Unique behavior of thickness dependence in the nonlinear wave-mixing process with a nematic thin film

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The dependence of the optimal electric biasing field and the diffraction efficiency on the thickness of the nematic thin film through the degenerate four-wave-mixing process is investigated. For relatively low optical pumps, the optimal bias monotonically increases with thickness, while the corresponding diffraction efficiency increases at first and then decays for a thickness even less than 150 μm, which is well within the phase-matching regime. For stronger pumps, the optimal bias can be doubly valued, and the curve of optimal bias versus thickness shows a closed loop. This behavior is unique to liquid crystals. The main mechanism is due to the twist effect of molecular orientation in addition to the scattering loss. Both the numerical and experimental results show these peculiar phenomena.

The physical origin as well as the efficiency of degenerate four-wave mixing[1-6] (DFWM) are consistently interesting subjects in nonlinear optics. For a common medium,[3] the nonlinear effects become more prominent when the interaction length is increased within the phase-matching regime. In nematic liquid crystals (NLC's), the deformation and fluctuation of the director are also involved in the nonlinear process. Therefore the threshold intensity, the nonlinear coefficient, and the linear loss are strongly dependent on the physical dimension of the medium. Armitage and Delwart[4] and Khoo and Lin[5] have reported that the diffraction efficiency increases and then decreases with respect to the sample thickness for DFWM in NLC's. However, they attributed this phenomenon to the scattering loss of the medium. The experimental results in Ref. 5 have shown that the curve of diffraction efficiency increases monotonically after the correction of scattering loss. The large optical nonlinearity based on molecular reorientation can be further enhanced by an applied static field. In previous studies[6] we have illustrated that the peak efficiency of DFWM with respect to the electric field can be obtained at a specific bias that is strongly dependent on the elastic deformation in NLC films. If the nonlinear phase shift is large enough, local maximum efficiency can be observed at two distinct voltages.

In this Letter we report the novel effects of NLC film thickness on the wave-mixing process. Instead of using multiple-layer NLC films,[5] we use a single-layer sample with spacers of different thicknesses. Increasing the thickness by layer numbers does not change the nonlinear coefficient of the sample film. It is emphasized that the nonlinear coefficient is no longer a constant in our samples of different thicknesses. It is illustrated that after correcting the scattering loss, there is still a drop in the plot of diffraction efficiency versus thickness. Moreover, the curve of optimal biasing field versus thickness shows a novel closed loop within the double-peak regime instead of the two divergent curves that are usually obtained with respect to other parameters.[6] The twist effect and nonlinear phase-shift accumulation in NLC are the crucial factors accounting for these effects, since the twist deformation (and the nonlinear coefficient) decreases and the phase-shift accumulation increases with an increase of thickness.

Figure 1 schematically depicts the problem under study. Consider a homeotropically aligned NLC cell of thickness d with an electric field (1 kHz) applied parallel to the unperturbed director $\hat{n}_o$. Two laser beams at $\lambda_0 = 514.5$ nm, with intensities I_1 and I_2, are nearly normally incident on and overlapped in the sample with a fairly small intersection angle α. The nematic substance is assumed to have negative dielectric anisotropies, namely, $\epsilon_{\parallel} < \epsilon_{\perp}$. For a sufficiently thin sample, DFWM can be treated as diffraction from the induced phase grating that is created under the steady illumination of an optical intensity $I(x,y,z) = I_1 + I_2 + 2\sqrt{I_1 I_2}\cos(2\pi x/\Lambda)$, with the grating period $\Lambda = \lambda_0/[2\sin(\alpha/2)]$. In general, the spatial distribution of molecular reorientation in NLC media is a local response to the distribution of the intensity grating. With hard boundaries assumed [i.e., $\theta(z=0) = \theta(z=d) = 0$], the angle of reorientation in the first-order approximation can be expressed as $\theta(x,z) = [\theta_1 + \theta_2\cos(2\pi x/\Lambda)]\sin(\pi z/d)$. The equilibrium values of the constants θ_1 and θ_2

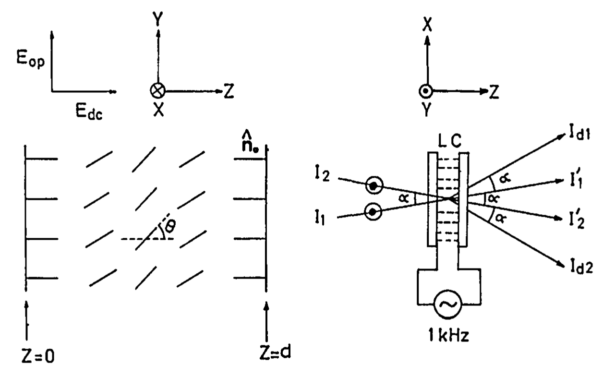

Fig. 1. Schematic of the nonlinear wave-mixing process.

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can be calculated from the minimization of total free energy F by letting $\partial F/\partial\theta_1 = 0$ and $\partial F/2\theta_2 = 0$. Under the assumptions that $\theta_1 < \pi/2$ and $\theta_2^2 \ll 1$, which are always satisfied in the following calculations and experiments, we have[6]

$$\theta_1 - KG_1(\theta_1, \theta_2) - (V/V_{\rm th})^2 G_2(\theta_1, \theta_2) - I_r G_3(\theta_1, \theta_2) + (I_t/I_{\rm th})G_4(\theta_1, \theta_2) = 0\,, \quad (1)$$

$$\theta_2 \simeq \frac{2I_r J_1(2\theta_1)}{1 + 2a - (1+b)[J_0(2\theta_1) - J_2(2\theta_1)] - KG_5(\theta_1, 0)}\,, \quad (2)$$

where $a \equiv 2(K_2/K_3)(d/\Lambda)^2$ is termed the twist ratio hereafter; $b \equiv I_t/I_{\rm th} + (V/V_{\rm th})^2 - 1$ is the reduced effective field; $K = 1 - K_1/K_3$, with K_1, K_2, and K_3 the splay, twist, and bend elastic constants, respectively; $G_i(\theta_1, \theta_2)$, $i = 1, 5$ are polynomial functions of θ_1 and θ_2; V is the applied voltage, and $V_{\rm th} = \pi(4\pi K_3/|\Delta\epsilon|)^{1/2}$ is the threshold voltage, with $\Delta\epsilon = \epsilon_\parallel - \epsilon_\perp$; $I_{\rm th} = (\pi/d)^2(cK_3/n_o u)$ is the threshold intensity, with $u = 1 - n_o^2/n_e^2$; c is the velocity of light in a vacuum; n_o and n_e are the ordinary and maximum extraordinary refractive indices, respectively; $I_r = \sqrt{I_1 I_2}/I_{\rm th}$ and $I_t = I_1 + I_2$; and $J_i(2\theta_1)$ is the Bessel function of the first kind, of order i.

If the local angle θ is known, the effective refractive index can then be obtained. The corresponding induced phase shift across the sample is $\delta(x) \cong \delta_0 + \delta_1 \cos(2\pi x/\Lambda)$, where δ_0 and δ_1 are the uniform phase retardation and the amplitude of this nonlinear phase grating, respectively. For $u \sin^2\theta \ll 1$ (for the usual nematics $u \ll 1$), we have

$$\delta_1 = (2\pi d/\lambda_0)\Delta\bar{n}_{\rm NL} = \pi u n_o d\theta_2 J_1(2\theta_1)/\lambda_0\,, \quad (3)$$

where $\Delta\bar{n}_{\rm NL}$ is the nonlinear refractive-index change averaged over sample thickness. The relative diffraction efficiency, referring to the fundamental diffraction beam I_{d_1}, is

$$\eta \equiv I_{d_1}/I_t \simeq r_1[J_1(\delta_1)]^2 + r_2[J_2(\delta_1)]^2\,, \quad (4)$$

where $r_1 = I_1/I_t$, $r_2 = I_2/I_t$, and $r_1 + r_2 = 1$. The behavior of the diffraction efficiency η is characterized by the property of the nonlinear phase shift δ_1. While the reduced optical intensity $I_t/I_{\rm th}$ is fixed, the optimal bias $b_{\rm pm}$ for the maximum phase amplitude δ_{1m} can be obtained by letting the first-order derivative of δ_1 be zero, i.e., $\partial\delta_1/\partial b = 0$. In the extreme case of $\theta_2^2 \ll \theta_1^2 \ll 1$, $a \ll 0.5$, and $K = 0$, the optimal field becomes

$$b_{\rm pm} \simeq \sqrt{K_2/K_3}(d/\Lambda)\,, \quad (5)$$

and the corresponding maximum phase is

$$\delta_{1m} = (2\pi d/\lambda_0)\Delta\bar{n}_{\rm NL}(b_{\rm pm}) \simeq (2\pi d/\lambda_0)(u n_o I_r)[1 - 4\sqrt{K_2/K_3}(d/\Lambda)]\,, \quad (6)$$

where $\Delta\bar{n}_{\rm NL}(b_{\rm pm})$ is the corresponding $\Delta\bar{n}_{\rm NL}$ at $b_{\rm pm}$ and is also the maximum value of $\Delta\bar{n}_{\rm NL}$ with respect to b. Note that $\Delta\bar{n}_{\rm NL}(b_{\rm pm})$ decreases with an increase of d. This is also true for the more general case as shown by the results of our numerical calculation.

Our numerical calculations have been made from relations (1)–(4) to illustrate the unique behavior of thickness dependence. The results are shown in Figs. 2 and 3(a). The maximum phase amplitude δ_{1m} and the corresponding average nonlinear refractive index $\Delta\bar{n}_{\rm NL}$ versus sample thickness d for $I_t/I_{\rm th} = 0.01$ are plotted in Fig. 2(a). Instead of monotonically increasing, the curve of δ_{1m} versus d has a peak value at optimal thickness d_m. This is obvious since the optical path increases and $\Delta\bar{n}_{\rm NL}$ decreases with increasing d. We attribute this phenomenon to the twist effect. The reason is that our grating is characterized by the twist angle θ_2, where the twist is along x. θ_2 decreases and so does $\Delta\bar{n}_{\rm NL}$ as the sample thickness increases. By letting $\partial\delta_{1m}/\partial d = 0$, we have the approximated optimal thickness $d_m \simeq 0.13\Lambda\sqrt{K_3/K_2}$ [as derived from relation (6)]. If the actual total intensity I_t rather than the reduced intensity $I_t/I_{\rm th}$ is kept constant, we can obtain another optimal value $d_m' \simeq 0.19\Lambda\sqrt{K_3/K_2}$. It favors a larger optimal thickness in the latter case. The reason

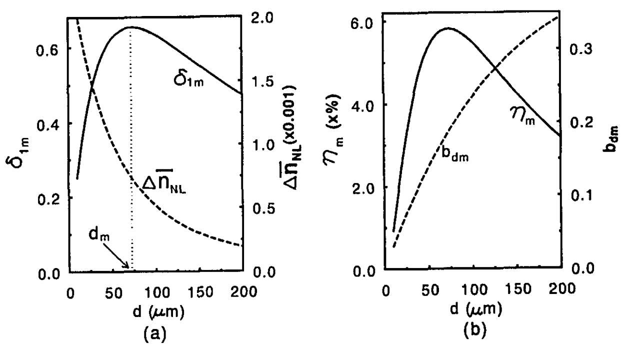

Fig. 2. Numerical results of (a) optimal phase amplitude δ_{1m} and the corresponding nonlinear refractive index $\Delta\bar{n}_{\rm NL}$ and (b) peak diffraction efficiency η_m and optimal field $b_{\rm dm}$ versus the sample thickness d. Parameters used are $n_e = 1.81$, $n_o = 1.57$, $|\Delta\epsilon| = 0.5$, $r_1 = 0.595$, $r_2 = 0.405$, $K = 0.23$, $K_2/K_3 = 4/7.5$, $\Lambda = 180$ μm, and $I_t/I_{\rm th} = 0.01$.

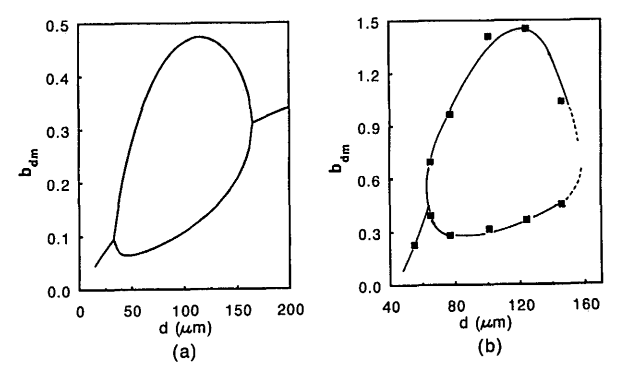

Fig. 3. Optimal field $b_{\rm dm}$ versus the sample thickness d in the double-peak regime. (a) Numerical results calculated with $I_t/I_{\rm th} = 0.04$; other parameters are the same as in Fig. 2. (b) Experimental results with $I_t/I_{\rm th} = 0.25$ and $\Lambda = 134$ μm. Both the solid and dashed curves are a guide for the eye. The environmental temperature is approximately 22 °C, and the weights are $r_1 = 0.595$ and $r_2 = 0.405$.

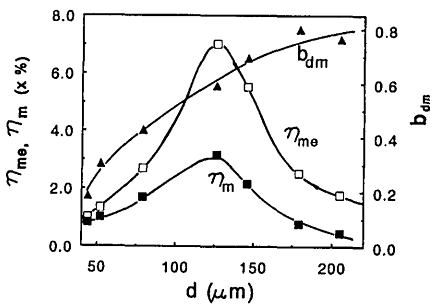

Fig. 4. Experimental results of peak diffraction efficiency η_m, effective peak efficiency η_{me}, and optimal field b_{dm} versus the sample thickness d in the single-peak regime. η_{me} is defined as $\eta_m/\exp(-\beta d)$, where β (cm^{-1}) is the measured loss coefficient. The curves are a guide for the eye. The environmental temperature is approximately 26 °C.

is that the effective pump I_t/I_{th} is simultaneously increased with d for fixed I_t, because the threshold intensity I_{th} is inversely proportional to the square of thickness.

In the case of $r_1 = 0.595$ and $r_2 = 0.405$, the diffraction efficiency η in Eq. (4) has a maximum with respect to the specific phase shift $\delta_1 = 2.075$. For $\delta_1 < 2.075$, η is a monotonically increasing function of δ_1. There is only one peak[6] in the plot of η versus b, and the optimal bias b_{dm} for the maximum efficiency η_m is the same as that (i.e., the term b_{pm}) for the maximum phase amplitude δ_{1m}. The calculated result of η_m versus d is plotted in Fig. 2(b), and its behavior is essentially the same as that of δ_{1m}. The optimal field b_{dm}, which is subjected to the twist ratio as illustrated in relation (5), increases with d. Since the general behavior of δ_1 versus b is increasing from zero and then decreasing to zero again, stronger pumps that actuate δ_{1m} to exceed 2.075 can result in the occurrence of double peaks[7] (where δ_1 equals 2.075) in the plot of diffraction efficiency versus electric bias. Referring to the general trend of the δ_{1m} curve in Fig. 2(a), double peaks can appear only in the middle of the thickness range, where δ_{1m} is larger than 2.075. This is illustrated by the numerical results shown in Fig. 3(a). It is obvious that the double peaks appear only in the middle of the thickness range and shows a novel closed loop instead of two divergent curves. The maximum phase δ_{1m} corresponding to both end points of the loop is exactly 2.075.

Fresh samples of good optical quality are prepared by sandwiching the nematic substance N-(4-methoxybenzylidene)-4-butylaniline between two glass windows. The sample thickness is controlled by a calibrated Mylar spacer. The diameter of the laser spot is larger than 1 mm. The detail of the experiments is essentially the same as that described in our previous report.[6] The threshold intensity and voltage, e.g., of a 92-μm-thick sample, are determined to be 464 W/cm^2 and 3.74 V, respectively. Figure 4 gives the observed peak efficiency η_m and optimal biasing field b_{dm} as a function of thickness in the single-peak regime. The effective peak efficiency η_{me}, as normalized to the linear scattering loss,[5] is also plotted in the figure. The grating period is fixed at 112 μm, and the incident intensities are kept at $I_1 = 0.0062 I_{th}$ and $I_2 = 0.0058 I_{th}$, respectively. b_{dm} increases with thickness d as it is subjected to the twist effect. It is unambiguously shown that both η_m and η_{me} have maximum values near 125 μm. It is obvious that the decreasing diffraction efficiency with respect to thickness is not only due to scattering loss but also due to the suppression of the twist deformation. The crucial influence of the sample thickness on the peak efficiency and optimal bias illustrated here agrees with the numerical results shown in Fig. 2(b). The experimental result in Fig. 3(b) exhibits the characteristic behavior of the numerical calculation in Fig. 3(a). Although the closed-loop curve of b_{dm}'s is not thoroughly presented under our experiments, the b_{dm}'s seem to come closer in the larger thickness range. It is almost impossible for us to determine the optimal bias for the relatively thick samples because the simultaneous occurrence of self-phase-modulation effects[8] (several rings are observed) makes it difficult to distinguish the lowest-order diffraction beam from the other spots.

In conclusion, we have shown that the sample thickness dependence of DFWM by means of molecular reorientation in liquid crystals is unique. That is, with weak input beams, the diffraction intensity exhibits a local maximum within the phase-matched regime instead of monotonically increasing as for a common nonlinear medium. The optimal biasing field for diffraction efficiency with respect to the thickness shows a novel closed loop in the double-peak regime with strong input beams, instead of two divergent curves that are usually obtained with respect to other parameters, e.g., the grating period. The twist effect and nonlinear phase-shift accumulation in NLC are the crucial factors that account for these phenomena.

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References

1. S. D. Durbin, S. M. Arakelian, and Y. R. Shen, Opt. Lett. **7**, 145 (1982).
2. N. V. Tabiryan, A. V. Sukhov, and B. Ya. Zel'dovich, Mol. Cryst. Liq. Cryst. **136**, 1 (1986).
3. See, e.g., R. A. Fisher, ed., *Optical Phase Conjugation* (Academic, New York, 1983).
4. D. Armitage and S. M. Delwart, Mol. Cryst. Liq. Cryst. **122**, 59 (1985).
5. I. C. Khoo and T. H. Liu, Phys. Rev. A **39**, 4036 (1989).
6. S.-H. Chen and C.-L. Kuo, Appl. Phys. Lett. **55**, 1820 (1989); C.-L. Kuo and S.-H. Chen, J. Appl. Phys. **68**, 4413 (1990); S.-H. Chen and C.-L. Kuo, Mol. Cryst. Liq. Cryst. **207**, 131 (1991).
7. C.-L. Kuo and S.-H. Chen, Opt. Lett. **15**, 610 (1990).
8. S. D. Durbin, S. M. Arakelian, and Y. R. Shen, Opt. Lett. **6**, 411 (1981).