Nonlinear light scattering by laser- and dc-field-induced molecular reorientations in nematic-liquid-crystal films

I. C. Khoo
Department of Physics and Astronomy, Wayne State University, Detroit, Michigan 48202
(Received 6 April 1981)

We present here a detailed experimental study of nonlinear optical scattering in nematic liquid crystals using low-power cw lasers. The scattering is effected by the phase grating created by two intersecting same-frequency lasers. These lasers interfere spatially to create an index modulation via their reorientation effects on the molecules. When aided by a dc magnetic field, the reorientation and nonlinear responses of the medium are enhanced. We have quantitatively measured the dependence of the diffraction efficiency as a function of the optical intensity and the magnetic field ranging from well below to far above the critical orientation threshold values. The scattering is also analyzed in terms of its dependence on the temperature, sample thickness, and the scattering geometry. The experimental results are in good agreement with theoretical expectations. We also point out some high-intensity effects and applications of this phase grating, e.g., in wave-front conjugation and holographic imaging.

I. INTRODUCTION

Light scattering properties of liquid crystals have been the subject of extensive fundamental and applied research.[1–10] It is well known that the responses of liquid crystals are extremely dependent on the temperature and the corresponding phase. In particular, it has been shown[8] that the nematic phase is characterized by a scattering (linear) cross section about a million times that of the isotropic phase. Furthermore, nematic films of small thicknesses and appropriate molecular alignments can be made[11] that will eliminate or reduce the unwanted spurious scattering associated with the random director-axis fluctuations. In light of this, it is perhaps surprising to note that nonlinear processes in the nematic phase are relatively unexplored. In our recent studies,[3] aimed at utilizing the much larger scattering ability of nematics, we have demonstrated that, indeed, four-wave mixing can occur with very high efficiency in a cross-beam experiment using low-power cw lasers.

The phase grating responsible for this wave-mixing experiment is the optically induced collective molecular reorientations, which can be further enhanced by an applied dc magnetic field. In the low optical intensity and small reorientation limit, the third-order nonlinearity can be analytically estimated to give a so-called third-order nonlinear susceptibility on the order of 10^{-3} esu. The observed diffraction efficiency, i.e., the ratio of the generated fourth wave to the input probe beam, was experimentally measured to agree with this value. For a pump beam intensity on the order of 10 W/cm^2, a diffraction efficiency of about 10^{-2} was obtained.

These preliminary studies clearly mandate further investigations. In this paper, we present the results of a detailed study of crossed-laser-beam experiments on nematic-liquid-crystal films. In particular, we have measured the diffraction from the phase grating generated by the crossed beams as a function of the optical and the applied dc magnetic fields. The magnetic field strengths used range from below the threshold fields (to create molecular reorientation) to well above the threshold. Since the primary molecular characteristics, i.e., the susceptibility anisotropies and the elastic constants (for bend, twist, and splay) involved in the molecular reorientation are known to be dependent on the temperature, the temperature dependence of the nonlinear diffraction is also studied. Effects associated with other parameters, such as geometry and sample thickness, will be discussed in the appropriate sections.

II. GENERAL THEORY

The relevant theory for our present study pertains to dc- and optical-field-induced molecular distortions and phase-grating formation due to two

crossed laser beams. Most of these theories have been worked out in the literature in various contexts.[3,4,11–14] We present a general review which serves as a convenient reference, and point out where new and important features will arise in the present context of nonlinear optical scattering. We are interested in the spatially varying dielectric constant as induced by two optical fields, in conjunction with an applied dc magnetic field. (An applied dc electric field could also be used, but its reorientational effect is equivalent to that by a magnetic field.) In the scattering geometry as shown in Fig. 1, the incident optical-field polarization is parallel to the magnetic field, and perpendicular to the director-axis unperturbed direction (i.e., the director axis is normal to the boundary planes of the sample for a homeotropically aligned nematic film). It is known that for this particular initial director configuration (i.e., $\theta=0$ at $Z=0$ and at $Z=d$), there is a threshold value of the applied field, below which θ is zero and above which θ is finite. Because of the dielectric anisotropy, the effective dielectric constant for optical propagation in the medium and the ability of the optical field to further perturb the molecular orientation (and create nonlinear scattering), clearly depend on the tilt angle. The problem of molecular distortion under the action of a dc field has been solved many times in the literature. Recently, Herman and Serinko[4] have included the effect of the optical field in conjunction with the dc magnetic field by defining an effective reorienting field. In a separate endeavor,[3] we have calculated the molecular reorientation by a purely optical field.

These calculations are by no means complete, since they are valid only for the region of field strength just above the threshold, when the tilt angle is assumed to be small. Under a large dc magnetic field that is substantially above the threshold (or so-called Freedericksz[12]) field, there are important new features. Above the threshold θ will rapidly assume a large value, whence an analytic expression for the phase grating (and therefore the wave-mixing efficiency) cannot be obtained. Nevertheless, the dependence of θ on the field strength can, and has been, numerically solved before, and will therefore enable us to analyze our experimental results.

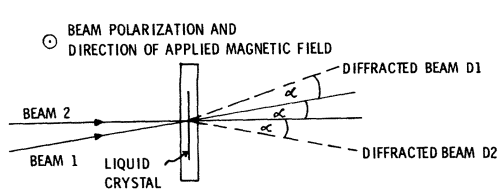

FIG. 1. Schematics of crossed-beam scattering geometry. The director-axis reorientation is in the XZ plane.

In brief, nematic molecular reorientation may be treated by solving for $\hat{n}(\vec{r})$ that will minimize the free energy

$$F=\frac{K}{2}\{[\vec{\nabla}\cdot\hat{n}(\vec{r})]^2+[\nabla\times\hat{n}(\vec{r})]^2\} -\nabla\chi[\vec{H}\cdot\hat{n}(\vec{r})]-\frac{\Delta\epsilon}{4\pi}[\vec{E}_{op}\cdot\hat{n}(\vec{r})]^2 , \quad (1)$$

where K is the elastic constant. (This is the so-called one-constant approximation.) For the homeotropically aligned system, the value for the elastic constant K should be that for bend distortion.

The application of the Euler-Lagrange equation immediately yields the familiar sine-Gordon equation for the torque balance:

$$\xi^2\frac{d^2\theta}{dz^2}+\sin\theta\cos\theta=0 , \quad (2)$$

where

$$\xi^2=\frac{4\pi}{E_{\text{eff}}^2}\frac{K}{\Delta\epsilon} , \quad (3)$$

and we have defined a total effective field E_{eff} analogous to Ref. 4:

$$E_{\text{eff}}^2=E_{\text{op}}^2+4\pi\frac{\Delta\chi}{\Delta\epsilon}H_{\text{dc}}^2 . \quad (4)$$

The optical field E_{op} comprises two same-frequency lasers propagating at two slightly crossed directions:

$$\vec{E}_{\text{op}}=\vec{E}_1\exp i(\vec{k}_1\cdot\vec{r}-\omega t) +\vec{E}_2\exp i(\vec{k}_2\cdot\vec{r}-\omega t) . \quad (5)$$

In Eq. (4), $\Delta\epsilon=\epsilon_{\parallel}-\epsilon_{\perp}$ is the dielectric anisotropy at (*at optical frequency*), where the symbols $\epsilon_{\parallel}$ and $\epsilon_{\perp}$ have their usual meaning. Similarly, $\Delta\chi$ is the magnetic anisotropy. For the liquid crystal MBBA (p-methoxybenzylidene-p'-n-butylaniline) both $\Delta\epsilon$ and $\Delta\chi$ are positive.

Equation (2) predicts the existence of a threshold (sometimes termed Freedericksz) field value E_f (or equivalently H_f, with the replacement of $\Delta\epsilon/4\pi$ by $\Delta\chi$ in the following expression):

$$E_f=\left[\frac{4\pi K}{\Delta\epsilon}\right]^{1/2}\frac{\pi}{d} \quad (6)$$

above which the molecule's tilt angle θ (which is initially equal to zero) is finite. In Refs. 3 and 4, the effective dielectric constant for applied fields just above the threshold have been analytically approximated to give a nonlinear dielectric constant.

$$\epsilon_{nL} \simeq \begin{cases} \left[\dfrac{\epsilon_\perp \Delta\epsilon}{\epsilon_{||} E_f^2}\right] E_{\rm op}^2 \quad \text{(purely optical field)}\,, & \text{(7a)} \\ \left[\dfrac{\epsilon_\perp (\Delta\epsilon)^2}{\epsilon_{||} 4\pi\Delta\chi H_f^2}\right] E_{\rm op}^2 \quad \text{(strong magnetic field)}\,. & \text{(7b)} \end{cases}$$

[For $H=0$ and for a tilted geometry (i.e., where the optical fields are not normally incident on the sample), it has been shown theoretically in Refs. 4 and 18 that the optically induced nonlinearity is also of the form given in (7a), multiplied by a geometrical factor. The importance of the geometrical factor is discussed in the Appendix.] This dielectric constant is obtained by approximating the exact expression for the effective dielectric constant $\epsilon_{\rm eff}$ obtained by solving Maxwell's equation

$$\epsilon_{\rm eff} \frac{\epsilon^2 - (\frac{1}{2}\Delta\epsilon)^2}{\epsilon - \frac{1}{2}\Delta\epsilon \cos 2\theta} \quad \epsilon = \tfrac{1}{2}(\epsilon_\perp + \epsilon_{||}) \tag{8}$$

in the limit $\theta << 1$.

For applied fields sufficiently above the threshold (e.g., 1.5 times), the molecular reorientation angle quickly assumes a large value and the dependence of the nonlinear index on the applied field is very complicated. The solutions for θ have been shown to be generalized Jacobian sine amplitude functions,[12] for which analytic expressions of ϵ_{nL} for high fields are not easily obtainable. However, a simple but instructive picture may be gained by examining the numerical solutions for θ. First, by symmetry consideration, θ is maximal at $Z=d/2$ (i.e., $\theta=\theta_m$ at $Z=d/2$). The equation for θ_m, and also for θ as a function of Z, can then be numerically integrated from Eq. (2). It suffices to consider only θ_m, since θ differs from θ_m only by a smoothly varying factor. Figure 2 shows a plot of θ_m as a function of the normalized field. θ_m is zero below E_f, rises abruptly above E_f with a typical critical behavior, and subsequently "saturates" to a maximum angle of 90° at very high fields. The susceptibility of the molecule to optical perturbation (i.e., the change of θ with respect to the external field) is reflected by the slope $d\theta/dE_{\rm op}$. We note here that $d\theta/dE_{\rm op}$ is particularly large just above the threshold, and therefore expect that the nonlinear optical responses of the molecules are largest just above the threshold. As the external field is increased, it begins to drop considerably (as depicted in the insert in Fig. 2). At above twice the threshold field, the variation of the molecular orientation with the external field is almost vanishing. Although a detailed numerical computation of the nonlinearity for various values of the applied field may appear desirable, we will content ourselves with these observations and their experimental confirmations.

For the present context of a phase-grating and nonlinear diffraction, we are interested only in the term involving the quadratic interference between the two incident beams:

Since the threshold field is a function of the sample thickness ($H_f \propto 1/d$), and the two key molecular anisotropies $\Delta\epsilon$ and $\Delta\chi$ are dependent on the temperature, we have also measured the temperature and the sample size dependences of the nonlinear diffraction efficiency.

III. EXPERIMENTAL DETAILS

The liquid crystal used was MBBA. The sample was prepared by the usual method of sandwiching

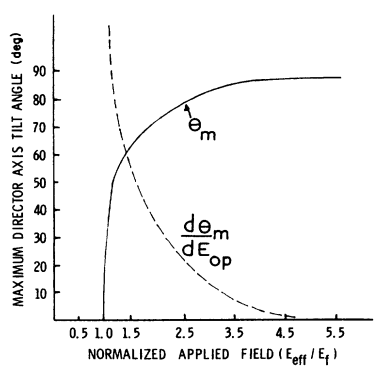

FIG. 2. Numerical solution of θ_m with the applied field. Dotted line shows qualitative dependence of $d\theta/dE_{\rm eff}$.

the nematics between two flat glass slides that had been treated with the surfactant HTAB (hexadecyltrimethylammonium bromide). The sample was sealed and was checked (visually by experience, and also by the optical conoscopic interference method[3]) to be uniformly homeotropic over a period of months. In practice, samples of thicknesses ranging from 50 to 100 μm were shown to be most desirable in terms of the signal to spurious side-scattering noise ratio, and the stability in alignment. The experimental setup is shown in Fig. 3. A single-line (5145 Å) argon laser was divided into two beams and recombined at a small angle on the sample. This angle is small (about 0.8°), and therefore phase mismatch is not important. For magnetic field study, the sample was placed in an electromagnet capable of 0.5-G fine tuning. From our experience with holographic imaging, the path lengths of the two beams should be carefully adjusted to maximize the interference modulations on the sample to ensure good diffracted signal, since the laser is operating in multimode. Temperature dependence of the scattering was measured by placing the sample in a temperature cell that allowed tuning from below the crystal nematic transition (~ 10 C) to above the nematic-isotropic temperature (~ 40 C). We have used samples of varying degrees of purity. The temperature effects associated with contaminants are quite drastic (a well-known property in chemistry). In all cases, the diffracted beams are clearly visible for incident laser beams on the order of 0.1 W in beam 1 and 0.5 W in beam 2. Both the intensity and the intensity distribution of all the exit beams are carefully monitored since we have observed self-focusing effects (Ref. 3) at high enough optical intensity.

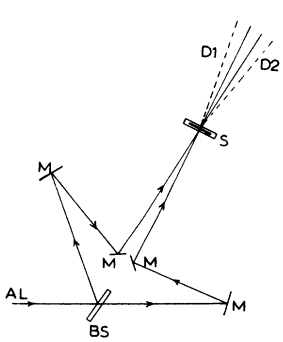

FIG. 3. Experimental setup. *BS*: beam splitter, *m*: mirror, *s*: sample, *AL*: argon laser, *D*1 and *D*2: diffracted beams.

IV. RESULTS AND ANALYSIS

As we mentioned earlier, the field-induced molecular reorientation depends on the interplay between the dc magnetic and the optical fields. We shall separate the discussion into three distinct cases: (a) $H=0$ and (b) strong magnetic field, defined by

$$\Delta\chi H_{\rm dc} >> (\Delta\epsilon/4\pi)(E_{\rm op})^2 ,$$

and (c) strong optical and magnetic fields.

Preliminary results for case (a) have been reported before. We report here on some of the newer observations. It has been observed that the diffraction is highly dependent on the relative tilt of the director axis with respect to the optical propagation as specially pointed out in the Appendix. Figure 4 shows the diffracted intensity as a function of the incident (undivided) argon laser intensity I_L for a tilting angle β of 0.16 rad. In the low-optical-intensity limit, the diffracted intensity obeys a four-wave—mixing picture very well, varying as the cubic function of I_L.

At high optical intensities, deviations from the cubic dependence is observed (cf. Fig. 4). At the same time, the beams also appeared to self-focus.[15] The exit beam diameter began to decrease with increasing intensity and subsequently shrunk to a

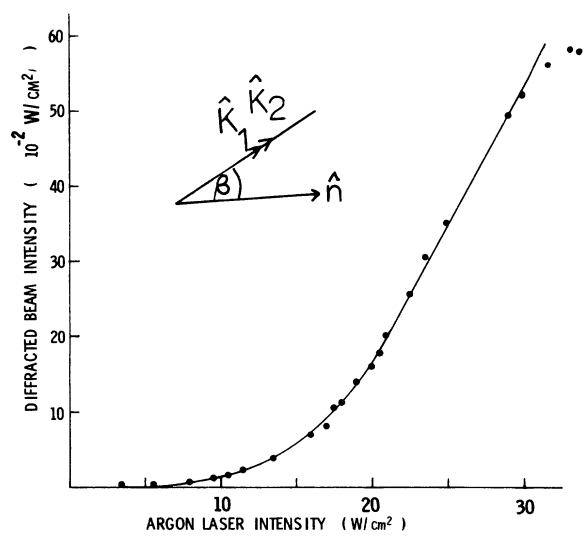

FIG. 4. Dependence of the diffracted intensity on the argon laser intensity in the absence of a magnetic field. Deviations from the four-wave—mixing cubic dependence are observed at a high optical intensity. Insert shows the relative orientation of the optical propagation vector, the optical field, and the nematic axis.

filamentlike beam of about 0.1 mm at the region of smallest beam waist. These occurred at an input beam power of about 2.2 W (for an input beam diameter of 2 mm) in a 100-μm thick sample. Self-focusing of light is an important subject in its own right which we will reserve for a detailed quantitative report in a future publication. It is important to note here that occasionally for samples subject to a much higher intensity than that mentioned above, optical illumination can give rise to damaging heating effects, whence the exit beams are very seriously distorted, accompanied by very wild spurious scatterings. These thermal effects are easily distinguishable from the high-intensity self-focusing and multiorder diffraction effects mentioned earlier.

The dependence of the diffraction efficiency on the sample thickness was noted to be very drastic. A 75-μm thick sample produced an easily visible diffraction at an incident optical intensity of about 10 W/cm^2, while a 50-μm thick sample produced almost vanishing diffraction. On the other hand, a 100-μm thick sample gave a diffraction more than 4 times that of the 75-μm thick sample. From (7a) or (7b) and (6) one can show that ϵ_{nL} varies as d^2. Thus, the diffracted field amplitude in the coherent wave-mixing process will vary as d^3, and the diffracted intensity will vary as d^6. Our measurements were not precise enough for checking this dependence, but our results clearly indicated that the diffracted intensity varied much faster than a d^2 and was somewhat similar to a d^6 dependence.

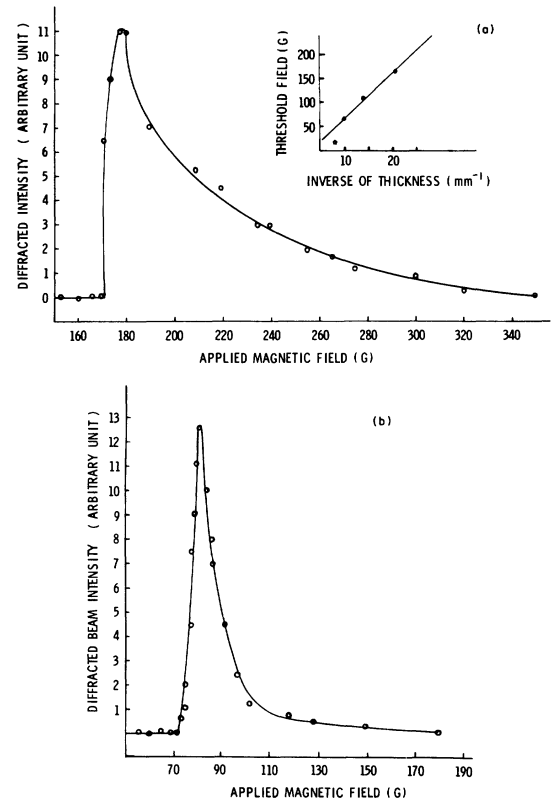

FIG. 5. Dependence of the diffracted intensity as a function of the magnetic field for two sample thicknesses. (a) $d=50$ μm and (b) $d=100$ μm. Insert in (a) shows the $1/d$ dependence of the Freedericksz field. Experimental conditions: $I_1=2.5$ W/cm^2 and $I_2=1.5$ W/cm^2. Peak diffraction efficiency (diffracted intensity/I_2) in percentage is 3% for (a) and 10% for (b).

A. Strong magnetic field

When a magnetic field was present and was strong enough to create molecular reorientation, the wave-mixing efficiency increased dramatically because of the larger effective reorienting field. Clearly visible diffracted beams were observed at a very low incident optical power (typically about 0.1 W, which in the absence of the magnetic field *would not* be able to generate visible diffractions). In this limit, the diffraction grating generated by the two incident beams scattered these beams in the two phase-matched directions (with appropriate intensity dependence) in accordance with the usual degenerate four-wave—mixing process (phase-grating diffraction). Beam D_1 varies as $I_1^2 I_2$, while beam D_2 varies as $I_2^2 I_1$. The ratio of the intensity of beam D_2 to beam D_1 goes as I_2:I_1 which was experimentally confirmed. Figures 5 show the typical diffraction efficiency (beam D_1) for samples of thicknesses $d=50$ μm and 100 μm, respectively. For all cases, the diffraction efficiency showed abrupt increases at very well-defined threshold fields (to within 0.5 G). These threshold fields were found to be exactly the same as those obtained by the optical conoscopic interference method, and they obey the d^{-1} dependence to within the accuracy in our measurements of the thickness [the insert in Fig. 5(a)]. The measured values of H_f are also in good agreement with those estimated from Eq. (6) (with $\Delta\epsilon \rightarrow \Delta\chi$).

The dependence of the diffraction efficiency on the magnetic field strength followed the predicted dependence from our theoretical analysis. Just above threshold, the molecules are particularly susceptible to further optical perturbations ($d\theta/d\epsilon_{\rm op}$ is large), and therefore the diffraction efficiency is very high. Depending on the sample thickness, the

diffraction efficiency decays to vanishing values at higher fields. If we examine these curves in terms of the normalized field strength (i.e., H/H_f), we note that the diffraction efficiencies decay to an almost vanishing value at $H \simeq 2H_f$, in agreement with the expected dependence as discussed in Sec. II. The results obtained for a 75-μm thick sample (not shown in these figures) are also consistent with these observations.

B. Strong magnetic and optical fields

At a higher optical intensity (about 30 W/cm^2) (i.e., visible diffractions were observed at zero magnetic field), the dependence of the diffraction efficiency on the magnetic field is similar to that given in Fig. 5, with a noted difference that some residue diffractions still persisted even at a high magnetic field (1500 G). We attribute these residue diffractions and the diffractions at zero magnetic field to be probably caused by the thermal grating effect (see Appendix and Ref. 18). The absolute magnitude of these diffractions is difficult to measure because of extremely large spurious side scatterings but is estimated to be comparable to the value at zero magnetic field.

We have also studied the case where the magnetic field is transverse to the optical polarizations with $\beta = 10°$ (cf. Fig. 4 insert). Figure 6 shows the dependence of the diffraction efficiency. One notes here that the diffraction efficiency abruptly drops to almost half its initial value at $H = H_f$, and proceeds to decrease very slowly at higher fields. The drop at $H = H_f$ is associated with the

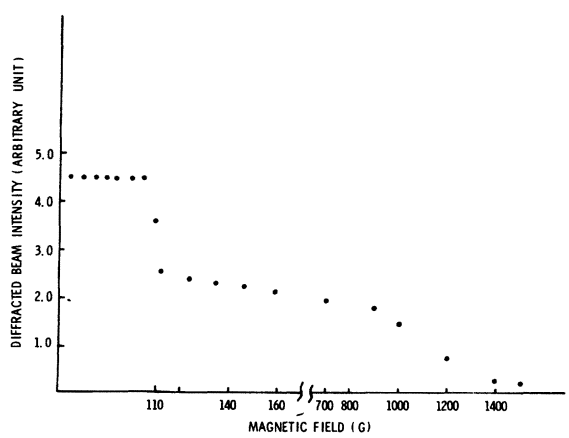

FIG. 6. Dependence of the diffracted intensity on a magnetic field perpendicular to the laser polarization. Diffraction at $H = 0$ is due to wave mixing in a tilted ($\beta \neq 0$) geometry.

reordering of the molecules in the direction of H, making it less susceptible to reorientation by the optical fields. More quantitatively speaking, the optical fields have to overcome both the molecules' elastic restoring force and the magnetic torque. It is interesting to note that the effect due to the magnetic field is only evident above the critical value H_f.

At a much higher magnetic field, the diffraction efficiency continues to decrease smoothly. At an applied field of 1500 G, the residue diffraction is found to be about 10% of the initial value.

For nematic films above the Freedericksz transition, and even in the case of purely optically induced ordering, the resultant optical nonlinearity is extraordinarily large, perhaps the largest of all known materials. In the weak optical intensity limit (corresponding to roughly the appearance of only the first-order diffracted beam and cubic dependence on the laser power), when the definition of a third-order susceptibility $\chi^{(3)}$ is valid, an experimentally observed value of $\chi^{(3)}$ of more than 10^{-3} esu is obtained. This is calculated based on the observed diffraction efficiency (I_D/I_2) of 10^{-2}, with an incident laser beam of intensity 10 W/cm^2 (for the case of purely optically induced orientation at $\beta = 0.16$). The diffraction efficiency is, in general, about an order of magnitude larger when a Freedericksz magnetic field is applied. Quantitative comparison of these observed wave-mixing efficiencies with theoretical expressions [e.g., Eqs. (7)] shows that the experimentally observed values are about three orders of magnitude lower in the case of purely optically induced effect ($H = 0$) and about two orders in the case of magnetic Freedericksz transition. The discrepancy of this magnitude is common among all four-wave–mixing experiments, and may be attributed to numerous factors (nonideal wave-mixing conditions, approximations made in the theoretical derivations and uncertainties in the actual values of the anisotropies . . . , etc.). In the case of the purely optically induced effect, the observed diffraction efficiency is lower also because we have not accounted for the geometry factor (dependence on β) in the theoretical estimate [which is based on (7a)]. If the geometrical factor is taken into account (see Ref. 4 or 18), the experimental and theoretical values are closer. In the present study, scattering loss is also a significant factor. Nevertheless, the excellent agreement of the thickness, the geometry, and the field *dependence* with theoretical expectations is well demonstrated.

V. TEMPERATURE DEPENDENCE

Since the physical properties of liquid crystals are highly dependent on the temperature, no study of liquid crystals is complete without a serious look at the temperature dependence. The dependence of the nonlinear scatterings (via the collective molecular reorientations) on the temperature is, however, not as clear-cut as the dependence on the magnetic field. In general, as the temperature is raised, the molecular anisotropies $\Delta\epsilon$ and $\Delta\chi$ as well as the elastic constant K will all be changed so that only qualitative dependence may be deduced. An instructive picture may be gained nevertheless, from the expression for the diffraction efficiency for small tilt angle θ and for a low optical field. In this limit, the diffracted (e.g., beam D_1) intensity can be shown to be given by

$$I_D \sim \frac{\Delta\epsilon^2}{K} I_1^2 I_2 \tag{8}$$

for both optical as well as optical-plus-dc-field-induced molecular reorientation, following the analysis of Refs. 3 and 4 [cf. Eqs. (7)]. (It may be deduced from Ref. 4 that the diffractions for a tilted geometry is also of this form, except for a geometrical factor which will be a constant throughout the temperature dependence experiment.) For simplicity, we shall concern ourselves with the case involving only the optical field. In this case, the two temperature-dependent variables are K and $\Delta\epsilon$: In the mean-field theory,[16] $\Delta\epsilon$ is proportional to the order parameter S, and K is proportional to S^2. Thus, while S is strongly temperature dependent, $\Delta\epsilon^2/K$ should not be (a fact that is well recognized in linear light scattering).[8] However, as one approaches the nematic-isotropic transition, we would expect the mean-field theory to break down. In more practical terms, we also expect that because of the dramatic decrease in the elastic constant K, the sample alignment will be severely disturbed and change the wave-mixing efficiency drastically.

Figure 7 shows a typical temperature dependence of the diffraction for a fairly pure sample in the absence of a magnetic field. Throughout the greater part of the nematic range the diffraction efficiency is almost flat. The less pure sample undergoes a phase transition at a lower temperature than does the purer one. In all cases, the diffraction efficiency increases dramatically near the transition temperature. The system, however, is not stable, and the diffraction efficiency often deteriorates if the sample is maintained near the transition temperature for a sufficiently long time. It is

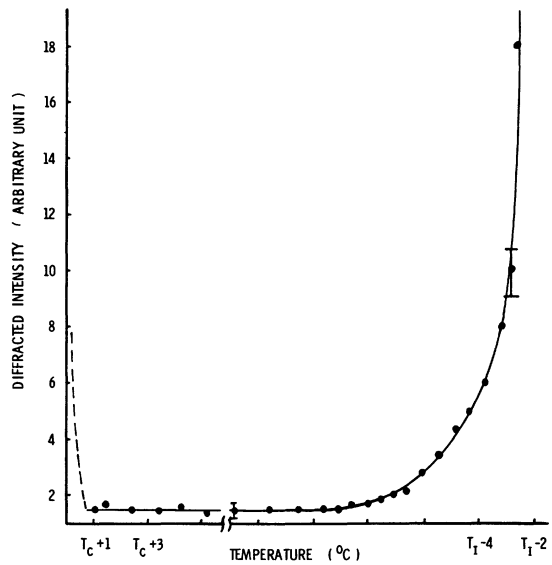

FIG. 7. Temperature dependence of the diffracted intensity for a 75-μm thick sample. Dramatic increases in the diffracted intensity occur near the nematic $\rightarrow$ isotropic and the nematic $\rightarrow$ crystal transitions T_i and T_c, respectively.

well known that purer samples (e.g., samples prepared under absolutely dry environment and free from contaminants from the surfactant treatment) are more stable at high temperatures, and we will not delve into this. More importantly, we note here that the heating from the incident laser beams are not appreciable (up to an intensity of 25 W/cm^2) as far as this dependence of the diffraction efficiency on the temperature and the nematic-isotropic transition point are concerned. As noted in Pershan's[9] work, e.g., the heating generated by a laser intensity of more than 50 W/cm^2 lowers the nematic-isotropic transition by only one or two degrees. The lowering of the transition is determined in our experiment to be mainly due to contaminants. In the other temperature extreme, i.e., approaching the nematic crystal transition, we also notice a very dramatic increase in the wave-mixing efficiency just before crystallization takes place. These results near the two transition points are expected from the large fluctuation in the molecules' orientations accompanying a phase transition. On the other hand, the thermal grating effect is also expected to become large near T_I, although it is shown to be small at room temperature in some recent work,[18] for $\beta \geq 0.15$. A detailed study of these temperature effects is rather complicated and is relegated to a future endeavor.

VI. CONCLUSION AND FURTHER REMARKS

We have presented a fairly complete experimental study and theoretical analysis of the nonlinear optical diffractions that are generated by crossed laser beams in a nematic materials. Although our study has also opened new questions, the qualitative and some quantitative agreement between theory and experiment as described are notable. The critical and large nonlinearity of nematics near (above) a dc Freedericksz transition is well demonstrated. The dependences on the geometry, on the thickness, and the temperature have provided further understanding and confirmation. The large nonlinearity of nematic liquid crystals gives rise to easily observable effects which, besides providing an excellent ground for studying fundamental properties of nematics, are of great potential application. Some of the results obtained here are clearly oriented towards the practical device application. A natural next step in our study is to use nematic film for wave-front conjugations and holographic imagings, which have been successfully conducted by this investigator.[17] Specifically, we have observed large, and easily processed conjugated signals by retroreflecting beam 1 and using a beam splitter in the path of beam 2 to view the conjugated signal and its intensity distribution. Phase distortion in the path of the object beam (beam 2) was found to be compensated. Currently, other holographic imaging studies are underway.

ACKNOWLEDGMENTS

The author acknowledges frequent constructive discussions with Professor Emmett Leith of the University of Michigan on phase-grating problems, and the technical assistance of the visiting scholar Mr. Shu-Lu Zhuang from the People's Republic of China. This research was supported by a grant from the Research Corporation, and a grant from the National Science Foundation under Contract No. ECS 8026775.

APPENDIX

In this section, we comment on some important differences associated with optical fields that are incident on the sample with non-normal incidence. From well-known dc-field-induced distortion on nematics (and also the theory given in Ref. 18), there is no threshold requirement on the (optical) electric field strength to induce finite molecular reorientation. This was indeed borne out in our experiment (Sec. IV A). For optical intensities on the order of 10 W/cm^2, there is no observable diffraction for strictly normal incidence ($\beta=0$). However, easily visible diffraction is observed for slight deviation from the normal ($\beta \gtrsim 0.15$) and increases in magnitude as β is increased as reported in Sec. IV A.

It is important to note here that these optical intensities required to produce diffractions are much lower than the so-called optical Freedericksz threshold intensity I_f $[I_f=(C/8\pi)E_f^2]$. For a 75-μm thick sample, and for MBBA, typically $\Delta\epsilon=0.4$ (not 0.1 as used in Ref. 3) and $K=10^{-7}$. This gives $I_f \approx 100$ W/cm^2, which is much larger than the intensities used here (and in Ref. 3) to generate observable diffractions. In retrospect, and reviewing the experimental conditions for Ref. 3 (where an *almost normal* incidence was used), it is likely therefore that the observed nonlinear diffractions as reported in Ref. 3 and in Sec. IV A pertains to a tilted geometry. Visible diffractions are generated for strictly *normal incidence* ($\beta=0$) but the required optical intensities are on the order of 50 W/cm^2. As reported before, self-focusing and irregular transmitted intensity patterns begin to appear at these intensities, making quantitative measurements very difficult.

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