PHYSICAL REVIEW A VOLUME 19, NUMBER 4 APRIL 1979

Nonlinear-optical processes in nematic liquid crystals near Freedericks transitions

R. M. Herman and R. J. Serinko
Department of Physics, The Pennsylvania State University, University Park, Pennsylvania 16802
(Received 19 October 1978)

Weak-beam amplification and diffracted-beam production are predicted under a variety of conditions, in which thin nematic-liquid-crystal samples maintained in an external magnetic field near Freedericks transitions are simultaneously illuminated by strong and weak beams derived from the same light source. It is shown that for 4-methoxybenzylidene-4'-butylaniline, substantial weak-beam amplification should occur under continuous illumination for modest (~ 0.1 W/cm^2) strong-beam intensities, for certain cell and beam geometries. If a small positive frequency difference between the strong and weak incident beams is maintained, weak-beam amplification is further enhanced, and the conditions on the angles of incidence for amplification are considerably relaxed.

I. INTRODUCTION

In recent years, nonlinear optical properties of nematic-liquid-crystal materials have been studied by a number of authors in an attempt to gain information on electronic and orientational contributions to nonlinear optical susceptibilities and to obtain relaxation rates and transport properties related to the orientational motions of the individual molecules.[1-4] Studies have been carried out both above and below the nematic-isotropic transition in an effort to understand the properties of these materials near the phase transition. In the nematic phase, also, low-intensity light scattering has been employed[5,6] to reveal the presence of a very narrow, intense Rayleigh component, corresponding to scattering from the director orientational fluctuations, which relax very slowly, in addition to at least two broader components (comprising the so-called Rayleigh wing) arising from fluctuations of the individual molecular alignments about the director. It is this narrow component that is primarily related to the self-focusing property of nematics[7] under continuous illumination. Other (nonorientational) slowly relaxing excitations in nematics have also been studied recently.[8] These have been ascribed to thermal- and mass-diffusion effects in liquid crystals.

In the present paper we shall again concentrate on the slowly varying components of molecular anisotropy that are related to the behavior of the director in the nematic phase. Our interest lies with nonlinearities that appear for relatively low light intensities. In seeking a method for enhancing the nonlinearities at low optical intensities, we shall consider the nematics as existing near Freedericks transitions associated with their placement in externally applied constant magnetic fields.[9-11] [We confine our interest to magnetic- (as opposed to electric-) field-induced transitions, for purposes of ultimate experimental expediency.] To obtain these transitions, a thin sample of nematic material is placed between parallel glass cell walls, treated such that at the walls the nematic director field is maintained in a fixed direction perpendicular to the applied magnetic field. Accordingly, at low fields, the director field is uniform throughout the sample, in that there is no systematic magnetic torque on the sample in its equilibrium configuration and for fluctuations away from equilibrium, elastic restoring torques exceed magnetic torques, which tend to orient the director field in a direction perpendicular to its low-field equilibrium configuration. As the magnetic field is increased, one encounters an abrupt change in behavior as the magnetic field becomes strong enough to prevent fluctuations from returning to their original (low-field) equilibrium value. In this case the original configuration now represents an unstable situation. Bistable equilibrium configurations then exist, with stable director orientations lying to either side of the low-field director orientation, in the plane defined by the latter and the magnetic field.

In the experiments that we shall propose, and the analysis that we carry out, we envision two beams of monochromatic light simultaneously incident on thin cells containing liquid-crystal material of positive dielectric anisotropy. The beams could have differing intensities, angles of incidence, polarizations, and frequencies. Throughout most of the present paper we shall concentrate on the lowest-order optical nonlinearities produced by beams of equal frequency, although the modifications resulting from nonzero frequency differences will be discussed toward the end of the paper. The results of the present analysis, and related experiments, could yield improved means of determining optical anisotropies, nonlinear optical properties, and nematic director relaxation

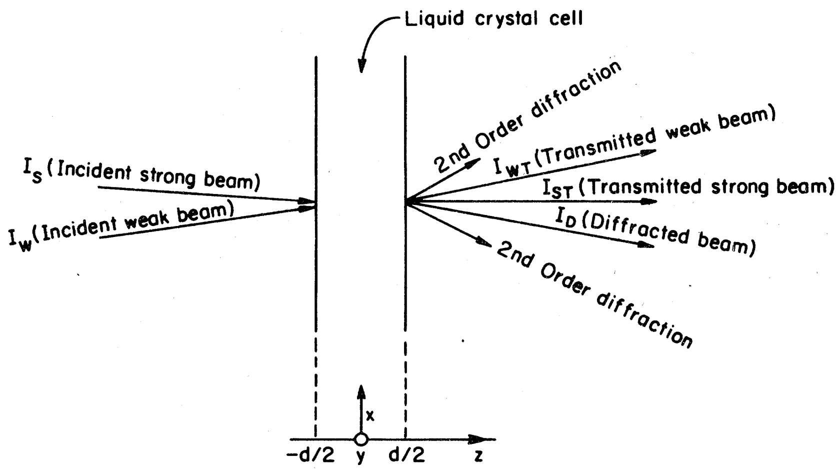

FIG. 1. Cell geometry for light scattering experiments.

rates. The phenomena discussed in the present paper could conceivably lead to direct applications in the area of optical amplification and display, using the birefringent phase holograms envisioned throughout the present analysis.

The general type of experimental configuration to which we address ourselves is shown schematically in Fig. 1. In the present work, we shall be concerned only with the first-order transmitted beams I_{WT} and I_D. The second-order beams indicated might appear as a result of higher-order nonlinearities, in practice, although we shall not deal with them further in the present paper.

The phenomena which we analyze would be most easily observable for the liquid crystal configurations depicted in Figs. 2(a) and 2(b). In this case—homeotropic alignment (director $\hat{n}$ parallel to the z axis in the absence of magnetic field) slightly above the Freedericks transition—the externally applied magnetic field tends to reorient the molecules as indicated schematically in Fig. 2(b) (or equivalently, with tilt angles θ opposite to those shown). At these fields, the director is quite susceptible to being further perturbed by the optical fields (in particular, by the interference signal between the strong and weak beams). As a result, liquid crystals at or near Freedericks transitions have large nonlinear optical susceptibilities. In fact, we shall show that with normal incidence, and strong-beam intensities as low as 2 W/cm^2, reasonably large weak beam optical amplifications (~100%) may be realized for certain angular separations between the incident beams, while for angles of incidence of the order of 45°, the same amplifications may be observed with much lower strong-beam intensities (~0.1 W/cm^2), for equal frequency strong and weak beams. The analysis of the distortions induced by magnetic fields perpendicular to the undistorted director field, both being parallel to the cell surfaces [Figs. 2(c) and 2(d)] is mathematically similar to the above case. Nonetheless, optical amplification in this case is less efficient, due to the form taken by the nonlinear susceptibility. Finally, it is shown that if there exists a nonzero frequency shift between the incident strong and weak beams, with the weak beam having (slightly) lower frequency, the optical amplification is increased while the angular specificity, which is present as a result of phase matching conditions for equal frequency input beams, is no longer present.

In Sec. II we analyze the situation in which the

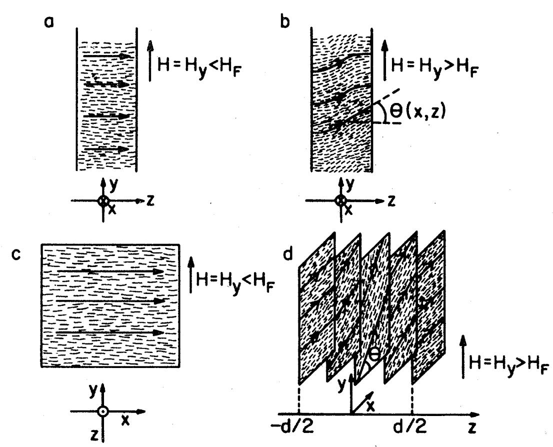

FIG. 2. Molecular alignment (–) and director field (→) for various cell geometries and magnetic fields. (a) Homeotropic alignment below the Freedericks transition. (b) Same initial alignment, above the Freedericks transition. (c) Zero field alignment parallel to x direction. (d) Same initial alignment, with $H_y > H_F$, for several fixed-z planes within the cell. [Equal, opposite values of θ are equivalent to those shown in (b) and (d).]

liquid crystal is aligned homeotropically ($\hat{n}_z$) at zero magnetic field, with applied magnetic field in the y direction (H_y) and with both strong and weak beams (approximately) normally incident with optical fields linearly polarized in the y direction. In Sec. III, we perform a similar analysis for the same configuration, except that now the beams are incident at angle ψ in the y-z plane, the optical fields making an angle Θ with the y axis, also in the y-z plane. In Sec. IV we make an analysis for initial orientation in the x direction ($\hat{n}_x$), applied magnetic field in the y direction, normal incidence, and linear polarization at angle Θ from the y axis in the x-y plane. In Sec. V, the effects of the weak and strong beams having differing polarizations are considered. Finally, in Sec. VI, the effects of frequency differences between the incident beams are derived.

II. NONLINEAR OPTICAL RESPONSE FOR THE CASE $\hat{n}_z, H_y, E_{S_y}, E_{W_y}$

In this and the following sections, we assume that the optical waves propagate with undistorted behavior in lowest approximation, for purposes of finding the nonlinear refractive indices. Following that, the spatially dependent phase retardation of the strong beam in propagation through the sample is calculated. This retardation is directly associated with the diffraction of the strong beam into the WT and D beams (Fig. 1). This corresponds to ones working in the first Born approximation, calculating the lowest-order distortion from the undistorted waveforms. The generalization to the case of continuously distorted waves is, in principle, straightforward and is presented in the Appendix. For weak interactions and thin samples, the more general treatment yields results that are equivalent to the phase-shift approach.

We shall consider working close to the Freedericks transition, so that the θ values for the director angle relative to the z axis [Fig. 2(b)] will be small. For simplicity, we shall work in the approximation that all three Franck elastic constants are equal (denoted by K). Ben-Abraham[12] has shown that this approximation leads only to very minor error in the calculation of the properties of 4-methoxybenzylidene-4′-butylaniline (MBBA), for example, to which reference is made in the numerical calculations of the present paper.

Let us begin by assuming a director angle profile throughout the thickness of the sample, $\theta(\vec{r}, z)$. Here $\vec{r}$ is the projection in the x-y plane of the vector extending from the origin to the points in the liquid crystal sample. The angle $\theta(\vec{r}, z)$ is determined by K, H_y (the applied field), the optical fields (in which interference terms between the incident weak and strong fields which vary with transverse displacement $\vec{r}$ play a prominent role) and the boundary conditions, $\theta(\vec{r}, z=\pm\frac{1}{2}d)=0$.

In an optical field propagating in the z direction, the electric displacement and the optical magnetic field must be transverse to the propagation vector in order to satisfy Maxwell's equations $\nabla\cdot\vec{D}=0$ and $\nabla\cdot\vec{B}=0$. In the present example, therefore, we consider the situation (B_x, D_y, k_z) and, by solving the other Maxwell equations, we find $\vec{E}$ (which satisfies the constitutive relation $\vec{D}=\underline{\epsilon}\cdot\vec{E}$) and k, the magnitude of the k vector. Assuming all wave forms of the type $e^{i(\omega t-kz)}$, $\vec{E}$ has components[13]

$$E_x=0\,,\qquad E_y=\left(\frac{\epsilon+\frac{1}{2}\Delta\epsilon\cos 2\theta}{\epsilon^2-(\frac{1}{2}\Delta\epsilon)^2}\right)D_y\,,\qquad E_z=\left(\frac{\frac{1}{2}\Delta\epsilon\sin 2\theta}{\epsilon^2-(\frac{1}{2}\Delta\epsilon)^2}\right)D_y\,, \tag{1}$$

while k has the form

$$k=\frac{\omega}{c}\left(\frac{\epsilon^2-(\frac{1}{2}\Delta\epsilon)^2}{\epsilon+(\frac{1}{2}\Delta\epsilon)\cos 2\theta}\right)=(\omega/c)n_{eff}\,, \tag{2}$$

in order to satisfy Maxwell's equations. In these equations, $\epsilon=\frac{1}{2}(\epsilon_\parallel+\epsilon_\perp)$ and $\Delta\epsilon=\epsilon_\parallel-\epsilon_\perp$, where $\epsilon_\parallel$ and $\epsilon_\perp$ are the dielectric coefficients that would be observed for fields polarized parallel and perpendicular to the director. (We shall assume positive anisotropy in this paper, $\epsilon_\parallel>\epsilon_\perp$.) For small angles, Eq. (2) reduces to

$$n_e^{1/2}\doteq\epsilon_\perp^{1/2}\left(1+\frac{\Delta\epsilon}{2\epsilon_\parallel}\theta^2(\vec{r},z)\right). \tag{3}$$

Accordingly the strong beam optical wave fronts, upon emergence from the sample, will be distorted, in that there is a nonlinear optical phase retardation which varies with $\vec{r}$, equal to

$$\delta(\vec{r})=k\frac{\Delta\epsilon}{2n_\parallel^2}\int_{-d/2}^{d/2}\theta^2(\vec{r},z)\,dz\,. \tag{4}$$

Provided that the interference components of $\delta(\vec{r})$ are small ($\ll 1$) the strong beam, emerging with distorted wave fronts, may easily be decomposed into plane-wave solutions having wave vectors as shown in Fig. 1, with sidebeams being weak compared with I_{ST}. If $\delta(\vec{r})$ variations are comparable to unity, the sidebeams could then be comparable in intensity to the central component, while higher-order diffraction components would be present. In calculating $\theta^2(\vec{r}, z)$ we employ the elastic continuum theory, according to the treatment of Sheng,[11] generalized to include the effects of the optical fields. The equation for θ is derived through minimization of the free-energy density, which, in the present case, is given through the expression

$$\mathfrak{F}(\vec{t},z)=\frac{K}{2}\left(\frac{d\theta(\vec{t},z)}{dz}\right)^2-\frac{\Delta\chi}{2}H^2\sin^2\theta-\frac{\Delta\epsilon}{8\pi}E^2(\vec{t})\sin^2\theta\,, \tag{5}$$

in which the relatively small derivatives of θ with respect to $\vec{t}$ have been ignored. The first term represents the contribution due to the bending of the director field, the second arises from the orientation of the director at angle $(\frac{1}{2}\pi-\theta)$ relative to the externally applied magnetic field, while the third represents the orientational energy in the slowly varying (actually time-independent, in the present case) components of squared optical electric fields. The latter effectively supplements the magnetic field such that in the present problem, the behavior of θ is the same as if there existed an effective applied magnetic field $H_{\rm eff}(\vec{t})$ of magnitude

$$H_{\rm eff}(\vec{t})=\left(H^2+\frac{\Delta\epsilon}{4\pi\Delta\chi}E^2(\vec{t})\right)^{1/2}. \tag{6}$$

In the above equations $\Delta\chi$ is the anisotropy in magnetic susceptibility while, of course, $\Delta\epsilon$ is the similarly defined anisotropy in dielectric constant for optical frequencies. The equation for $\theta(\vec{t},z)$ as inferred through minimization of the free energy has the form

$$\xi^2(\vec{t})\frac{d^2\theta}{dz^2}+\sin\theta\cos\theta=0\,, \tag{7}$$

subject to the boundary conditions $\theta(\vec{t},z=\pm\frac{1}{2}d)=0$. The coherence length $\xi(\vec{t})$ is defined as

$$\xi(\vec{t})=\frac{1}{H_{\rm eff}}\left(\frac{K}{\Delta\chi}\right)^{1/2}. \tag{8}$$

We now represent E in the form

$$E=E_y=\tfrac{1}{2}\left(\{E_S\exp[-i(kz-\omega t)]+E_W\exp[-i(\sqrt{k^2-k_t^2}\,z+\vec{k}_t\cdot\vec{t}-\omega t)]\}+\text{c.c.}\right), \tag{9}$$

where c.c. denotes the complex conjugate, E_S and E_W representing the strong-and weak-field amplitudes, which may, indeed, be complex. The neglect of differences between the actual weak-field wave-vector component in the z direction, $\sqrt{k^2-k_t^2}$ and k, is justified, provided that $(k_t^2d/k)\ll 1$. This is equivalent to the condition that the angular separation between the input beams be less than $(kd)^{-1/2}$. (In the Appendix, we show that there exists an optimal angle for scattering of the strong beam into the weak and diffracted beams, which is determined, in part, by the phase mismatching associated with the difference in z components of wave vector, for the wider angles of separation between incoming beams. The efficiency for conversion into the weak beam remains high, even for large angles, in the event that the weak beam has slightly lower frequency than the strong, however. This important aspect of the scattering process is also described in the Appendix.) For wider angular separations (20°, say) d should therefore not exceed one optical wavelength (in the liquid crystal medium); for small angular separations (~1°) d may be as large as a few hundred wavelengths. Assuming that the transverse wave vector component $\vec{k}_t$ is small compared with the total wave vector $\vec{k}$, we obtain

$$E^2\cong\tfrac{1}{2}\{|E_S|^2+|E_W|^2+[E_S^*E_W\exp(-i\vec{k}_t\cdot\vec{t})+\text{c.c.}]\}. \tag{10}$$

Finally, assuming that H is nearly equal to the Freedericks transition field H_F; that $\Delta\chi H^2\gg\Delta\epsilon E^2$; and that $E_W\ll E_S$, $H_{\rm eff}$ may be expressed in the approximate form

$$H_{\rm eff}\cong H+\frac{\Delta\epsilon}{16\pi\Delta\chi H_F}\times\{|E_S|^2+[E_S^*E_W\exp(-i\vec{k}_t\cdot\vec{t})+\text{c.c.}]\}. \tag{11}$$

The solution for Eq. (7) is well known. For small director angles, $\theta(\vec{t},z)$ has the form

$$\theta(\vec{t},z)\cong\theta_M(\vec{t})\cos(\pi z/d)\,. \tag{12}$$

The maximum angle of inclination (which occurs at $z=0$) has the approximate form

$$\theta_M(\vec{t})\cong\begin{cases}2\{[H_{\rm eff}(\vec{t})-H_F]/H_F\}^{1/2}, & H>H_F\\ 0, & H<H_F,\end{cases} \tag{13}$$

with H_F, the Freedericks field, being equal to $(\pi/d)(K/\Delta\chi)^{1/2}$. Restricting our interest to situations in which $H\geqslant H_F$, therefore, the phase shift is found by integration over z in accordance with Eq. (4), with the result

$$\delta(\vec{t})\cong\frac{kd\Delta\epsilon}{n_\parallel^2}\left(\frac{H-H_F}{H_F}\right)+\frac{kd(\Delta\epsilon)^2}{16\pi n_\parallel^2\Delta\chi}\,\frac{\{|E_S|^2+[E_S^*E_W\exp(-i\vec{k}_t\cdot\vec{t})+\text{c.c.}]\}}{H_F^2}. \tag{14}$$

The first term arises from the magnetic-field-induced change in effective index of refraction; the term in $|E_S|^2$ leads to self-focusing effects, which will be neglected in the present treatment; and, finally, the terms modulated with wave vector $\vec{k}_t$ give rise to a contribution to the weak transmitted beam and to the diffractive component.

To obtain the sidebeam intensities from $\delta(\vec{t})$

we simply write the laser field, following transmission through the sample, as

$$E_S \exp\{i[\omega t - kz - \delta(\vec{t})]\} = e^{-i\delta_0}\Big[E_S \exp[i(\omega t - kz)] \quad (15a)$$

$$- \left(\frac{ikd\Delta\epsilon^2}{16\pi n_\parallel^2 \Delta\chi}\right)\frac{|E_S|^2}{H_F^2} E_W \exp[i(\omega t - kz - \vec{k}_t \cdot \vec{t})] \quad (15b)$$

$$- \left(\frac{ikd\Delta\epsilon^2}{16\pi n_\parallel^2 \Delta\chi}\right)\frac{E_S^2}{H_F^2} E_W^* \exp[i(\omega t - kz + \vec{k}_t \cdot \vec{t})]\Big] \;. \quad (15c)$$

having incorporated the terms not modulated with $\vec{k}_t$ into δ_0. The first term (15a) represents the simply transmitted beam, traveling in the initial strong-beam direction, while (15b) represents a supplement to the weak-transmitted beam and (15c) represents the diffractive component. The electric field amplitude for the simply transmitted weak beam (which would have been present in the absence of the nonlinearities) must be added to (15b) to obtain the resultant field, which, in turn, must be squared to obtain the intensity. The imaginary number i in the latter ensures that there is no interference and that the intensities should be added. (This is not the case when one allows for frequency differences to exist between E_W and E_S, as will be discussed below.) Accordingly,

$$I_{WT} = I_W\left[1 + \left(\frac{kd\Delta\epsilon^2 |E_S|^2}{16\pi n_\parallel^2 \Delta\chi H_F^2}\right)^2\right], \quad (16)$$

ignoring reflection losses at the faces of the sample holder. By contrast the diffracted intensity, obtained by squaring (15c), is simply

$$I_D = (I_{WT} - I_W), \quad (17)$$

where in Eqs. (16) and (17) I_W is the weak transmitted intensity in the absence of the strong field, as is consistant with the general behavior of 4-photon scattering processes,[14,15] as explained in the Appendix.

Considering, as an example, a 100-μm-thick sample of MBBA, the diffracted intensity is obtainable in the form $0.25 I_S^2 I_W$, with I_S measured in W/cm^2; thus for strong beams having intensities in the neighborhood of 2 W/cm^2, one expects a doubling in weak transmitted intensity. One may operate somewhat above, as opposed to just above, the Freedericks transition, as can be seen by the absence of any factor which depends sensitively on $(H - H_F)$ in Eqs. (16) and (17). This arises from the fact that it is θ^2 and not θ itself that is responsible for the diffractive process, and θ^2 varies directly as $(H_{eff} - H_F)$, so long as H_{eff} does not greatly exceed H_F ($H_{eff} < 1.5 H_F$, say).

III. $\hat{n}_z$, H_y, E_S, AND E_W POLARIZED AT ANGLE Θ FROM THE y AXIS IN THE y-z PLANE

The configuration may be achieved through homeotropic alignment in the z direction with optical fields propagating at some angle Θ with respect to the z axis, with linear polarization in the y-z plane as shown in Fig. 3. In order to achieve angle Θ, the incident beams must propagate at angle ψ through the sample, to be determined. The differences between the nonlinear optical phenomena in this case and those described above arise from two effects: (i) the differences in behavior of the liquid crystal in tilted fields and (ii) the differences in propagation of the strong beam having a different direction of polarization relative to the (perturbed) liquid crystal director. In contrast to the results of Sec. II, it will now be seen that θ is linearly dependent on the interference component of the optical intensity while the index variations will similarly depend linearly on θ. To find $\theta(z)$ we note that Eq. (5) is now replaced by

$$\mathfrak{F} = \frac{K}{2}\left(\frac{d\theta(z,\vec{t})}{dz}\right)^2 - \frac{\Delta\chi}{2} H^2 \sin^2\theta - \frac{\Delta\epsilon}{8\pi} E^2 \sin^2(\theta + \Theta). \quad (18)$$

This is equivalent to the expression

$$\mathfrak{F} = \frac{K}{2}\left(\frac{d\theta(z,\vec{t})}{dz}\right)^2 - \frac{\Delta\chi}{2} H^2 (1 + \sigma^2 + 2\sigma\cos 2\Theta)^{1/2} \times \sin^2(\theta + \alpha) + (\text{const}) E^2, \quad (19)$$

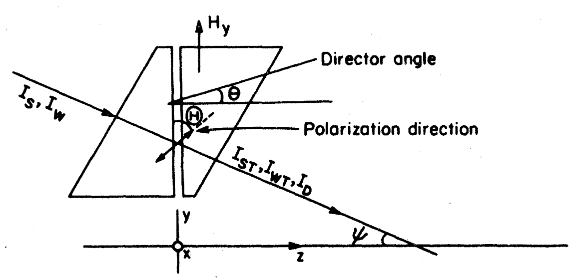

FIG. 3. Optical configuration for obtaining angle ψ between the incident light beam and undistorted director, corresponding to angle Θ between the polarization direction and the y axis.

where

$$\sigma = \Delta\epsilon E^2/4\pi\Delta\chi H^2 \tag{20}$$

and

$$\sin 2\alpha = \sigma \sin 2\Theta/(1+\sigma\cos 2\Theta) . \tag{21}$$

The free energy of orientation in the combined fields is maximized at angle $-\alpha$.

At this point, two effects must be considered. The less dominant is simply contained in the modulation factor for free energy, $(1+2\sigma\cos 2\Theta+\sigma^2)^{1/2}$. This factor, by itself, leads to orientational disturbances that are equivalent, in lowest order, to those obtained for the normally incident light, except that the (approximate) factor $\cos 2\Theta$ now multiplies each occurrence of $|E_S|^2, E_S^* E_W$, etc., in Eq. (11) for H_{eff}. Nevertheless, the more interesting effect, which is of primary importance, is provided by the maximization of the orientational free-energy component at angle $-\alpha$. In analyzing its effects we shall, for the moment, replace the modulating factor $(1+2\sigma\cos 2\Theta+\sigma^2)^{1/2}$ by unity. Then the differential equation (7) may be replaced by an equivalent one in the angle $\theta+\alpha$, subject to the boundary conditions $\theta=0$ at $z=\pm\frac{1}{2}d$. If we define $\beta=\theta+\alpha$, then the differential equation in β has a form identical with Eq. (7) and the boundary conditions become $\beta=\alpha$ at $z=\pm\frac{1}{2}d$. The solution to this equation is, then,

$$\frac{1}{\xi_F}\frac{H}{H_F}\left(\frac{d}{2}+z\right)\sin\beta_m = \int_\alpha^\beta d\beta'\left[1-\left(\frac{\sin\beta'}{\sin\beta_m}\right)^2\right]^{-1/2}, \tag{22}$$

where, of course, α, β, and β_m are functions of the transverse components of position $\vec{t}$ through the interference effects of the optical fields on α. One should view Eq. (22) as applicable in the domain $-\frac{1}{2}d<z<0$, with the understanding that the solutions are symmetric about $z=0$. In all cases α will be viewed as being small, inasmuch as $\sigma \ll 1$ [Eqs. (20) and (21)]. If at the same time we restrict our attention to reasonably small β-values, Eq. (22) may be solved, approximately, through expansion of $\sin\beta'$ and $\sin\beta_m$. To find β_m, we take the case $z=0, \beta=\beta_m$ in which case

$$\frac{\pi}{2}\frac{H}{H_F}\sin\beta_m = \int_\alpha^{\beta_m} d\beta\left[1-\left(\frac{\sin\beta}{\sin\beta_m}\right)^2\right]^{-1/2} \tag{23}$$

holds. This integral may be rendered into the form of an elliptic integral through the transformation

$$\sin\phi = \sin\beta/\sin\beta_m , \tag{24}$$

in which case Eq. (23) reduces to

$$\frac{\pi}{2}\frac{H}{H_F} = \int_{\phi(\alpha)}^{\pi/2} d\phi\, [1-(\sin\beta_m\sin\phi)^2]^{-1/2} . \tag{25}$$

The above is now an elliptic integral, in view of the fact that $\sin\beta_m<1$, having the solution[14]

$$\frac{\pi}{2}\frac{H}{H_F} = K(\sin\beta_m) - F(\phi(\alpha), \sin\beta_m) , \tag{26}$$

with

$$\phi(\alpha) = \sin^{-1}(\sin\alpha/\sin\beta_m) , \tag{27}$$

K and F being complete and incomplete elliptic integrals of the first kind. They may be expanded in the form

$$K(k) = \frac{\pi}{2}\sum_{n=0}^{\infty}\left(\frac{(2n-1)!!}{2n!!}\right)^2 k^{2n}$$

and

$$F(\phi,k) = \sum_{n=0}^{\infty}\left(\frac{(2n-1)!!}{2n!!}\right)\gamma_{2n}k^{2n} ,$$

with

$$\gamma_{2n} = \int_0^\phi \sin^{2n}\phi\, d\phi ,$$

provided that we interpret the term $((-1)!!/0!!)$ as unity. The physically interesting cases will, of course, occur for H nearly equal to H_F, in which case $\alpha \ll \beta_m \ll 1$. Accordingly, we retain terms only to first powers in α and second in β_m with the result that Eq. (26) may be reduced to the form

$$4(H/H_F-1) \cong \beta_m^2 - 8\alpha/\pi\beta_m \tag{28}$$

having approximated $\phi(\alpha)$, from Eq. (27), simply as $(\alpha/\sin\beta_m)$. In the case $\alpha=0$, β_m has real solutions, approximately equal to $\pm 2[(H-H_F)/H_F]^{1/2}$, reproducing the result of Sec. II. This remains essentially the case for fields substantially greater than H_F in that the solution to the above equation yields β_m approximately equal to $2[(H-H_F)/H_F]^{1/2} + (\alpha/\pi)[(H-H_F)/H_F]^{-1}$ for the condition $(H-H_F)/H_F \gg (\alpha/\pi)^2$. For smaller fields (including $H \lesssim H_F$), of course, the α-dependent term becomes relatively more important. At $H=H_F$, for example, β_m is equal to $(8\alpha/\pi)^{1/3}$, which is typically very much larger than α, yet not as large as it would be at the larger fields. Notice, at this point, that there is no negative root for β at $H \leqslant H_F$; in fact, the negative root is possible only so long as $(H-H_F)/H_F \geqslant 3(\alpha/2\pi)^{2/3}$. (With this in mind, the root which appears to be dominant, physically, is the positive one and we shall therefore focus our attention upon that root.) For $H<H_F$ such that $(H_F-H)/H_F \gg \frac{1}{2}(\alpha/\pi)^{1/3}$, β_m is approximately equal to $(2\alpha/\pi)[(H_F-H)/H_F]^{-1}$ provided, still, that $(H_F-H)/H_F \ll 1$. To summarize, therefore, the positive root has the following approximate form:

$$\beta_m \cong \begin{cases} 2\left(\frac{H-H_F}{H_F}\right)^{1/2} + \frac{\alpha}{\pi}\left(\frac{H-H_F}{H_F}\right)^{-1}, & 1 \gg \frac{H-H_F}{H_F} \gg \left(\frac{\alpha}{\pi}\right)^2, \quad (29a) \\ \left(\frac{8\alpha}{\pi}\right)^{1/3}, & \frac{H-H_F}{H_F} = 0, \quad (29b) \\ \frac{2\alpha}{\pi}\left(\frac{H_F-H}{H_F}\right)^{-1}, & 1 \gg \frac{H_F-H}{H_F} \gg \frac{1}{2}\left(\frac{\alpha}{\pi}\right)^{1/3}. \quad (29c) \end{cases}$$

In order to assess the relative importance of various influences on θ_m, we recall that $\theta_m = \beta_m - \alpha$, and that we are ultimately interested in dependencies of each of the above terms on squared optical fields as they enter through the parameter σ. At this point, we may restore the modulation factor $(1+2\sigma\cos 2\Theta + \sigma^2)^{1/2}$. To lowest order in σ, H should be replaced by $H_{\text{eff}} \cong H(1+\sigma\cos 2\Theta)$ while $\alpha \cong \sigma \sin 2\Theta$. Accordingly, if we examine Eq. (29a), we find

$$\left(\frac{d\theta_m}{d\sigma}\right) \cong \frac{H}{H_F}\cos 2\Theta\left(\frac{H-H_F}{H_F}\right)^{-1/2} + \sin 2\Theta\left[\frac{1}{\pi}\left(\frac{H-H_F}{H_F}\right)^{-1} - 1\right], \quad (30a)$$

$$1 \gg \frac{H-H_F}{H_F} \gg \left(\frac{\alpha}{\pi}\right)^2.$$

It is clear that the second term, which arises from the α dependence of θ_m, is dominant especially near the Freedericks transition. It is clear from (29c), moreover, that just below the Freedericks transition, the dependence of θ_m on interference terms in the optical fields is of the same order of magnitude as it is just above the transition—in fact, it is virtually twice as large,

$$\frac{d\theta_m}{d\sigma} \cong \sin 2\Theta\left[\frac{2}{\pi}\left(\frac{H-H_F}{H_F}\right)^{-1} - 1\right], \quad 1 \gg \frac{H-H_F}{H_F} \gg \frac{1}{2}\left(\frac{\alpha}{\pi}\right)^{1/3}. \quad (30b)$$

Obviously, expressions (29a) and (29c) are not valid at the transition itself in which case one would examine the properties of (29b), whose derivatives diverge as $\alpha \to 0$—that is, as the optical fields themselves diminish. Further analysis of this feature would most likely reveal the simultaneous presence of many orders or nonlinear optical processes; however, one presumably would work somewhat away from the transition itself in order to achieve optical stability.

The other aspect of the problem that now has importance, since the optical fields are polarized at angle Θ with respect to the y axis, is the manner in which first order changes in θ can affect the phase shifts. In fact, for arbitrary ψ values, θ in Eq. (2) must now be replaced by $\theta+\psi$, and the optical-field-induced components of n_{eff} which we shall denote as $n_{\text{t.i.}}$ become

$$n_{\text{t.i.}} = \frac{[\epsilon^2 - (\frac{1}{2}\Delta\epsilon)^2]^{1/2}}{(\epsilon + \frac{1}{2}\Delta\epsilon\cos 2\psi)^{3/2}} \frac{\Delta\epsilon}{2} \sin 2\psi \frac{d\theta}{d\sigma}\sigma. \quad (31)$$

We must, therefore, find $\theta(z)$ in terms of θ_m, and ultimately integrate over the cell thickness. Returning to Eq. (22), assuming that $H/H_F \cong 1$, and confining our interest to lowest order terms, we see that

$$\frac{\pi z \beta_m}{d} \cong -\int_\beta^{\beta_m} d\beta' \left[1 - \left(\frac{\beta'}{\beta_m}\right)^2\right]^{-1/2}$$

whose solution is, then

$$\frac{\pi z}{d} \cong \sin^{-1}(\beta/\beta_m) - \tfrac{1}{2}\pi.$$

With $\beta \gg \alpha$ we find, once again [cf. Eq. (12)]

$$\theta(z) \cong \theta_m \cos(\pi z/d). \quad (32)$$

The phase shift, $\delta(\vec{t})$ is now obtainable through the expression

$$\delta(\vec{t}) = k_0 \int_{-d/2}^{d/2} n_{\text{t.i.}} \frac{dz}{\cos\psi}, \quad (33)$$

where k_0 is the vacuum wave vector and the factor $1/\cos\psi$ represents the lengthening of the path due to nonnormal incidence. Using Eqs. (20), (30b), (31), and (32), we obtain

$$\delta(\vec{t}) = \frac{k_0 d(\Delta\epsilon)^2}{4\pi^2\Delta\chi} \frac{[\epsilon^2 - (\frac{1}{2}\Delta\epsilon)^2]^{1/2}}{(\epsilon+\frac{1}{2}\Delta\epsilon\cos 2\psi)^{3/2}} \times\left[\frac{2}{\pi}\left(\frac{H_F-H}{H_F}\right)^{-1} - 1\right] \times\left(\frac{\sin 2\psi \sin 2\Theta}{\cos\psi}\right)\frac{E^2(\vec{t})}{H_F^2}, \quad (34)$$

with Θ and ψ related by the expression

$$\Theta = \psi + \tan^{-1}\left(\frac{\frac{1}{2}\Delta\epsilon\sin 2\psi}{(\epsilon+\frac{1}{2}\Delta\epsilon\cos 2\psi)}\right), \quad (35)$$

according to (1). By comparison with Eq. (14), we see that the effect of tilting the optical fields is one of further enhancing the magnitude of the diffracted intensity by the factor

$$\left\{\frac{2n_{\parallel}^3}{\pi}\left(\epsilon+\frac{\Delta\epsilon}{2}\cos 2\psi\right)^{-3/2}\times\left[\frac{2}{\pi}\left(\frac{H_F-H}{H_F}\right)^{-1}-1\right]\frac{\sin 2\psi\sin 2\Theta}{\cos\psi}\right\}^2$$

over the normal-incidence case. For MBBA, with $H=0.9H_F$ and $\psi=\frac{1}{4}\pi$, this factor is 28.7, so that one should observe 100% weak-beam amplification at strong-beam intensities of order 0.1 W/cm^2. In addition, the fact that one may operate below the Freedericks transition may lead to much greater optical stabilities than would be found for the case $H\gtrsim H_F$.

IV. $\hat{n}_x, H_y, E_S$, AND E_W POLARIZED PARALLEL TO ONE ANOTHER, AT ANGLE Θ FROM THE y AXIS IN THE x-y PLANE

This alignment is achieved through treating the glass surfaces in various ways so as to cause an alignment at the cell walls in the x direction that is parallel to the walls. With the new definition for Θ (i.e., in the x-y plane, as opposed to the y-z plane) and with the elastic free energy term being $\frac{1}{2}K(d\theta/dz)^2$, with K now representing the Franck elastic constant for bending and θ being defined as the director angle in the x-y plane relative to the x direction, the equation governing the director and the boundary conditions are identical to those of Sec. II for $\Theta=0$ and Sec. III for $\Theta\neq 0$. The differences arise solely from the manner in which the light scattering takes place. To illustrate this, let us analyze the case $\Theta\neq 0$, and compare results with those for Sec. III. In that section, we found diffracted amplitudes of order $E_S\delta$ which, in turn, are of order

$$k_0 d n_{t.1.} E_S \sim \Delta\epsilon \sin 2\psi (d\theta_m/d\sigma)\sigma k_0 d E_S\,, \qquad (36)$$

according to Eq. (31).

In contrast, for the present case, if we approximate $\theta(z,\vec{t})$ by $\theta_m(\vec{t})$ for the moment, the effects of the birefringence grating are obtained by analyzing the optical electric field following transmission through the medium in the following manner. First, we decompose E_S into components parallel and perpendicular to the director,

$$E_S=E_S\hat{n}_y=E_S\{\cos[\Theta+\theta_m(\vec{t})]\hat{n}_\perp+\sin[\Theta+\theta_m(\vec{t})]\hat{n}_\parallel\}.$$

Following passage through the sample, this field then becomes

$$\sim E_S\{\cos[\Theta+\theta_m(\vec{t})]\hat{n}_\perp\exp(-ik_0n_\perp d)+\cos[\Theta+\theta_m(\vec{t})]\hat{n}_\parallel\exp(-ik_0n_\parallel d)\}.$$

This may be rewritten, assuming $\theta_m\ll\Theta$ and representing the modulational components of $\theta_m(\vec{t})$ by $(d\theta_m/d\sigma)\sigma(\vec{t})$, in the form

$$E_S[\cos\Theta\,\hat{n}_\perp\exp(-ik_0n_\perp d)+\sin\Theta\,\hat{n}_\parallel\exp(-ik_0n_\parallel d)]+(d\theta_m/d\sigma)\sigma(\vec{t})E_S[\hat{n}_\parallel\cos\Theta\exp(-ik_0n_\parallel d)-\hat{n}_\perp\sin\Theta\exp(-ik_0n_\perp d)].$$

The first component is obviously the transmitted strong beam, which has now been elliptically polarized according to the relative phase $k_0(n_\parallel-n_\perp)d$. The second term represents the modulational component, proportional to $\sigma(\vec{t})$, which contains both the nonlinear optical contribution to E_{WT} as well as E_D. It is important to note that the sidebeams have polarization orthogonal to that of the transmitted strong beam—a fact that could ultimately be important in optical processing. Nonetheless, for present purposes, the amplification factor is relatively small, having amplitude

$$E_D\sim(d\theta_m/d\sigma)\sigma E_S\,,$$

as compared with expression (36). The principal numerical difference lies in the absence of the factor k_0d from the above expression which, for 100-μm thicknesses, is of the order 10^3. Accordingly we shall not pursue this line of investigation further but shall, instead, return to the configuration associated with Secs. II and III.

V. SAME GEOMETRY AS SEC. III, EXCEPT THAT VARIOUS POLARIZATIONS OF E_S AND E_W ARE CONSIDERED

In Sec. III, we treated the case $\hat{n}_z$, H_y, E in y-z plane, at angle Θ with respect to y axis, and we have found that so far, this configuration produces optimal light scattering for Θ in the neighborhood of $\frac{1}{4}\pi$. In order to achieve this configuration the propagation vector is oriented at angle ψ from the z direction. We shall now consider the same direction of propagation, with the possibility that the E fields could have various polarizations. A qualitative description will suffice to characterize the phenomena. The point of difference is that now the electric field components in the x direction will couple to no other field components, nor will they influence the director angles, simply because they are oriented perpendicular to the y-z plane in which the director is constrained by the dominant forces in the $\hat{n}_z H_y$ configuration. Hence, these components will propagate undisturbed through the sample, taking on the usual phase factor, $\exp(-ik_0n_\perp d)$. Accordingly, only the components of E_W and E_S in the plane of incidence interact and scatter nonlinearly; the diffracted field will be completely polarized in the plane of incidence, as would the amplification component of the weak beam. Consequently, the weak beam, as amplified, and the diffracted beam would have polarizations which simply differ from the transmitted strong beam, with the result that one could conceivably perform downstream selection of the radiational components through an appropriate combination of analyzers.

VI. FREQUENCY DIFFERENCES IN E_W AND E_S

Let us now modify the incident beams so that there exists a frequency difference

$$\Omega = \omega_W - \omega_S$$

between the weak and strong beams. The squared optical field replacing that given by Eq. (10) is then

$$E^2 = \tfrac{1}{2}\left(|E_S|^2 + |E_W|^2 + \{E_S^* E_W \exp[-i(\vec{k}_t\cdot\vec{t} - \Omega t)] + \text{c.c.}\}\right). \tag{37}$$

The frequency differences envisioned are small—of the order of the inverse relaxation time τ^{-1} for the optically induced excitations. The equation governing the excitations would then be of the form

$$\frac{d\theta_m(\vec{t}, t)}{dt} = \frac{1}{\tau}\left(\theta_m^{\text{s.s.}}(\vec{t}, t) - \theta_m(\vec{t}, t)\right), \tag{38}$$

assuming a single relaxation time, with $\theta_m^{\text{s.s.}}(\vec{t}, t)$ being the steady state value of θ_m, having been obtained in previous sections of the present paper, with $E_S^* E_W$ now multiplied by $e^{i\Omega t}$,

$$\theta_m^{\text{s.s.}}(\vec{t}, t) = \frac{d\theta_m}{d\sigma}\frac{\Delta\epsilon}{8\pi\Delta\chi}\left(\frac{|E_S|^2 + E_S^* E_W \exp[-i(\vec{k}_t\cdot\vec{t} - \Omega t)] + \text{c.c.}}{H_F^2}\right). \tag{39}$$

The solution of Eq. (38) is then

$$\theta_m(\vec{t}, t) = \frac{d\theta_m}{d\sigma}\frac{\Delta\epsilon}{8\pi\Delta\chi}\left[\frac{|E_S|^2}{H_F^2} + \frac{1}{H_F^2}\left(\frac{E_S^* E_W \exp[-i(\vec{k}_t\cdot\vec{t} - \Omega t)]}{1 + i\Omega t} + \text{c.c.}\right)\right]. \tag{40}$$

The corresponding expression for the dielectric tensor is given by Eq. (A17) of the Appendix. Accordingly, the spatially modulated terms in the phase retardation, δ [Eqs. (14) and (34)], now acquire the phase factor $e^{i\Omega t}/(1+i\Omega t)$, or its complex conjugate, with the result that to lowest order in the phase modulation, the emergent optical field is given by

$$E_S \exp\{i[\omega_S t - kz - \delta(\vec{t}, t)]\} = e^{-i\delta_0}\left[E_S \exp[i(\omega_S t - kz)] - i\frac{d\delta}{d\sigma}\frac{\Delta\epsilon}{8\pi\Delta\chi H_F^2}\right.$$
$$\left.\times\left(\frac{|E_S|^2 E_W \exp[i(\omega_W t - \vec{k}_W\cdot\vec{x})]}{1 + i\Omega t} + \frac{E_S^2 E_W^* \exp\{i[(\omega_S - \Omega)t - \vec{k}_D\cdot\vec{x}]\}}{1 - i\Omega t}\right)\right], \tag{41}$$

in analogy to Eq. (15) of Sec. II or its counterpart in other sections. Accordingly, while the diffracted amplitudes are effectively decreased, the lowest-order contribution to the transmitted weak amplitude has a component which is in phase with the simply transmitted weak component (that which is present in the absence of nonlinearities). Accordingly, a form of optical heterodyning occurs, which leads to a true optical gain characterizing two-photon interactions[15] as explained in the Appendix. The optical amplification factor for traversal of the cell by the weak beam is

$$\frac{I_W(\frac{1}{2}d)}{I_W(-\frac{1}{2}d)} = \exp\left(-\frac{2\Omega t}{1+(\Omega t)^2}\frac{d\delta}{d\sigma}\frac{\Delta\epsilon|E_S|^2}{8\pi\Delta\chi H_F^2}\right) \tag{42}$$

for wide-angle separations between the incident beams. The amplification maximizes for frequency difference $\Omega = -\tau^{-1}$ (Stokes-shifted weak beam) in agreement with that for nonlinear orientational scattering in liquids. This frequency difference would be quite small, particularly near the Freedericks transition. The small frequency differences simply act to provide a spatial phase difference between $\theta_m(\vec{t}, t)$ and the fields which give rise to it. Such frequency differences typically would lie well within the laser linewidth itself, but could nonetheless be provided by a slight Doppler shifting of one of the incident beams.

The measurement of optical amplification as a function of Ω could be useful to the determination of nematic relaxation times for well-specified excitations near Freedericks transitions. As an example of the effectiveness of the optical heterodyning implicit in Eq. (43), if one worked with strong beam intensities such that for zero frequency difference one obtained a 1% weak-beam enhancement, for $\Omega\tau = -1$ one would now obtain a 10% increase. Even more important, however, may be the fact that for $\Omega = 0$ one must work at $\theta \cong \theta_{\text{opt}}$, as explained in the Appendix, while the amplification takes place for a wide range of angles and the diffracted component tends to disappear for $\Omega \neq 0$.

VII. CONCLUSION

Several polarization cases have been examined for nonlinear optical amplification and diffraction of light beams transmitted through thin nematic liquid-crystal samples. It is found that substantial coupling between weak and strong light beams having the same frequency should occur for relatively low-level CW reference beam intensities (~0.1 W/cm^2 for cell thickness ~100 μm) for homeotropically aligned samples maintained near (slightly above or below) the Freedericks transition, with externally applied magnetic field lying parallel to the cell walls in the plane of incidence. The incident optical beam should also have electric fields polarized in the plane of incidence, the angle of incidence being approximately 45°. If, moreover, a frequency difference exists between the beams there will exist an optical heterodyning effect that serves to enhance the lower-frequency beam at the expense of the higher. This effect maximizes for angular frequency separations equal to the inverse relaxation time for the nematic director-field excitations generated by optical interference signals, and would take place for a wide range of angular separations between the incident beams.

ACKNOWLEDGMENT

The authors take pleasure in thanking T. A. Wiggins and W. Doane for stimulating discussions during the course of this research. This research was supported in part by the U.S. Office of Naval Research.

APPENDIX: APPROXIMATE SELF-CONSISTENT SOLUTIONS FOR MAXWELL'S EQUATIONS IN NONLINEAR BIREFRINGENT MEDIA

We wish to prove our contention that the phase integral of Sec. III, Eq. (33), is the important quantity for producing weak beam amplification (as well as the diffraction component) for equal-frequency incident beams, and to gain a deeper insight into the scattering dynamics in general. In addition, we shall analyze the effects of frequency separations between the incident beams. We begin by examining Maxwell's equations under the assumption that the incident beams propagate in a direction forming (nonzero) angle ψ with the unperturbed director as illustrated in Fig. 3, and described in Sec. III. Moreover, we shall confine our interest to the case $H<H_F$, although other cases may be handled similarly. In any event, we assume $\vec{B}=\vec{H}$ ($\mu=1$), while

$$\vec{D}=\underline{\epsilon}\cdot\vec{E}$$

with $\underline{\epsilon}$, a second-rank Cartesian tensor, assumed to have the form

$$\underline{\epsilon}=\underline{\epsilon}_0+\tfrac{1}{2}\underline{\epsilon}_2(z)\left\{|E_S|^2+\left[E_S^*E_W(z)\exp\left(-i\int(\vec{k}_W-\vec{k}_S)\cdot d\vec{x}\right)+\text{c.c.}\right]+\left[E_S^*E_D(z)\exp\left(-i\int(\vec{k}_D-\vec{k}_S)\cdot d\vec{x}\right)+\text{c.c.}\right]\right\}. \quad (A1)$$

Here $\underline{\epsilon}_2$ is linearly proportional [cf. discussion preceding Eq. (31)] to the director tilt angle $\theta(z)$, which would have resulted were the illumination intensity constant in z; hence the perturbational components of the dielectric tensor are assumed proportional to their value in uniform intensity optical fields times the actual intensity as a function of $(\vec{t},z)$. This assumption is justified by noting that θ_m [and therefore $\theta(z)$] depends linearly on α [Eq. (29c)] for z-independent intensity; that α is only very weakly dependent on z by virtue of the small magnitudes of $E_W(z)$ and $E_D(z)$ as compared with E_S; and that in the differential equations resulting from the minimization of $\mathfrak{F}$ [Eq. (19)], the assumption that θ is now multiplied by the ratio $\alpha(z)/\alpha(z=0)$ yields a differential equation sensibly equivalent to the form which is found for constant intensity. [This amounts to neglecting relative changes with z in $\alpha(z)$ compared with those in $\theta(z)$.] As usual, we have neglected terms in $|E_W|^2$ and $|E_D|^2$, although we are obviously including the major effects of both the diffracted component and the weak beam on $\underline{\epsilon}$. The propagation vectors for the three beams are assumed to be spatially varying, the phase of the weak beam, for example, being $-\int\vec{k}_W\cdot d\vec{x}$ where the line integral in each case, is taken on a path from the entrance window of the cell to the field point along the direction of k_W (etc. for k_D, k_S).

Inasmuch as the term $\frac{1}{2}\underline{\epsilon}_2(z)|E_S|^2$ is common in the propagation of all three fields, we shall henceforth incorporate it into $\underline{\epsilon}_0$. (Since $\underline{\epsilon}_2$ varies with z, $\underline{\epsilon}_0$ must now be regarded as having a slight spatial variation as well. The propagation vector $\vec{k}_S$ will be used to define the z direction, $\vec{k}_W$ and $\vec{k}_D$ lying on opposite sides of $\vec{k}_S$, forming small angles with $\vec{k}_S$.) The three optical fields now satisfy the following equations,

$$-\nabla\times\nabla\times\vec{E}_S\exp\left[-i\left(\int_{-d/2}^{z}k_S\,dz-\omega t\right)\right]=\frac{\partial^2}{c^2\partial t^2}\underline{\epsilon}_0\cdot\vec{E}_S\exp\left[-i\left(\int_{-d/2}^{z}k\,dz-\omega t\right)\right], \quad (A2)$$

$$-\nabla\times\nabla\times\vec{E}_W\exp\left[-i\left(\int\vec{k}_W\cdot d\vec{x}-\omega t\right)\right]=\frac{\partial^2}{c^2\partial t^2}(\underline{\epsilon}_0+\tfrac{1}{2}\underline{\epsilon}_2\,|E_S|^2)\cdot\vec{E}_W\exp\left[-i\left(\int\vec{k}_W\cdot d\vec{x}-\omega t\right)\right]$$
$$+\frac{\partial^2}{c^2\partial t^2}(\tfrac{1}{2}\,\underline{\epsilon}_2E_S^2\cdot\vec{E}_D^*)\exp\left[-i\left(\int(2\vec{k}_S-\vec{k}_D)\cdot d\vec{x}-\omega t\right)\right],\qquad (A3)$$

$$-\nabla\times\nabla\times\vec{E}_D\exp\left[-i\left(\int\vec{k}_D\cdot d\vec{x}-\omega t\right)\right]=\frac{\partial^2}{c^2\partial t^2}(\underline{\epsilon}_0+\tfrac{1}{2}\,\underline{\epsilon}_2|E_S|^2)E_D\exp\left[-i\left(\int\vec{k}_D\cdot d\vec{x}-\omega t\right)\right]$$
$$+\frac{\partial^2}{c^2\partial t^2}(\tfrac{1}{2}\,\underline{\epsilon}_2E_S^2E_W^*)\exp\left[-i\left(\int(2\vec{k}_S-\vec{k}_W)\cdot d\vec{x}-\omega t\right)\right].\qquad (A4)$$

Here we have ignored possible small effects on $\underline{\epsilon}_2$ associated with slight polarization direction differences in $\vec{E}_S$, $\vec{E}_W$, and $\vec{E}_D$ associated with their different directions of propagation. (These differences disappear in case the polarization direction is chosen as perpendicular to the plane containing $\vec{k}_S$, $\vec{k}_W$, and $\vec{k}_D$ at any rate.) Inasmuch as $\vec{D}$ must be perpendicular to the propagation vector for each of its component fields (since $\nabla\cdot\vec{D}=0$), the $\vec{E}$ fields in each of the three equations above has a direction which, then, renders the right-hand sides of the three equations perpendicular to the respective propagation directions. [Since $\underline{\epsilon}$ and the various field strengths change throughout the sample the optical electric fields (and secondarily the propagation vectors) change direction slightly as they propagate through the sample. Such changes will be regarded as very small in their effects, and will be ignored in this treatment.] Accordingly, we keep only the perpendicular components of all terms in Eqs. (A2)–(A4). Multiplying Eq. (A3) by the factor $E_W/E_{W\perp}$, we note that the left side of the equation would then be

$$\frac{E_W}{E_{W_\perp}}(-\nabla\times\nabla\times\vec{E}_W)=\frac{\hat{n}_\perp E_W}{E_{W_\perp}}\nabla^2E_{W_\perp}=(\nabla^2E_W)\hat{n}_\perp$$

while the various perpendicular components of the right-hand side of Eq. (A3) become

$$\frac{E_W}{E_{W_\perp}}(\underline{\epsilon}_0\cdot\vec{E}_W)_\perp=\epsilon_0E_W,$$

$$\frac{E_W}{E_{W_\perp}}(\underline{\epsilon}_2\cdot\vec{E}_W)_\perp=\epsilon_2E_W,$$

$$\frac{E_W}{E_{W_\perp}}(\underline{\epsilon}_2\cdot\vec{E}_D)_\perp=\epsilon_2E_D,$$

etc. Accordingly, with explicit evaluation of the time derivatives, Eqs. (A2)–(A4) now become

$$\nabla^2\left[E_S\exp\left(-i\int k_Sdz\right)\right]=-\left(\frac{\omega}{c}\right)^2\epsilon_0E_S\exp\left(-i\int k_Sdz\right),\qquad (A5)$$

$$\nabla^2\left[E_W\exp\left(-i\int\vec{k}_W\cdot d\vec{x}\right)\right]=-\left(\frac{\omega}{c}\right)^2(\epsilon_0+\tfrac{1}{2}\,\epsilon_2|E_S|^2)E\exp\left(-i\int\vec{k}_W\cdot d\vec{x}\right)$$
$$-\left(\frac{\omega}{c}\right)^2\epsilon_2E_S^2E_D^*\exp\left[-i\int(2\vec{k}_S-\vec{k}_D)\cdot d\vec{x}\right],\qquad (A6)$$

together with a third equation having $(E_W,\vec{k}_W)$, interchanged with $(E_D,\vec{k}_D)$. We now define

$$k_S(z)=(\omega/c)\epsilon_0(z)^{1/2}$$

and

$$k_W^2=k_D^2(z)=k_S^2(1+\epsilon_2|E_S|^2/2\epsilon_0),$$

such that

$$k_W(z)=k_D(z)\cong k_S(z)+g(z)$$

with

$$g(z)=(\epsilon_2(z)\,|E_S|^2/4\epsilon_0)k_S.$$

The quantity $\epsilon_2|E_S|^2/4\epsilon_0$ is the field-induced index that would be produced by E_S acting alone [given by Eq. (31), provided that α contained only $|E_S|^2$ terms]. Assuming no depletion of the strong beam, $\nabla E_S=0$, Eq. (A6) and its counterpart reduce to the coupled linear equations

$$\nabla_{\vec{k}_W}E_W\cong-ig(z)\frac{E_S{}^2}{|E_S|^2}E_D^*\exp\left(-i\int_{-d/2}^{z}\mathcal{K}(z)\,dz\right)\qquad (A7)$$

and

$$\nabla_{\vec{k}_D}E_D^*\cong ig(z)\frac{E_S^{*2}}{|E_S|^2}E_W\exp\left(i\int_{-d/2}^{z}\mathcal{K}(z)dz\right),\qquad (A8)$$

with

$$\vec{\mathcal{K}}(z) = 2\vec{k}_S - \vec{k}_W - \vec{k}_D ,$$

being directed parallel to k_S (z direction), having magnitude

$$\mathcal{K}(z) = k_S\theta^2 - 2g(z) ,$$

with θ $(= k_t/k_S)$ being the angle separating $\vec{k}_W$ and $\vec{k}_S$ (not to be compared with director angles as defined in the main body of the text).

Equations (A7) and (A8) must now be solved, subject to the boundary conditions

$$E_W(z = -\tfrac{1}{2}d) = E_W^{(0)} , \quad E_D(z = -\tfrac{1}{2}d) = 0 .$$

Quite obviously, for rapid growth in E_D^* and E_W to take place $\int_{-d/2}^{z}\mathcal{K}\,dz$ should be slowly varying in z whenever $g(z)$ is itself sizeable. Were g not spatially varying (through the spatial variation of $\underline{\epsilon}_2$ in the present problem), one could choose θ in a manner such that K would be zero. There would then be perfect phase matching, and the conversion of energy from E_S to E_W and E_D would be maximized at $\theta = \theta_{\text{opt}} = \pm(2gk_S^{-1})^{1/2}$ as is well known[14,15] in four-photon interactions in Kerr liquids. In the present problem, we choose θ so as to minimize $\mathcal{K}$ throughout most of the region of largest g,

$$\theta \cong \pm[2g(z=0)k_S^{-1}]^{1/2} .$$

Before proceeding, let us remark that if we examine intensities I_W and I_D, which are proportional to $|E_W|^2$ and $|E_D|^2$, Eqs. (A7) and (A8) rather directly reveal that

$$\nabla(|E_W|^2) = \nabla(|E_D|^2) ,$$

so that the intensities are equal, within a constant, and

$$I_W(z) = I_W(z = -\tfrac{1}{2}d) + I_D(z) , \tag{A9}$$

as is characteristic of all four-photon nonlinear optical processes[15,16] in which, ultimately, two laser photons become converted by the effectively passive medium to a pair of photons, one belonging to each of the W and D beams. As a result of Eq. (A9) it is sufficient, for present purposes, simply to obtain $E_D(z = \tfrac{1}{2}d)$, for example, which one may approach through an iterative solution of Eqs. (A7) and (A8). Our present interest concerns situations in which $\int g\,dz$ is in the neighborhood of unity, with g reaching its maximum value at $z = 0$. Choosing θ as outlined above, the phase factors in Eqs. (A7) and (A8) reduce, approximately, to the form $\exp(\pm i\int_{-d/2}^{0}\mathcal{K}(z)\,dz)$ and the solution for Eqs. (A7) and (A8), consistent with the initial conditions, may then be approximated by

$$E_W(z) \cong E_W^{(0)} \cosh \int_{-d/2}^{z} g(z)\,dz , \tag{A10}$$

$$E_D^*(z) \cong E_W^{(0)} \left[\frac{E_S^{*2}}{|E_S|^2}\exp\left(-i\int_{-d/2}^{0}\mathcal{K}(z)\,dz\right)\right] \times \sinh \int_{-d/2}^{z} g(z)\,dz . \tag{A11}$$

The intensities, following traversal of the cell, are then

$$I_D\left(\frac{d}{2}\right) = I_W\left(\frac{d}{2}\right) - I_W\left(-\frac{d}{2}\right) \cong I_W - \left(\frac{d}{2}\right)\sinh^2 \int_{-d/2}^{d/2} g(z)\,dz , \tag{A12}$$

$$\theta \cong (2g(z=0)k_s^{-1})^{1/2} .$$

If one were to choose very thin cells, for which $\int g(z)\,dz \ll 1$, and angles restricted to $\theta \ll (k_s d)^{-1/2}$ [and hence $\int \mathcal{K}(z)\,dz \ll 1$] one would obtain a response which does not vary significantly with θ for small angles

$$I_D\left(\frac{d}{2}\right) = I_W\left(\frac{d}{2}\right) - I_W\left(-\frac{d}{2}\right) \cong I_W\left(-\frac{d}{2}\right)\left(\int_{-d/2}^{d/2} g(z)\,dz\right)^2 , \tag{A13}$$

$$\int_{-d/2}^{d/2} g(z)\,dz \ll 1 , \quad \theta \ll (k_S d)^{-1/2} .$$

The integral $\int_{-d/2}^{d/2} g(z)\,dz$ appearing in this equation is, in fact, the phase shift to which was attributed the scattering described in the main body of the text.

The high degree of angular specificity implicit in the present results [Eq. (A12)] might be regarded as an undesirable feature in many applications. This specificity is reduced, in large measure, if a frequency difference is imposed upon the incident weak and strong beams, as may be inferred from the work of Chaio *et al.*[15] In applying their calculation, we assume that the relaxation of the nonlinear part of the dielectric tensor $\underline{\epsilon}_2$ is characterized by a single relaxation time τ. (This time could be very long compared with relaxation times in more usual nonlinear optical processes.) Accordingly

$$\frac{\partial \underline{\epsilon}_2(z,t)}{\partial t} = \frac{1}{\tau}\left[\underline{\epsilon}_2^{\text{s.s.}}(z) - \underline{\epsilon}_2(z,t)\right] \tag{A14}$$

represents an appropriate equation for describing the relaxation, with the superscript s.s. indicating the steady-state value. For a frequency difference between the weak and strong beams

$$\Omega = \omega_W - \omega_S ,$$

and with the diffracted beam shifted through frequency $-\Omega$ from the strong beam, $\underline{\epsilon}$ then becomes

$$\underline{\epsilon}(z,t)=\underline{\epsilon}_0+\tfrac{1}{2}\underline{\epsilon}_2^{s\cdot s\cdot}(z)\left\{|E_S|^2+\left(\frac{E_S^*E_W\exp[-i(\int(\vec{k}_W-\vec{k}_S)\cdot d\vec{x}-\Omega t)]}{1+i\Omega t}+\text{c.c.}\right)+\left(\frac{E_S^*E_D\exp[-i(\int(\vec{k}_D-\vec{k}_S)\cdot d\vec{x}+\Omega t)]}{1-i\Omega t}+\text{c.c.}\right)\right\}. \quad \text{(A15)}$$

With this expression, the methods of Ref. 14 lead directly to a two-photon scattering optical gain function for weak beam intensity

$$G(z,\Omega)=-\left(\frac{2\Omega t}{1+(\Omega t)^2}\right)g(z). \quad \text{(A16)}$$

The optical gain leads directly to exponential growth in the weak beam (and the corresponding suppression of the diffracted beam) for negative Ω (Stokes-shifted weak beam),

$$I_W(z,\Omega)=I_W\left(-\frac{d}{2}\right)\times\exp\left\{-\left(\frac{2\Omega t}{1+(\Omega t)^2}\right)\int_{-d/2}^{d/2}g(z)\,dz\right\}. \quad \text{(A17)}$$

[which is equivalent to Eq. (42) of Sec. VI] for angles in excess of $[2g(z=0)k_s^{-1}]^{1/2}$. This result is remarkable in that there exists no sensitive dependence either on θ or d, provided that the above condition on θ is met, which should provide for some ease of observation of the weak beam amplification.

[1]G. K. L. Wong and Y. R. Shen, Phys. Rev. Lett. 30, 895 (1973).

[2]G. K. L. Wong and Y. R. Shen, Phys. Rev. A 10, 1277 (1974).

[3]D. V. G. L. N. Rao and S. Jayaraman, Phys. Rev. A 10, 2457 (1974).

[4]J. R. LaLanne, Phys. Lett. A 51, 74 (1975).

[5]G. R. Alms, T. D. Gierke, and W. H. Flygare, J. Chem. Phys. 61, 4083 (1974).

[6]N. M. Amer, Y. S. Lin, and Y. R. Shen, Solid State Commun. 16, 1157 (1975).

[7]A. Saupe (private communication).

[8]H. Hervet, W. Urbach, and F. Rondelez, J. Chem. Phys. 68, 2725 (1978).

[9]V. Freedericks and A. Repiewa, Z. Phys. 42, 532 (1927).

[10]P. G. de Gennes, *The Physics of Liquid Crystals* (Clarendon, Oxford, 1974).

[11]P. Sheng, in *Introduction to Liquid Crystals*, edited by E. B. Priestly, P. J. Wojtowicz, and P. Sheng (Plenum, New York, 1974).

[12]S. I. Ben-Abraham, Phys. Rev. A 14, 1251 (1976).

[13]M. Born and E. Wolf, *Principles of Optics* (Pergamon, New York, 1959).

[14]L. M. Milne-Thomson, in *Handbook of Mathematical Functions*, edited by M. Abramowitz and I. A. Stegun (National Bureau of Standards, Washington, 1970).

[15]R. Y. Chaio, P. L. Kelley, and E. Garmire, Phys. Rev. Lett. 17, 1158 (1966).

[16]R. L. Carman, R. Y. Chaio, and P. L. Kelley, Phys. Rev. Lett. 17, 1281 (1966).