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 (DFWM) are consistently interesting subjects in nonlinear optics. For a common medium, 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 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 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, 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, 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.

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 . Two laser beams at , with intensities and , 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, . 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 , with the grating period . 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., , the angle of reorientation in the first-order approximation can be expressed as . The equilibrium values of the constants and are

![Fig. 1. Schematic of the nonlinear wave-mixing process.](image-url)
can be calculated from the minimization of total free energy $F$ by letting $\partial F/\partial \theta_1 = 0$ and $\partial F/\partial \theta_2 = 0$. 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

$$\theta_2 = \frac{2I_r J_0(2\theta_1)}{1 + 2a - (1 + b)[J_0(2\theta_1) - J_2(2\theta_1)] - KG_0(\theta_1, 0)},$$

where $a = 2(K_2/K_3)(d/\lambda)^2$ is termed the twist ratio hereafter; $b = I_s/I_{th} + (V/V_{th})^2 - 1$ 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; $G_0(\theta_1, \theta_2)$, $i = 1, 5$ are polynomial functions of $\theta_1$ and $\theta_2$; $V$ is the applied voltage, and $V_{th} = \pi(4\pi K_3/\Delta e)^{1/2}$ is the threshold voltage, with $\Delta e = e_1 - e_2$. \(I_{th} = (d/\lambda)^2(cK_3/n_0)\) is the threshold intensity, with $u = 1 - n_0^2/n_2^2$; $c$ is the velocity of light in a vacuum; $n_0$ and $n_2$ are the ordinary and maximum extraordinary refractive indices, respectively; $I_s = \sqrt{I_1 I_2}/I_{th}$ and $I_r = I_1 + I_2$; and $J_i(2\theta_1)$ is the Bessel function of the first kind, of order $i$.

If the local angle $\theta$ is known, the effective refractive index can then be obtained. The corresponding induced phase shift across the sample is $\delta(x) = \delta_0 + \delta_1 \cos(2\pi x/\Lambda)$, where $\delta_0$ and $\delta_1$ 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

$$\delta_1 = (2\pi d/\lambda_0)\Delta n_{NL} = \pi u n_0 d \theta_0 J_1(2\theta_1)/\lambda_0,$$

where $\Delta n_{NL} = \Delta n_{NL}(d, \lambda)$ is the nonlinear refractive-index change averaged over sample thickness. The relative diffraction efficiency, referring to the fundamental diffraction beam $I_{d1}$, is

$$\eta = I_{d1}/I_s = r_1 J_1(\delta_1)^2 + r_2 J_2(\delta_1)^2,$$

where $r_1 = I_1/I_{th}$, $r_2 = I_2/I_{th}$, and $r_1 + r_2 = 1$. The behavior of the diffraction efficiency $\eta$ is characterized by the property of the nonlinear phase shift $\delta_1$. While the reduced optical intensity $I_s/I_{th}$ is fixed, the optimal bias $b_{pm}$ for the maximum phase amplitude $\delta_m$ can be obtained by letting the first-order derivative of $\delta_1$ 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

$$b_{pm} = \sqrt{K_2/K_3}(d/\lambda),$$

and the corresponding maximum phase is

$$\delta_m = (2\pi d/\lambda_0)\Delta n_{NL}(b_{pm}) = (2\pi d/\lambda_0)(u n_0 I_s)[1 - 4\sqrt{K_2/K_3}(d/\lambda)],$$

where $\Delta n_{NL}(b_{pm})$ is the corresponding $\Delta n_{NL}$ at $b_{pm}$ and is also the maximum value of $\Delta n_{NL}$ with respect to $b$. Note that $\Delta n_{NL}(b_{pm})$ 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 $\delta_m$ and the corresponding average nonlinear refractive index $\Delta n_{NL}$ versus sample thickness $d$ for $I_s/I_{th} = 0.01$ are plotted in Fig. 2(a). Instead of monotonically increasing, the curve of $\delta_m$ versus $d$ has a peak value at optimal thickness $d_m$. This is obvious since the optical path increases and $\Delta n_{NL}$ decreases with increasing $d$. We attribute this phenomenon to the twist effect. The reason is that our grating is characterized by the twist angle $\theta_2$, where the twist is along $x$. $\theta_2$ decreases and so does $\Delta n_{NL}$ as the sample thickness increases. By letting $\partial \delta_m/\partial d = 0$, we have

$$\Delta n_{NL} = 0.13 \sqrt{K_2/K_3} \text{ [as derived from relation (6)].}$$

If the actual total intensity $I_s$ rather than the reduced intensity $I_s/I_{th}$ is kept constant, we can obtain another optimal value $d_m' = 0.19 \sqrt{K_2/K_3}$. It favors a larger optimal thickness in the latter case. The reason
is that the effective pump $I_1/I_{th}$ is simultaneously increased with $d$ for fixed $I_1$, 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 $\eta$ in Eq. (4) has a maximum with respect to the specific phase shift $\delta_1 = 2.075$. For $\delta_1 < 2.075$, $\eta$ is a monotonically increasing function of $\delta_1$. There is only one peak of $\eta$ versus $b$, and the optimal bias $b_{dm}$ for the maximum efficiency $\eta_m$ is the same as that (i.e., the term $b_{dm}$) for the maximum phase amplitude $\delta_{1m}$. The calculated result of $\eta_m$ versus $d$ is plotted in Fig. 2(b), and its behavior is essentially the same as that of $\delta_{1m}$. 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 $\delta_1$ versus $b$ is increasing from zero and then decreasing to zero again, stronger pumps that actuate $\delta_{1m}$ to exceed 2.075 can result in the occurrence of double peaks (where $\delta_1$ equals 2.075) in the plot of diffraction efficiency versus electric bias. Referring to the general trend of the $\delta_{1m}$ curve in Fig. 2(a), double peaks can appear only in the middle of the thickness range, where $\delta_{1m}$ 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 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