , ψ^m , and d^m , the concentration of acceptor impurities N_a was assumed to be typical for BSO: $N_a = 10^{22}$ m⁻³ [\[2\].](#page-4-2) However, in the course of our calculations it was found that variation of the concentration of the acceptor impurities in the range $10^{21} - 4 \times 10^{22}$ m⁻³ hardly altered the dependences plotted in Figs 3b, 3c, and 3d. However, as can be seen from Fig. 4, the optimal Bragg angle φ ^m depended significantly on N_a .

Figure 4. Dependence of the optimal Bragg angle φ ^m on the concentration of acceptor impurities N_a .

Knowing the acceptor concentration in the BSO crystal, it is possible to determine the optimal Bragg angle φ ^m with the aid of Fig. 4 and then, by using the dependences plotted in Figs 3b, 3c, and 3d, to obtain the remaining parameters θ^m , ψ^m , and d^m for which the relative intensity of the signal optical wave is maximised simultaneously with respect to four of the above parameters. We may note that γ^m increases monotonically with increase in the acceptor impurity concentration N_a .

5. Conclusions

The problem of maximising the relative intensity of the signal optical wave in a BSO crystal simultaneously with respect to four parameters (namely the polarisation and orientation angles, the crystal thickness, and the Bragg angle) was solved on the basis of a phenomenological model of the holographic process in cubic photorefractive gyrotropic piezoelectric crystals. The model is based on a numerical solution of a system of coupled equations with stepwise variation of the interference pattern visibility as a result of a redistribution of the optical wave energy between elementary constant-visibility holographic gratings.

It was established that there are two sets of the optimal parameters for which the maximum relative intensity is virtually the same. Taking into account the self-diffraction we found that, at an acceptor concentration $N_a = 10^{22}$ m⁻³ typical for BSO and for the wavelength of 632.8 nm, the parameters were as follows: $\varphi^m \approx 11^{\circ}$, $\theta^m \approx 39.1^{\circ}$, $\varphi^m \approx$ 98.85°, and $d^m \approx 7.11$ mm (for the first maximum) and $\varphi^m \approx 11^\circ$, $\theta^m \approx 320.9^\circ$, $\psi^m \approx 54.15^\circ$, and $d^m \approx 7.11$ mm (for the second maximum). In addition, the dependence of these optimal parameters on the acceptor concentration in the BSO crystal was determined in the range 10^{21} – 4×10^{22} m⁻³ .

The theoretical results obtained in the constant-visibility approximation, with the visibility fixed for each initial polarisation azimuth of the optical waves, agreed satisfactorily with the available experimental data [\[12\],](#page-4-0) improving the agreement between the theoretical and experimental relationships compared with the approximation in which the electric-field amplitude of the grating is independent of the polarisation azimuths of the optical waves.

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