Double peaks of fundamental diffraction efficiency versus biasing of electric fields through the degenerate four-wave mixing process in nematic liquid-crystal films

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The observation of double peaks of diffraction efficiency through the degenerate four-wave mixing process as a function of the biasing electric-field strength in a nematic liquid-crystal film is described. This behavior is due to the high optical nonlinearity of liquid-crystal media and the dramatic modulating effect of an ac voltage on the reorientational Freedericksz transition. The dependence of the peak occurrence on grating periods, electric fields, and optical intensities is presented. Theoretical calculations show the same characteristic behavior as do the experimental results.

Optical wave mixing in nematic liquid-crystal (NLC) films, such as wave-front conjugation[1] and self-diffraction,[2–7] has been studied by scientists for several years. Because of the fairly large optical nonlinearity, the distribution of the optical intensities of various beams in the medium must be calculated from a set of coupled-wave equations in general. At the same time the molecular reorientation as well as the coupling coefficient, which depend on the experimental conditions, must be determined by minimization of the total free energy[8] in the NLC film. These will make the theoretical calculations fairly complicated. However, for a sufficiently thin sample,[3] degenerate four-wave mixing (DFWM) can be simply described by diffraction from an induced phase grating derived from the free energy by neglecting the higher-order gratings and the energy variation of incident beams in the sample. In addition to the optical pumps, DFWM in a NLC medium can be strongly influenced by a low-frequency electric or magnetic field. While only a few reports are available,[3,6,7] none of them has adequately discussed the behavior of the diffraction efficiency with respect to the biasing field.

In this Letter we report the first observation, to our knowledge, of double peaks in the plot of the diffraction efficiency of DFWM versus biasing electric fields. By our theoretical model derived from the continuum theory,[8] these phenomena can be explained by the large induced nonlinear refractive-index change and the dramatic modulating effect of the biasing field in the reorientational Freedericksz transition. Prominent double diffraction peaks are obtained by both experimental and numerical results.

The experimental geometry is shown in the inset of Fig. 1. A quasi-static electric field at 1 kHz is applied along the unperturbed molecular direction $\hat{n}$ of the homeotropically aligned NLC film, which is assumed to have negative dielectric anisotropy, namely, $\epsilon_{\parallel} < \epsilon_{\perp}$. Two mutually coherent laser beams at $\lambda_0 = 514.5$ nm, with intensities I_1 and I_2, derived from the same Ar^+ source are nearly normally incident upon and overlapped in the sample with an intersection angle α. The interference of two beams will create a periodically distorted orientational structure, which then gives risk to an induced phase grating, with the same period $\Lambda = \lambda_0/[2\sin(\alpha/2)]$ in the sample. In equilibrium, the reorientational angle $\theta(x, z)$ can be obtained by minimizing the total free energy F of the sample, $F = \int \mathcal{F} dv$. $\mathcal{F}$ is the free-energy density and is given by[9] $\mathcal{F} = K_2(\partial\theta/\partial x)^2/2 + K_3(1 - K\sin^2\theta)(\partial\theta/\partial z)^2/2 - D_z^2/8\pi\epsilon_{\parallel}(1 - w\sin^2\theta) + In_o/c(1 - u\sin^2\theta)^{1/2}$, where K is defined as $1 - K_1/K_3$, with K_1, K_2, and K_3 the splay, twist, and bend elastic constants, respectively; $w = 1 - \epsilon_{\perp}/\epsilon_{\parallel}$; D_z is the z component of the electric displacement; $I = I_1 + I_2 + \sqrt{I_1 I_2}\cos(2\pi x/\Lambda)$ is the optical intensity; c is the velocity of light in vacuum; and u is defined as $1 - n_o^2/n_e^2$, with n_o and n_e representing the ordinary and maximum extraordinary refractive indices, respectively. Under the assumption of infinite incident plane waves, $\theta(x, z)$ can be expressed as

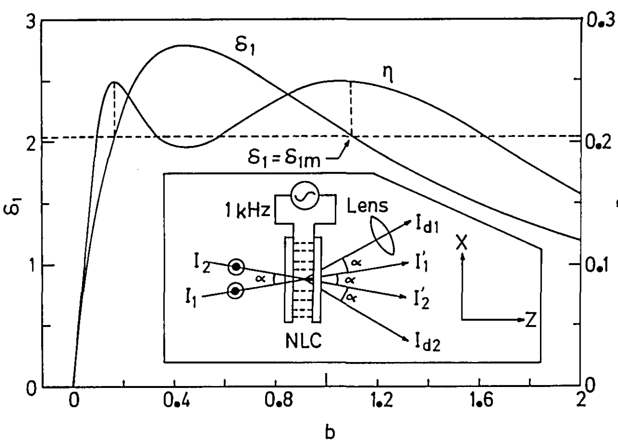

Fig. 1. Phase amplitude δ_1 and diffraction efficiency η versus the reduced effective field b with $\delta_{1m} = 2.075$, $\eta_m = 0.25$, and $\delta_{max} = 2.78$. The inset shows the experimental geometry.

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$$\theta(x, z) = \left[\theta_1 + \theta_2 \cos\left(\frac{2\pi x}{\Lambda}\right)\right]\sin\left(\frac{\pi z}{d}\right), \quad (1)$$

which represents the lowest-order Fourier component consistent with the boundary conditions $\theta(x, 0) = 0$ and $\theta(x, d) = 0$, where d is the sample thickness. The equilibrium values of the constants θ_1 and θ_2 can be calculated from minimization of the total free energy F by letting $\partial F/\partial\theta_1 = 0$ and $\partial F/\partial\theta_2 = 0$. In order to avoid too many truncation errors in calculating F, we keep terms associated with θ_1 and θ_2 to the second order under the assumption of $\theta_2{}^2 \ll 1$, which is satisfied in the following calculations and experiments. Then we have

$$\theta_2 \simeq \frac{2I_r J_1(2\theta_1) + KJ_2(2\theta_1)}{1 + (K_2/K_3)(2d/\Lambda)^2 - \left[\frac{I_t}{I_{\mathrm{th}}} + \left(\frac{V}{V_{\mathrm{th}}}\right)^2\right][J_0(2\theta_1) - J_2(2\theta_1)] - Kf(\theta_1)}, \quad (2)$$

where $I_{\mathrm{th}} = (\pi/d)^2 cK_3/un_o$ is the threshold intensity, $I_r = \sqrt{I_1 I_2}/I_{\mathrm{th}}$, $I_t = I_1 + I_2$, V is the applied voltage, $V_{\mathrm{th}} = \pi(4\pi K_3/|\Delta\epsilon|)^{1/2}$ is the threshold voltage with $\Delta\epsilon = \epsilon_\parallel - \epsilon_\perp$, J_0, J_1, and J_2 are the zeroth-, first-, and second-order Bessel functions, respectively, and $f(\theta_1)$ depends on θ_1 and has a positive functional value. Here we point out that the larger the constant K is (which means less splay restoring force is present), the larger the distortion angle θ_2 becomes.

By neglecting the higher-order terms in the Taylor series, the effective refractive index $n(x)$ for a uniaxial birefringent medium has the functional form

$$n(x) = \bar{n} + \Delta\bar{n}_{\mathrm{NL}} \cos\left(\frac{2\pi x}{\Lambda}\right), \quad (3)$$

where $\bar{n}$ is the spatially uniform refractive index and $\Delta\bar{n}_{\mathrm{NL}} = un_o\theta_2 J_1(2\theta_1)/2$ is the modulation index of this grating. Both $\bar{n}$ and $\Delta\bar{n}_{\mathrm{NL}}$ are the mean values over the sample thickness. The corresponding induced phase shift across the sample is

$$\delta(x) = \delta_0 + \delta_1 \cos\left(\frac{2\pi x}{\Lambda}\right), \quad (4)$$

where $\delta_0 = \bar{n}(2\pi/\lambda_0)d$ is the uniform phase shift and $\delta_1 = \Delta\bar{n}_{\mathrm{NL}}(2\pi/\lambda_0)d$ is the modulation amplitude of the corresponding phase grating.

By satisfying the thin-grating condition, i.e., $2\pi^2(\Delta\bar{n}_{\mathrm{NL}}/\bar{n})(d/\Lambda)^2 < 1$, the discussion can be confined in the Raman–Nath regime[10] with the process of the Fraunhofer diffraction[11] from a sinusoidal phase grating. The total power of I_{d1}, which is depicted in the inset of Fig. 1, comes from the first-order diffraction of E_1 and the second-order diffraction of E_2. The diffraction efficiency, which is discussed throughout this Letter, is defined as

$$\eta \equiv \frac{I_{d1}}{I_t} = r_1[J_1(\delta_1)]^2 + r_2[J_2(\delta_1)]^2, \quad (5)$$

where $r_1 = I_1/I_t$, $r_2 = I_2/I_t$, and $r_1 + r_2 = 1$. There exists a maximum diffraction efficiency η_m in Eq. (5) with respect to the unique optimal phase amplitude δ_{1m} that depends on the weights r_1 and r_2 only. In the case of $\theta_2{}^2 \ll \theta_1{}^2 \ll 1$ and $K = 0$, we can treat[12] the grating structure as a perturbation on the uniform molecular reorientation induced by the spatial average effective field and find that $\theta_1 \simeq \sqrt{2b}$ and $\theta_2 \simeq 2I_r J_1(2\theta_1)/\{1 + 2a - (1 + b)[J_0(2\theta_1) - J_2(2\theta_1)]\}$, where $b \equiv I_t/I_{\mathrm{th}} + (V/V_{\mathrm{th}})^2 - 1$ is defined as the reduced effective field and $a = (K_2/K_3)(2d/\Lambda)^2/2$. By keeping the lowest two orders in the Taylor series expansion, δ_1 is approximately equal to $\phi I_r(2b - 4b^2)/(a + b)$, where $\phi = \pi un_o d/\lambda_0$. The reduced effective fields for maximum diffraction efficiency can be obtained as

$$b = b_{dm} = \{\phi a I_r - \delta_{1m}/2) \pm [(\phi I_r - \delta_{1m}/2)^2 - 4a\delta_{1m}\phi I_r]^{1/2}\}/4\phi I_r. \quad (6)$$

The difference of these two peak fields is $[(\phi I_r - \delta_{1m}/2)^2 - 4a\delta_{1m}\phi I_r]^{1/2}/2\phi I_r$.

In order to illustrate the double-peak phenomena, δ_1 and η versus the effective reduced field b calculated from relations (2)–(5) are shown in Fig. 1 with $d = 110$ μm, $\Lambda = 132$ μm, and a fixed reduced optical field $I_t/I_{\mathrm{th}} = 0.05$. The parameters used in all calculations are $r_1 = 0.595$, $r_2 = 0.405$, $n_o = 1.57$, $n_e = 1.81$, $|\Delta\epsilon| = 0.5$, $K_1 = 7.125 \times 10^{-7}$ dyn, $K_2 = 4 \times 10^{-7}$ dyn, and $K_3 = 7.5 \times 10^{-7}$ dyn. As one can see from Fig. 1, before the threshold condition ($b = 0$) is attained, δ_1 is zero. As b is raised slightly, and so is the biasing field, the phase-modulation amplitude δ_1 increases rapidly, which reflects the critical behavior in this reorientational transition, to a maximum value $\delta_{\max}$, and then decreases gradually while b is increasing. The diffraction efficiency η is monotonically increasing with δ_1 if $\delta_1 \le \delta_{1m}$. When δ_1 exceeds δ_{1m}, η decreases, then reaches its local minimum at $\delta_1 = \delta_{\max}$, where $\Delta\bar{n}_{\mathrm{NL}}$ has its maximum value (=0.002). As δ_1 decreases from $\delta_{\max}$ and passes δ_{1m} again, the second peak occurs.

The dependence of the peak field b_{dm} on the reciprocal of the grating period is illustrated in Fig. 2(a). It is obvious that there are two regimes, double peak and

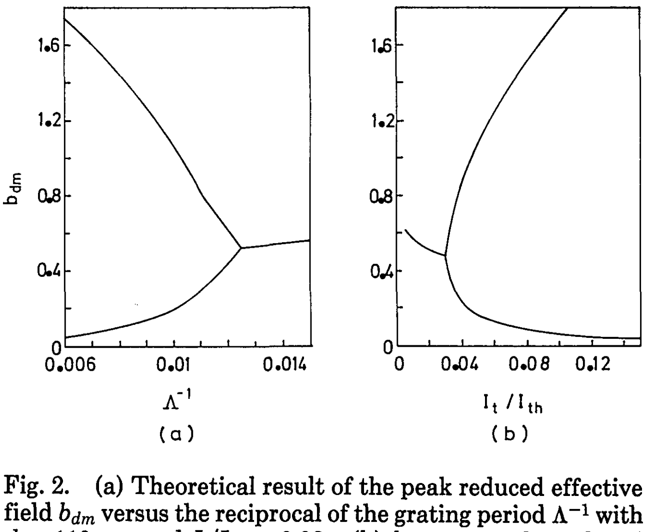

Fig. 2. (a) Theoretical result of the peak reduced effective field b_{dm} versus the reciprocal of the grating period Λ^{-1} with $d = 110$ μm and $I_t/I_{\mathrm{th}} = 0.08$. (b) b_{dm} versus the reduced optical field I_t/I_{th} with $d = 110$ μm and $\Lambda = 132$ μm.

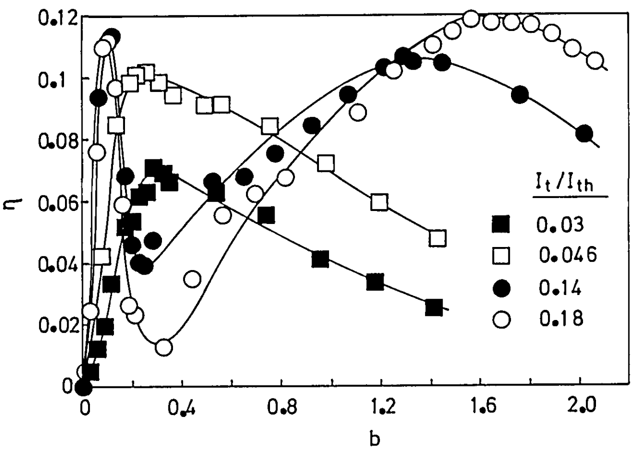

Fig. 3. Experimental results of the diffraction efficiency versus the reduced effective field for various optical intensities. The solid curves are only a visual aid.

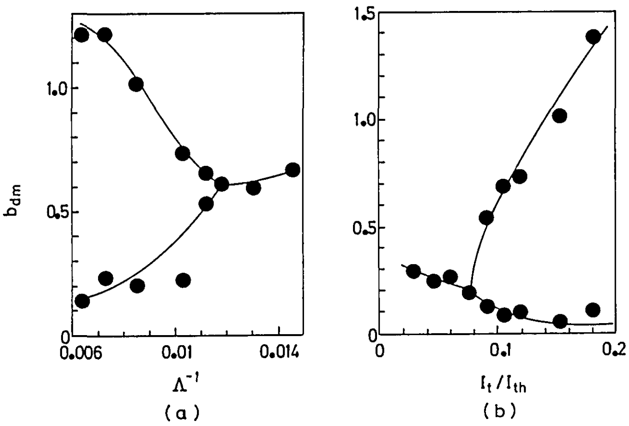

Fig. 4. (a) b_{dm} versus Λ^{-1} with d = 110 μm and $I_t/I_{\rm th}$ = 0.209; $I_{\rm th}$ and $V_{\rm th}$ are determined to be 3.74 V and 325 W/cm^2, respectively. (b) b_{dm} versus $I_t/I_{\rm th}$ with d = 92 μm and Λ = 146 μm; $I_{\rm th}$ and $V_{\rm th}$ are determined to be 464 W/cm^2 and 3.74 V, respectively. The solid curves are only a visual aid.

single peak, which correspond to the situation of $\delta_{\rm max}$ greater or less than the optimal phase amplitude δ_{1m}, respectively. The critical point where two peaks merge reveals that $\delta_{\rm max}$ is equal to δ_{1m}. In the double-peak regime, one can see that the larger the reciprocal of the grating period is, the smaller the separation of the two peaks becomes, as predicted in Eq. (6). This can be explained by the twist-deformation effect discussed in our previous paper,[12] i.e., the term $(K_2/K_3)(2d/\Lambda)^2$ in relation (2), since the smaller the grating period is, the less deep the grating modulates. According to the behavior of the phase–amplitude curve shown in Fig. 1, as the whole curve is suppressed owing to the decrease of the grating modulation, the separation of the two points with $\delta_1 = \delta_{1m}$ is decreased. Similar behavior is shown in Fig. 2(b) for the dependence on the pump intensity, and it can be understood that a stronger pump permits a deeper phase modulation.

The detail of this experiment is essentially the same as described previously,[6] and typical results are shown in Figs. 3 and 4. A fresh sample of N-(p-methoxybenzylidene)-p-butylaniline with d = 92 μm and Λ = 146 μm is used in Fig. 3. The threshold fields $V_{\rm th}$ and $I_{\rm th}$ are determined to be 3.74 V and 464 W/cm^2, respectively. It can be seen that the diffraction-efficiency curves increase as the pump is increased. With stronger incident intensities, double peaks occur and spread out accordingly. The local minimum that corresponds to the maximum phase $\delta_{\rm max}$ is lowered to 10% of the maximum value for $I_t/I_{\rm th}$ = 0.18. The maximum efficiency η_m has an average value of 11%, which is smaller than that predicted by theoretical calculation. Nevertheless, it is reasonable since a large part of the beam energy is contained in the spurious background scattering.[6] The dependence of the peak occurrence on Λ^{-1} and I_t is shown in Figs. 4(a) and 4(b), respectively; these curves show the same behavior as the numerical results in Figs. 2(a) and 2(b), respectively. The conditions needed for the critical point, i.e., $\delta_{\rm max} = \delta_{1m}$ = 2.075 are Λ = 84 μm and $(V/V_{\rm th})^2$ = 1.39 for d = 110 μm and $I_t/I_{\rm th}$ = 0.209, as shown in Fig. 4(a).

In conclusion, we have illustrated for the first time, to our knowledge, that the maximum efficiency of the fundamental diffraction through the DFWM process can be obtained under two distinct biasing fields. As the grating period or pump intensity increases, the difference of these two fields increases accordingly, as predicted by the analytical solution. The critical point where the two peaks merge, corresponding to a specific phase amplitude $\delta_{\rm max} = \delta_{1m}$, is pointed out. Prominent double peaks of the diffraction efficiency are shown by both experimental and numerical results.

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