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Nonlinear refractive-index phenomena in isotropic media subjected to a dc electric field: Exact solutions

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An exact, closed-form solution is presented for the propagation characteristics of an intense, arbitrarily polarized optical beam in an isotropic medium which is subjected to a dc electric field. The related phenomena of dc-electric-field-induced optical rectification and second-harmonic generation are examined as well as the propagation characteristics of a weak beam in an isotropic medium in the presence of both the intense optical and dc fields. One fundamental and noteworthy aspect of the results presented here is that both the formalism and the resulting solutions for these third-order nonlinear optical phenomena are given explicitly and entirely in terms of the set of real, observable Stokes parameters rather than in terms of the electric field amplitudes and phases.

I. INTRODUCTION

The optical Kerr effect (OKE), first unambigously predicted by Buckingham[1] prior to the advent of the laser, was one of the earliest nonlinear optical phenomena to be observed experimentally.[2,3] The OKE or nonlinear refractive index has since been and remains a topic of much interest and study given its practical and important consequences for laser propagation characteristics[4–8] as well as its use as the basis for a wide variety of applications.[9]

It is noteworthy that the very first OKE observations, reported by Mayer and Gires[2] and by Maker, Terhune, and Savage,[3] offered a clear delineation between two characteristically distinct, almost dichotomous manifestations of the same effect. Thus, as demonstrated by the former authors, propagating an intense, linearly polarized optical pulse within an isotropic medium induces a transitory birefringence which is formally and optically equivalent to that of a (positive) uniaxial crystal with its optic axis parallel to the polarization vector of the intense "pump" beam. In such a configuration, which is literally the optical electric field analog of the dc Kerr effect, an arbitrarily polarized, weak "probe" beam will have the orientation angle, shape, and handedness of its polarization ellipse altered by the optically induced birefringence. However, as shown by Maker, Terhune, and Savage,[3] an elliptically polarized pump pulse propagating within an isotropic medium induces a refractive-index change which results in a continuous precession of the orientation angle of the polarization ellipse while leaving its shape and handedness (assuming no dichroism) unchanged.[3,8] This particular manifestation of the OKE is aptly referred to as self-induced ellipse rotation (SIER). The form of the refractive-index change induced by such an elliptically polarized pump beam does not, however, correspond to circular birefringence but rather, as will be shown in this paper, to elliptical birefringence with the important consequence that an arbitrarily polarized probe beam will, in addition to reorientation, experience change in the shape and handedness of its polarization ellipse. While the optical properties and their resulting effect on propagation characteristics have long been known and examined in detail[8,10–12] for each of these morphisms of the OKE, the more general case of simultaneous dc and optical Kerr effects has hitherto gone unsolved.

This paper presents a closed-form solution for the propagation characteristics of an arbitrarily polarized, intense optical beam within a nonabsorbing isotropic medium subjected to an arbitrary dc electric field. This solution is both general in that no restrictions are placed upon the relative strengths of the dc and optical fields as well as exact insofar as the resulting set of coupled, nonlinear equations describing the phenomenon are solved completely in terms of known transcendental functions. This general result, which contains those for SIER, the linearly polarized OKE, and the dc Kerr effect as special cases, reveals an intimate and somewhat surprising coupling between the diverse propagation characteristics of these individual nonlinear optical phenomena.

Given the sparseness of exact, closed-form solutions to general problems in the field of nonlinear optics, the mere finding of this solution is in itself noteworthy. There is, however, an ulterior motivation for possessing such a solution which further enhances its value. It is a well-established[13–16] fact that, in media which have a center-of-inversion symmetry and, in particular, non-optically-active isotropic media, the (electric dipole) second-order nonlinear susceptibility tensor is identically zero with the consequence that phenomena such as second-harmonic generation and optical rectification are normally forbidden[17] in such media. The application of a dc electric field, however, removes the center-of-inversion symmetry and thus gives rise to a number of dc-electric-field-induced nonlinear optical phenomena, the principal and most investigated of these being the phenomena of dc-induced optical rectification[18–22] (DCIOR) and dc-induced second-harmonic generation[21–28] (DCISHG). The necessary and basic prerequisite to obtaining general, exact solutions describing these effects is precisely that solution which is the subject of this paper. The equations describing DCIOR and DCISHG in isotropic media will be formulated and the nature of their solutions will be discussed with particular reference to previous experimental observations of these phenomena.

Section II deals solely with the formulation of the relevant equations which govern the propagation of an intense optical beam in an isotropic medium subjected to a dc field as well as the related equations for DCIOR and DCISHG. An essential and important feature of this development is that the final set of nonlinear differential equations are formulated not in terms of electric field amplitudes but rather in terms of a set of real, directly observable variables. Among the various candidates for such a set, the particular variables adopted in the present work are the familiar Stokes parameters. Section III presents the solution for the propagation characteristics of an intense optical wave in an isotropic medium subjected to a dc field in the case where the nonlinear susceptibility tensor components are strictly real. Several specific examples are illustrated and the nature of the solutions is discussed in some detail. Section IV examines the propagation characteristics of a probe beam in the presence of the intense optical field as well as the phenomena of DCIOR and DCISHG. Section V concludes with a discussion of the broader significance and implications of the solutions presented here to other nonlinear optical studies and in particular to the role played by the mathematical eigenfunctions described succinctly in Sec. III.

II. GENERAL FORMALISM

A. Fields and the nonlinear wave equation

The Kerr cell and coordinate geometry is illustrated in Fig. 1. A right-handed Cartesian coordinate system is defined such that the optical beams propagate parallel to and in the direction of the $+z$ axis with the entrance and exit faces of the cell lying in the $z=0$ and $z=L$ planes, respectively. The dc electric field is assumed to be uniform throughout the region $0\leq z\leq L$ and, without loss of generality, parallel to the x axis.

The real electric field within the cell, consisting of a dc field, an intense pump beam of frequency ω, and a weak probe beam of frequency ω', is expressed as

$$\vec{E}(z,t)=E_0\hat{x}+\mathrm{Re}[\vec{E}(\omega,z)\exp(ikz-i\omega t)+\vec{E}(\omega',z)\exp(ik'z-i\omega' t)]\ , \tag{1}$$

where $k=k(\omega)=n(\omega)\omega/c$, E_0 is the dc field amplitude, and where the Fourier spectral amplitudes $\vec{E}(\omega,z)$ and

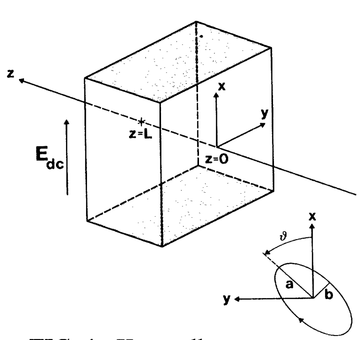

FIG. 1. Kerr cell geometry.

$\vec{E}(\omega',z)$ [with $|\vec{E}(\omega',z)|\ll|\vec{E}(\omega,z)|$] are assumed to be slowly varying with respect to the phase factors $\exp(ikz)$ and $\exp(ik'z)$. Although only the z dependence is explicitly expressed, it is permissible, within limitations,[29,30] to assume an implicit, slowly varying x,y dependence for the spectral amplitudes. The reality of the electric field requires that the spectral amplitudes obey the condition $[\vec{E}(\omega,z)]^*=\vec{E}(-\omega,z)$.

The nonlinear wave equation for the electric field in a nonmagnetic dielectric in the absence of currents or free charges is[13–16,22]

$$\frac{\partial}{\partial z}\vec{E}(\omega_q,z)=\frac{2\pi i\omega}{n(\omega)c}\vec{P}^{\,\mathrm{nl}}(\omega_q,z)\ , \tag{2}$$

where $\vec{P}^{\,\mathrm{nl}}(\omega_q,z)$ is the spectral amplitude of the nonlinear polarization. In isotropic media, the dominant nonlinearity is the third-order, electric dipole polarization given by the expression[15,16]

$$P_i^{\mathrm{nl}}(\omega_q,z)=D_{rst}\chi_3^{ijkl}(\omega_q;\omega_r,\omega_s,\omega_t)E_j(\omega_r,z)\times E_k(\omega_s,z)E_l(\omega_t,z)e^{i\Delta kz}\ , \tag{3}$$

where

$$\begin{aligned}\omega_q&=\omega_r+\omega_s+\omega_t\ ,\\ \Delta k&=k_r+k_s+k_t-k_q\ ,\end{aligned} \tag{4}$$

and where the degeneracy factor D_{rst} is

$$D_{rst}=6+4\delta_{rs}\delta_{rt}\delta_{st}-3(\delta_{rs}+\delta_{rt}+\delta_{st})\ , \tag{5}$$

with $D_{rst}=6$, 3, or 1 according to whether none, two, or all three of the frequencies are equal, respectively. With the inclusion of this factor, it is implicitly understood that expansion of Eq. (3) is to include only distinct combinations of ω_r, ω_s, and ω_t and not their permutations.

The third-order nonlinear susceptibility tensor $\chi_3(\omega_q;\omega_r,\omega_s,\omega_t)$ obeys a reality condition[15]

$$[\chi_3^{ijkl}(\omega_q;\omega_r,\omega_s,\omega_t)]^*=\chi_3^{ijkl}(-\omega_q;-\omega_r,-\omega_s,-\omega_t) \tag{6}$$

and possesses an intrinsic permutation symmetry[13–16,31,32] which states that its value is invariant under the 3! permutations of the pairs $j\omega_r$, $k\omega_s$, and $l\omega_t$ (this symmetry may be extended to the 4! permutations which include the pair $i\omega_q$ if and only if none of the four frequencies coincides with a resonant frequency of the medium). In addition, the susceptibility tensor must conform to the spatial symmetries of the medium it describes. Lists of the nonzero and independent elements for tensor orders 1 to 3 in all crystallographic groups have been tabulated by Butcher,[15] Kielich,[33] and Flytzanis.[31] In isotropic media, there are 21 nonzero elements of χ_3 of which only three are independent. Degeneracy in the frequencies will further reduce the number of independent elements to two in the case of self-induced phenomena ($\omega_r=\omega_s\doteq-\omega_t$) and to one for the completely degenerate case of third-harmonic generation ($\omega_r=\omega_s=\omega_t$). Detailed descriptions of the third-order nonlinear susceptibility tensor which, within a classical framework, delineate the nuclear and electronic contributions to χ_3 have been given by Flytzanis,[31] Hellwarth,[32] Owyoung,[12] and by Kasprowicz-Kielich and Kielich.[34]

B. Polarization and the Stokes parameters

The transversely polarized, complex Fourier spectral amplitude at ω is expressed as

$$E_{x,y}(\omega,z)=A_{x,y}(\omega,z)\exp[i\epsilon_{x,y}(\omega,z)] \,, \tag{7}$$

where the functions A_x, A_y, ϵ_x, and ϵ_y are strictly real and where, with reference to Eq. (1), the phase angles are defined positive with respect to $kz-\omega t$. The amplitude of $E(\omega,z)$ is defined as

$$\begin{aligned} A(\omega,z)&=[\,|E_x(\omega,z)|^2+|E_y(\omega,z)|^2]^{1/2}\\ &=[A_x^2(\omega,z)+A_y^2(\omega,z)]^{1/2} \end{aligned} \tag{8}$$

so that $A(\omega,z)\geq 0$. When $A_0=A(\omega,0)$ is >0, it is possible to normalize the spectral amplitude components to their values at $z=0$, i.e.,

$$E_{x,y}(\omega,z)/A_0=a_{x,y}(\omega,z)\exp[i\epsilon_{x,y}(\omega,z)] \,, \tag{9}$$

with a_x and a_y referred to as the normalized (dimensionless) amplitudes.

An alternate representation for $E(\omega,z)$, and the one favored in the present work, is in terms of its normalized right- and left-circularly polarized amplitudes[35] given by

$$\begin{aligned} E_r(\omega,z)/A_0&=a_r(\omega,z)\exp[i\epsilon_r(\omega,z)]\\ &=2^{-1/2}[E_x(\omega,z)+iE_y(\omega,z)]/A_0 \,,\\ E_l(\omega,z)/A_0&=a_l(\omega,z)\exp[i\epsilon_l(\omega,z)]\\ &=2^{-1/2}[E_x(\omega,z)-iE_y(\omega,z)]/A_0 \,, \end{aligned} \tag{10}$$

with corresponding unit vectors[36]

$$\begin{aligned} \hat{e}_r&=2^{-1/2}(\hat{x}-i\hat{y}) \,,\\ \hat{e}_l&=2^{-1/2}(\hat{x}+i\hat{y}) \end{aligned} \tag{11}$$

so that

$$\vec{E}(\omega,z)=E_x(\omega,z)\hat{x}+E_y(\omega,z)\hat{y}=E_r(\omega,z)\hat{e}_r+E_l(\omega,z)\hat{e}_l \,.$$

The normalized Stokes parameters of the optical wave at frequency ω are defined to be[37]

$$\begin{aligned} s_0(\omega,z)&=a_x^2+a_y^2=a_r^2+a_l^2 \,,\\ s_1(\omega,z)&=a_x^2-a_y^2=2a_ra_l\cos\delta \,,\\ s_2(\omega,z)&=2a_xa_y\cos\gamma=2a_ra_l\sin\delta \,,\\ s_3(\omega,z)&=2a_xa_y\sin\gamma=a_l^2-a_r^2 \,, \end{aligned} \tag{12}$$

where

$$\begin{aligned} \gamma(\omega,z)&=\epsilon_y(\omega,z)-\epsilon_x(\omega,z) \,,\\ \delta(\omega,z)&=\epsilon_r(\omega,z)-\epsilon_l(\omega,z) \end{aligned} \tag{13}$$

are the phase differences. Only three of the four Stokes parameters are independent by virture of the identity[38]

$$s_0^2=s_1^2+s_2^2+s_3^2 \,. \tag{14}$$

The polarization state, i.e., the shape, orientation, and handedness of the polarization ellipse, is given uniquely and simply in terms of the Stokes parameters. Thus the polarization parameter defined as

$$P(\omega,z)=s_3/s_0, \quad -1\leq P\leq +1 \tag{15}$$

is >0 for right-handed elliptical, <0 for left-handed elliptical, $=0$ for linear polarization, and $+1$ and -1 for right- and left-handed circular, respectively. The ratio of the semiminor axis to the semimajor axis is given by

$$b/a=\tan(\tfrac{1}{2}\sin^{-1}|s_3/s_0|\,), \quad 0\leq b/a\leq 1 \,. \tag{16}$$

The angle of the ellipse major axis, measured positive from the $+x$ axis towards the $+y$ axis, is specified by

$$\vartheta(\omega,z)=\tfrac{1}{2}\delta(\omega,z)=\tfrac{1}{2}\tan^{-1}(s_2/s_1), \quad -\frac{\pi}{2}\leq\vartheta\leq+\frac{\pi}{2} \tag{17}$$

where the individual signs of s_1 and s_2 must be taken into consideration in correctly evaluating the arctan function.

In the case where $A_0=0$, Eq. (12) is used to define the unnormalized Stokes parameters S_0, S_1, S_2, and S_3 by simply replacing all the normalized a amplitudes with their absolute A counterparts, noting that Eqs. (14)—(17) are valid for either set of parameters.

C. Propagation of the pump wave

The nonlinear polarization at ω is composed of self-induced contributions via $\chi_3(\omega;\omega,\omega,-\omega)$ which, for the present, is allowed to be complex valued, and dc-induced contributions via $\chi_3(\omega;\omega,0,0)$ which is assumed strictly real. Expanding Eq. (3) with $\omega_q=\omega$ results in

$$\begin{aligned} P_x^{\rm nl}(\omega,z)=&\,6\chi_3^{1122}(\omega;\omega,\omega,-\omega)\,|\vec{E}(\omega,z)|^2E_x(\omega,z)+3\chi_3^{1221}(\omega;\omega,\omega,-\omega)[\vec{E}(\omega,z)]^2E_x^*(\omega,z)\\ &+[6\chi_3^{1221}(\omega;\omega,0,0)+3\chi_3^{1122}(\omega;\omega,0,0)]E_0^2E_x(\omega,z) \,, \end{aligned} \tag{18}$$

$$\begin{aligned} P_y^{\rm nl}(\omega,z)=&\,6\chi_3^{1122}(\omega;\omega,\omega,-\omega)\,|\vec{E}(\omega,z)|^2E_y(\omega,z)+3\chi_3^{1221}(\omega;\omega,\omega,-\omega)[\vec{E}(\omega,z)]^2E_y^*(\omega,z)\\ &+3\chi_3^{1122}(\omega;\omega,0,0)E_0^2E_y(\omega,z) \end{aligned} \tag{19}$$

which in turn, using the relation $[\vec{E}(\omega,z)]^2=2E_r(\omega,z)E_l(\omega,z)$, leads directly to the expression

$$\begin{aligned} P_{r,l}^{\rm nl}(\omega,z)=&\,6\chi_3^{1122}(\omega;\omega,\omega,-\omega)[\,|E_r(\omega,z)|^2+|E_l(\omega,z)|^2]E_{r,l}(\omega,z)\\ &+6\chi_3^{1221}(\omega;\omega,\omega,-\omega)\,|E_{l,r}(\omega,z)|^2E_{r,l}(\omega,z)+3\chi_3^{1122}(\omega;\omega,0,0)E_0^2E_{r,l}(\omega,z)\\ &+3\chi_3^{1221}(\omega;\omega,0,0)E_0^2[E_r(\omega,z)+E_l(\omega,z)] \,. \end{aligned} \tag{20}$$

Since the effect of DCIOR on the magnitude of E_0 is negligible,[20] the value of E_0 is assumed constant in these expressions. Note that alternate, equivalent expressions for $\chi_3(\omega;\omega,0,0)$ are possible, e.g., $\chi_3^{1221}(\omega;0,0,\omega)=\chi_3^{1122}(\omega;\omega,0,0)$ and $\chi_3^{1122}(\omega;0,0,\omega)=\chi_3^{1221}(\omega;\omega,0,0)$. Also note the degeneracy $\chi_3^{1212}(\omega;\omega,\omega,-\omega)=\chi_3^{1122}(\omega;\omega,\omega,-\omega)$.

The first step towards obtaining differential equations in terms of the Stokes parameters consists of substituting Eq. (20) into Eq. (2) and then separating the results into its real and imaginary parts for the amplitude and phase functions defined by Eq. (10). This gives

$$\frac{d}{dz}a_r(\omega,z)=-J_3(a_r^2+a_l^2)a_r-J_1a_l^2a_r+\tfrac{1}{2}R_0a_l\sin\delta \,, \tag{21a}$$

$$\frac{d}{dz}a_l(\omega,z)=-J_3(a_r^2+a_l^2)a_l-J_1a_r^2a_l-\tfrac{1}{2}R_0a_r\sin\delta \,, \tag{21b}$$

$$\frac{d}{dz}\delta(\omega,z)=R_1(a_l^2-a_r^2)+\tfrac{1}{2}R_0(a_l^2-a_r^2)(\cos\delta)/(a_ra_l) \,, \tag{21c}$$

where the derivative is written as d/dz in place of the strictly rigorous partial $\partial/\partial z$ (i.e., the frequency is treated purely as a parameter). Although not needed in the immediate case, the equation governing the sum of the phases, $\sigma(\omega,z)=\epsilon_r(\omega,z)+\epsilon_l(\omega,z)$, is also given at this point (this result is used for the DCISHG equations).

$$\frac{d}{dz}\sigma(\omega,z)=R_0+R_4+(R_1+2R_3)(a_r^2+a_l^2)+\tfrac{1}{2}R_0(a_r^2+a_l^2)(\cos\delta)/(a_ra_l) \,, \tag{21d}$$

where, for notational brevity, the constants

$$\begin{aligned}
R_0&=\frac{12\pi\omega}{n(\omega)c}\chi_3^{1221}(\omega;\omega,0,0)E_0^2 \,,\\
R_1&=\frac{12\pi\omega}{n(\omega)c}\mathrm{Re}[\chi_3^{1221}(\omega;\omega,\omega,-\omega)]A_0^2 \,,\\
R_3&=\frac{12\pi\omega}{n(\omega)c}\mathrm{Re}[\chi_3^{1212}(\omega;\omega,\omega,-\omega)]A_0^2 \,,\\
R_4&=\frac{12\pi\omega}{n(\omega)c}\chi_3^{1122}(\omega;\omega,0,0)E_0^2 \,,\\
J_1&=\frac{12\pi\omega}{n(\omega)c}\mathrm{Im}[\chi_3^{1221}(\omega;\omega,\omega,-\omega)]A_0^2 \,,\\
J_3&=\frac{12\pi\omega}{n(\omega)c}\mathrm{Im}[\chi_3^{1212}(\omega;\omega,\omega,-\omega)]A_0^2
\end{aligned} \tag{22}$$

have been introduced with the R_i's denoting terms which contain the real or real parts of the susceptibilities and the J_i's incorporating the imaginary parts of $\chi_3(\omega;\omega,\omega,-\omega)$. The dimensions of the R and J coefficients are $(\text{length})^{-1}$.

With Eqs. (21) in hand, it is a straightforward task to derive the final nonlinear differential equations expressed in terms of the normalized Stokes parameters of Eq. (12). They are

$$\frac{d}{dz}s_0(\omega,z)=-(J_1+2J_3)s_0^2+J_1s_3^2 \,, \tag{23a}$$

$$\frac{d}{dz}s_1(\omega,z)=-(J_1+2J_3)s_0s_1-R_1s_2s_3 \,, \tag{23b}$$

$$\frac{d}{dz}s_2(\omega,z)=-(J_1+2J_3)s_0s_2+R_0s_3+R_1s_1s_3 \,, \tag{23c}$$

$$\frac{d}{dz}s_3(\omega,z)=-2J_3s_0s_3-R_0s_2 \,. \tag{23d}$$

The detailed solution to this set of nonlinear equations in the case of strictly real $\chi_3(\omega;\omega,\omega,-\omega)$ is dealt with in Sec. III. It is both significant and encouraging that the transformation of the nonlinear equations from dependence on field amplitudes and phases to dependence on the Stokes parameters is in fact a complete transformation.

D. Propagation of the probe wave

The nonlinear polarization at the probe frequency ω' is given by

$$\begin{aligned}
P_{r,l}^{\mathrm{nl}}(\omega',z)&=3(2-\delta_{\omega\omega'})\chi_3^{1122}(\omega';\omega',\omega,-\omega)\,|\vec{\mathrm{E}}(\omega,z)|^2E_{r,l}(\omega',z)\\
&\quad+3(2-\delta_{\omega\omega'})\chi_3^{1212}(\omega';\omega',\omega,-\omega)[\vec{\mathrm{E}}(\omega',z)\cdot\vec{\mathrm{E}}^*(\omega,z)]E_{r,l}(\omega,z)\\
&\quad+3(2-\delta_{\omega\omega'})\chi_3^{1221}(\omega';\omega',\omega,-\omega)[\vec{\mathrm{E}}(\omega',z)\cdot\vec{\mathrm{E}}(\omega,z)]E_{l,r}^*(\omega,z)\\
&\quad+3\chi_3^{1122}(\omega';\omega',0,0)E_0^2E_{r,l}(\omega',z)+3\chi_3^{1221}(\omega';\omega',0,0)E_0^2[E_r(\omega',z)+E_l(\omega',z)] \,.
\end{aligned} \tag{24}$$

The equations governing the propagation of the wave at frequency ω' are derived in a manner exactly parallel to that outlined above for the case of the pump wave. The final result takes the form

$$\frac{d}{dz}s_0'=-\tfrac{1}{2}(J_1'+J_3'+2J_5')s_0s_0'+\tfrac{1}{2}(J_1'-J_3')s_3s_3'-\tfrac{1}{2}(J_1'+J_3')(s_1s_1'+s_2s_2') \,, \tag{25a}$$

$$\frac{d}{dz}s_1'=-\tfrac{1}{2}(J_1'+J_3'+2J_5')s_0s_1'-\tfrac{1}{2}(J_1'+J_3')s_1s_0'-\tfrac{1}{2}(R_1'+R_3')s_2s_3'-\tfrac{1}{2}(R_1'-R_3')s_3s_2' \,, \tag{25b}$$

$$\frac{d}{dz}s_2'=-\tfrac{1}{2}(J_1'+J_3'+2J_5')s_0s_2'-\tfrac{1}{2}(J_1'+J_3')s_2s_0'+R_0's_3'+\tfrac{1}{2}(R_1'+R_3')s_1s_3'+\tfrac{1}{2}(R_1'-R_3')s_3s_1' \,, \tag{25c}$$

$$\frac{d}{dz}s_3' = -\tfrac{1}{2}(J_1' + J_3' + 2J_5')s_0 s_3' + \tfrac{1}{2}(J_1' - J_3')s_3 s_0' - R_0' s_2' - \tfrac{1}{2}(R_1' + R_3')(s_1 s_2' - s_2 s_1') \;, \tag{25d}$$

where $s_i' = s_i(\omega',z)$ and where the constants R_i' and J_i' are

$$\begin{aligned}
R_0' &= \frac{12\pi\omega'}{n(\omega')c}\chi_3^{1221}(\omega';\omega',0,0)E_0^2 \;,\\
R_1' &= \frac{12\pi\omega'}{n(\omega')c}(2-\delta_{\omega\omega'})\mathrm{Re}[\chi_3^{1221}(\omega';\omega',\omega,-\omega)]A_0^2 \;,\\
R_3' &= \frac{12\pi\omega'}{n(\omega')c}(2-\delta_{\omega\omega'})\mathrm{Re}[\chi_3^{1212}(\omega';\omega',\omega,-\omega)]A_0^2 \;,\\
J_1' &= \frac{12\pi\omega'}{n(\omega')c}(2-\delta_{\omega\omega'})\mathrm{Im}[\chi_3^{1221}(\omega';\omega',\omega,-\omega)]A_0^2 \;,\\
J_3' &= \frac{12\pi\omega'}{n(\omega')c}(2-\delta_{\omega\omega'})\mathrm{Im}[\chi_3^{1212}(\omega';\omega',\omega,-\omega)]A_0^2 \;,\\
J_5' &= \frac{12\pi\omega'}{n(\omega')c}(2-\delta_{\omega\omega'})\mathrm{Im}[\chi_3^{1122}(\omega';\omega',\omega,-\omega)]A_0^2 \;.
\end{aligned} \tag{26}$$

The degeneracy term $(2-\delta_{\omega\omega'})$ is included in these definitions to ensure that Eqs. (25) reduce to Eqs. (23) in the limit of $\omega'=\omega$.

E. dc-induced second-harmonic generation

The normally forbidden phenomenon of second-harmonic generation in an isotropic medium is made possible by the presence of the dc electric field and the nonlinear susceptibility $\chi_3(2\omega;0,\omega,\omega)$. The circular components of the nonlinear polarization at 2ω are given by

$$e^{-i\Delta kz}P_{r,l}^{\mathrm{nl}}(2\omega,z) = 2^{1/2}3\chi_3^{1122}(2\omega;0,\omega,\omega)E_0E_r(\omega,z)E_l(\omega,z) + 2^{1/2}3\chi_3^{1221}(2\omega;0,\omega,\omega)E_0E_{r,l}(\omega,z)[E_r(\omega,z)+E_l(\omega,z)] \;, \tag{27}$$

where

$$\Delta k = (2\omega/c)[n(\omega)-n(2\omega)] \;. \tag{28}$$

Taking $\chi_3(2\omega;0,\omega,\omega)$ to be strictly real and ignoring pump depletion, the nonlinear wave equation (2) at the second-harmonic frequency 2ω, when separated into its real and imaginary parts, leads to

$$\frac{d}{dz}\{A_r(2\omega,z)\cos[\epsilon_r(2\omega,z)]\} = -F_1(z) = -V_1a_r^2\sin(\sigma+\delta+\Delta kz) - (V_1+V_2)a_ra_l\sin(\sigma+\Delta kz) \;, \tag{29a}$$

$$\frac{d}{dz}\{A_r(2\omega,z)\sin[\epsilon_r(2\omega,z)]\} = F_2(z) = V_1a_r^2\cos(\sigma+\delta+\Delta kz) + (V_1+V_2)a_ra_l\cos(\sigma+\Delta kz) \;, \tag{29b}$$

$$\frac{d}{dz}\{A_l(2\omega,z)\cos[\epsilon_l(2\omega,z)]\} = -F_3(z) = -V_1a_l^2\sin(\sigma-\delta+\Delta kz) - (V_1+V_2)a_ra_l\sin(\sigma+\Delta kz) \;, \tag{29c}$$

$$\frac{d}{dz}\{A_l(2\omega,z)\sin[\epsilon_l(2\omega,z)]\} = F_4(z) = V_1a_l^2\cos(\sigma-\delta+\Delta kz) + (V_1+V_2)a_ra_l\cos(\sigma+\Delta kz) \;, \tag{29d}$$

where

$$\begin{aligned}
V_1 &= \frac{2^{1/2}12\pi\omega}{n(2\omega)c}\chi_3^{1221}(2\omega;0,\omega,\omega)E_0A_0^2 \;,\\
V_2 &= \frac{2^{1/2}12\pi\omega}{n(2\omega)c}\chi_3^{1122}(2\omega;0,\omega,\omega)E_0A_0^2 \;.
\end{aligned} \tag{30}$$

In principal, Eqs. (29) may be integrated to give $A_{r,l}(2\omega,z)$ and $\epsilon_{r,l}(2\omega,z)$ once the Stokes parameters as functions of z and hence the functions F_1–F_4 are known.

F. dc-induced optical rectification

The intense optical wave at ω induces a dc polarization through $\chi_3(0;0,\omega,-\omega)$, here assumed strictly real, for which the component parallel to $\vec{E}_0$ is given by

$$P_x^{\mathrm{nl}}(0,z) = 6[\chi_3^{1212}(0;0,\omega,-\omega)+\chi_3^{1221}(0;0,\omega,-\omega)] \times E_0E_x(\omega,z)E_x^*(\omega,z) + 6\chi_3^{1122}(0;0,\omega,-\omega) \times E_0\,|\vec{E}(\omega,z)|^2 \;. \tag{31}$$

Since the beam at ω is a cw source, the observed dc voltage resulting from this polarization is proportional to the integral of Eq. (31), i.e.,

$$V_{\mathrm{dc}} = 3LE_0A_0^2g\left[(\chi_3^{1111}+\chi_3^{1122})\frac{1}{L}\int_0^L s_0\,dz + (\chi_3^{1111}-\chi_3^{1122})\frac{1}{L}\int_0^L s_1\,dz\right] \;, \tag{32}$$

where g is factor dependent upon the cell and beam geometry.[15,18–20]

III. EXACT RESULTS FOR REAL $\chi_3(\omega;\omega,\omega,-\omega)$

A. General solution

For a medium transparent at frequencies ω and 2ω, then $\chi_3(\omega;\omega,\omega,-\omega)$ is strictly real ($J_1=J_3=0$) whence $s_0(\omega,z)$ is constant and $=1$. Equations (23) simplify to

$$\frac{d}{dz}s_1(\omega,z) = -R_1s_2s_3 \;, \tag{33a}$$

$$\frac{d}{dz}s_2(\omega,z) = R_1s_1s_3 + R_0s_3 \;, \tag{33b}$$

$$\frac{d}{dz}s_3(\omega,z) = -R_0s_2 \;, \tag{33c}$$

with

$$s_1^2+s_2^2+s_3^2=1 \ . \tag{34}$$

An immediate consequence of Eqs. (33a) and (33c) is the identity

$$\frac{d}{dz}(2R_0s_1-R_1s_3^2)=0 \ , \tag{35a}$$

that is,

$$2R_0(s_1-s_{10})=R_1(s_3^2-s_{30}^2) \ , \tag{35b}$$

where s_{i0} denotes $s_i(\omega,0)$. Together, Eqs. (34) and (35) reveal that only one of the three Stokes parameters of Eq. (33) is independent. The following will work specifically with s_3 although an exactly similar procedure would be followed for either s_1 or s_2.

Squaring Eq. (33c) and invoking Eqs. (34) and (35) to eliminate s_1 and s_2 leads to the first-order, second-degree nonlinear equation

$$\left[\frac{d}{dz}s_3\right]^2=-B_1s_3^4-B_2s_3^2+B_3 \ , \tag{36}$$

where

$$\begin{aligned} B_1&=\tfrac{1}{4}R_1^2 \ , \\ B_2&=R_0^2+R_1(R_0s_{10}-\tfrac{1}{2}R_1s_{30}^2) \ , \\ B_3&=R_0^2-(R_0s_{10}-\tfrac{1}{2}R_1s_{30}^2)^2 \ . \end{aligned} \tag{37}$$

The following properties of these B_i coefficients are noted as

$$B_1\geq 0 \ , \tag{38a}$$

$$B_2^2+4B_1B_3\geq 0 \ , \tag{38b}$$

$$B_3<0\rightarrow B_2<0 \ . \tag{38c}$$

The second-order, first-degree nonlinear differential equation for s_3 equivalent to Eq. (36) is

$$\frac{d^2}{dz^2}s_3=-2B_1s_3^3-B_2s_3 \ , \tag{39}$$

which illustrates clearly that the essential nonlinearity is that arising from the R_1 term, i.e., the self-induced contribution.

Factoring the right-hand side, Eq. (36) may be rewritten as

$$\left[\frac{d}{dz}s_3\right]^2=-B_1(s_3^2-\alpha_1)(s_3^2-\alpha_2) \ , \tag{40}$$

where the roots $\alpha_{1,2}$ are

$$\begin{aligned} \alpha_{1,2}&=-(B_2/2B_1)\pm[(B_2/2B_1)^2+(B_3/B_1)]^{1/2} \\ &=[r^2s_{30}^2-2(1+rs_{10})]/r^2 \\ &\qquad\pm(2/r^2)[(1+rs_{10})^2+r^2s_{20}^2]^{1/2} \end{aligned} \tag{41}$$

and where the dimensionless ratio r is defined to be

$$\begin{aligned} r&=R_1/R_0 \\ &=[\chi_3^{1221}(\omega;\omega,\omega,-\omega)A_0^2]/[\chi_3^{1221}(\omega;\omega,0,0)E_0^2] \ . \end{aligned} \tag{42}$$

It may be assumed, without loss of generality, that $r\geq 0$ (this point is discussed in more detail in Sec. V). It follows from Eqs. (38) that both α_1 and α_2 are strictly real and that $0\leq\alpha_1\leq 1$ for all $r\geq 0$. When $s_{10}>0$, then α_1 has no local extremum but simply increases or decreases monotonically from $\alpha_1(r=0)=1-s_{10}^2$ to $\alpha_1(r=\infty)=s_{30}^2$. If $s_{10}<0$, however, then $\alpha_1=1$ when $r=-2s_{10}/(1-s_{30}^2)$ for $s_{30}\neq 1$. The behavior of α_2 is somewhat simpler with $\alpha_2(r=0)=-\infty$, $\alpha_2(r=\infty)=s_{30}^2$, and $d\alpha_2/dr>0$ for all $r\geq 0$. Thus α_2 is a strictly monotonically increasing function of r which changes its sign at $r=2(1+s_{10})/s_{30}^2$ when $s_{30}\neq 0$. One specific case which merits special attention is that of $s_{20}=0$ for which (a) $s_{20}=0$, $s_{10}>0$

$$\begin{aligned} \alpha_1&=s_{30}^2 \ , \\ \alpha_2&=s_{30}^2-4(1+rs_{10})/r^2 \ , \end{aligned} \tag{43}$$

and (b) $s_{20}=0$, $s_{10}<0$

$$\begin{aligned} \alpha_1&=s_{30}^2 \ , \\ \alpha_2&=s_{30}^2-4(1+rs_{10})/r^2, \quad r<-1/s_{10} \\ \alpha_1&=s_{30}^2-4(1+rs_{10})/r^2 \ , \\ \alpha_2&=s_{30}^2, \quad r>-1/s_{10} \end{aligned} \tag{44}$$

noting in particular the "flip-flop" behavior of $\alpha_{1,2}$ when $s_{10}<0$.

The solution for s_3, which follows directly upon integration of Eq. (40) and the subsequent inversion of the resulting elliptic integral,[39] takes the form of the Jacobian elliptic function[39–41] cn [the sn solution is ruled out by the condition (38a)]. Specifically, the result is found to be

$$rs_3(\omega,z)=2pkf\,\mathrm{cn}(fR_0z+c;m) \ , \tag{45}$$

where

$$\begin{aligned} f&=[(1+rs_{10})^2+r^2s_{20}^2]^{1/4} \ , \\ m&=\alpha_1/(\alpha_1-\alpha_2)=\tfrac{1}{2}+(r^2-1-f^4)/4f^2 \ , \end{aligned} \tag{46}$$

and

$$-\mathrm{Re}[K(m)]\leq c\leq +\mathrm{Re}[K(m)] \ , \tag{47}$$

with $K(m)$ denoting the Jacobian quarter-period, $k=m^{1/2}$, $p=\pm 1=\mathrm{sgn}(s_{30})$, and with the sign function defined as $\mathrm{sgn}(x)=1$ for $x\geq 0$ and $\mathrm{sgn}(x)=-1$ for $x<0$. As points of notation, note that the Jacobian elliptic functions and periods are here expressed as dependent upon the Jacobian parameter m (many authors write this dependence in terms of the modulus $k=+m^{1/2}$). Secondly, although deference to tradition requires that the symbol k be used to represent both the wave number as well as the Jacobian modulus, its meaning will always be clear by context. The solutions for s_1 and s_2 follow immediately from Eqs. (35b) and (33c) and take the form

$$rs_1(\omega,z)=f^2[1-2m\,\mathrm{sn}^2(fR_0z+c;m)]-1 \ , \tag{48}$$

$$rs_2(\omega,z)=2pkf^2\,\mathrm{sn}(fR_0z+c;m)\mathrm{dn}(fR_0z+c;m) \tag{49}$$

so that the constant c is given in terms of the initial conditions according to

$$\mathrm{cn}(c;m)=r\,|s_{30}|/2kf\ ,$$
$$\mathrm{sgn}(c)=\mathrm{sgn}(ps_{20})\ . \tag{50}$$

The propagation characteristics of the optical beam depend critically upon the Jacobian parameter m which in turn is dependent on the initial values of the Stokes parameters and the ratio r according to

$$m=\tfrac{1}{2}+\frac{r^2s_{30}^2-2(1+rs_{10})}{4[(1+rs_{10})^2+r^2s_{20}^2]^{1/2}} \tag{51}$$

which, for small values of r, may be expanded as

$$m=\tfrac{1}{4}(1-s_{10}^2)r^2-\tfrac{1}{2}s_{10}(1-s_{10}^2-\tfrac{1}{2}s_{30}^2)r^3+\cdots\ . \tag{52}$$

It follows from the discussion above of the $\alpha_1(r)$ and $\alpha_2(r)$ dependences that $m\geq 0$ for all values of $r\geq 0$. However, values of $m>1$ do occur and it is for this reason that Eq. (47) stipulates the real part of the Jacobian quarter-period $K(m)$. These solutions as given are valid for all initial conditions and values of r for which m is defined; the only instance for which m is not defined is the special case [from Eq. (44)] $s_{20}=0$, $s_{10}<0$, $r=-1/s_{10}$ although, even in this case, Eqs. (45)–(50) yield the correct (trivial) solution by taking the limit as r approaches $-1/s_{10}$. The variation of the parameter m with r is illustrated in Fig. 2 for five different initial polarization states. Note, in particular, examples 4 and 5 in this figure which illustrate the two cases where $s_{20}=0$, Eqs. (43) and (44), respectively; the radically different $m(r)$ dependences result from simply changing the sign of s_{10} (i.e., the initial polarization ellipse is rotated by $\pi/2$).

It is possible to express the solutions for the Stokes parameters in a very concise form in terms of the "Chebyshev-like (Jacobian) elliptic functions" (or CLEF) con and son defined as

$$\mathrm{con}_j(x;m)=\cos\{j[\mathrm{am}(x;m)]\}\ ,\qquad j=0,\pm1,\pm2,\ldots$$
$$\mathrm{son}_j(x;m)=\sin\{j[\mathrm{am}(x;m)]\}\ , \tag{53}$$

where $\mathrm{am}(x;m)$ is the Jacobian amplitude function.[39–42] The CLEF constitute an eigenfunction sequence which is complete and orthogonal on $L^2(-2\,\mathrm{Re}[K(m)],\ 2\mathrm{Re}[K(m)])$ but, unlike its degenerate ($m=0$) trigonometric counterpart $\{\cos(nx),\sin(nx);\ n=0,\pm1,\pm2,\ldots\}$ does not exhibit geometrically decreasing harmonic periods for its elements. In terms of the CLEF, the solutions given above may be reexpressed compactly as

$$rs_1(\omega,z)=f^2\,\mathrm{con}_2((fR_0/k)z+c;1/m)-1\ ,$$
$$rs_2(\omega,z)=pf^2\,\mathrm{son}_2((fR_0/k)z+c;1/m)\ , \tag{54}$$
$$rs_3(\omega,z)=2pkf\,\mathrm{con}_1(fR_0z+c;m)\ .$$

The use of these functions amounts to much more than simply a form of mathematical "shorthand." Particular elements of this CLEF sequence ($j=0$–3) have appeared as solutions to problems in nonlinear optics (and, indeed, in a number of other nonlinear studies) with the earliest example being the general solutions reported by Armstrong *et al.*[13] In addition, degenerate cases of the CLEF, most particularly when $m=1$, have also been "invoked" in several studies including those concerned primarily with soliton solutions to propagation problems.[43] Such results lend corroboration to the underlying belief that the CLEF defined in Eq. (53) may prove to play a fundamental and important role in describing nonlinear optical phenomena. A detailed description of these special functions will be published separately.

The case of SIER only follows by allowing the parameter r to approach ∞, with the result

$$s_1(\omega,z)=2a_{r0}a_{l0}\cos(\delta_0-R_1s_{30}z)\ ,$$
$$s_2(\omega,z)=2a_{r0}a_{l0}\sin(\delta_0-R_1s_{30}z)\ , \tag{55}$$
$$s_3(\omega,z)=s_{30}\ ,$$

indicating, as stated in the Introduction, that the polarization ellipse for ω is rotated uniformly without distortion. The case of the dc Kerr effect only follows in the limit $r=0$, with the result

$$s_1(\omega,z)=s_{10}\ ,$$
$$s_2(\omega,z)=2a_{x0}a_{y0}\cos(\gamma_0+R_0z)\ , \tag{56}$$
$$s_3(\omega,z)=2a_{x0}a_{y0}\sin(\gamma_0+R_0z)\ ,$$

revealing that, in general, the shape, handedness, and orientation of the beam at ω will be altered. A final special case considered at this point treats r as arbitrary but assumes that the incident beam is linearly polarized, i.e., $s_{30}=0$. For this particular initial condition, the solution takes the form

$$s_1(\omega,z)=s_{10}+(rs_{20}^2/2f^2)\mathrm{sd}^2(fR_0z;m)\ ,$$
$$s_2(\omega,z)=s_{20}\mathrm{cd}(fR_0z;m)\mathrm{nd}(fR_0z;m)\ , \tag{57}$$
$$s_3(\omega,z)=-(s_{20}/f)\mathrm{sd}(fR_0z;m)\ ,$$

noting that $0\leq m<1$ when $s_{30}=0$.

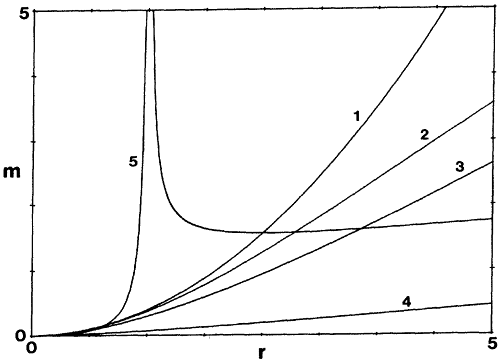

FIG. 2. Jacobian parameter m as a function of the ratio r for initial polarization states s_{10}, s_{20}, and s_{30} of (1) 0,0,1; (2) 0,0.28,0.96; (3) 0.2,0.2,0.9592; (4) 0.8,0,0.6; (5) -0.8,0,0.6.

B. Numerical examples

Propagation characteristics for various values of the ratio r (with $R_0L=2\pi$) are illustrated in Figs. 3–9 for a pump beam with an initial polarization state $s_{10}=0$, $s_{20}=0.28$, and $s_{30}=0.96$ corresponding to a right-hand-elliptically polarized wave having $b/a=0.75$ and an orientation $\vartheta=+\pi/4$ (the variation of the Jacobian parameter

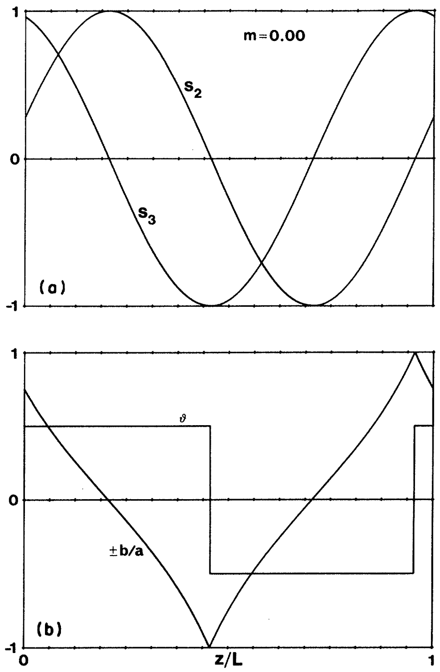

FIG. 3. Stokes parameters and the polarization ellipse descriptors as functions of z for the initial polarization state $s_{10}=0$, $s_{20}=0.28$, and $s_{30}=0.96$. The dependence of $m(r)$ for this initial state is given by curve 2 in Fig. 2. In this figure, $r=0$ (dc Kerr effect only) and $R_0L=2\pi$.

m with the ratio r for this initial state is given as curve 2 in Fig. 2). Each figure shows the variation of the Stokes parameters with the dimensionless distance z/L, as well as the variation of the orientation angle of the polarization ellipse (more correctly, the value shown is the normalized angle $2\vartheta/\pi$) and the signed quantity $\pm b/a = (b/a)\text{sgn}(s_3)$ describing both the shape and handedness

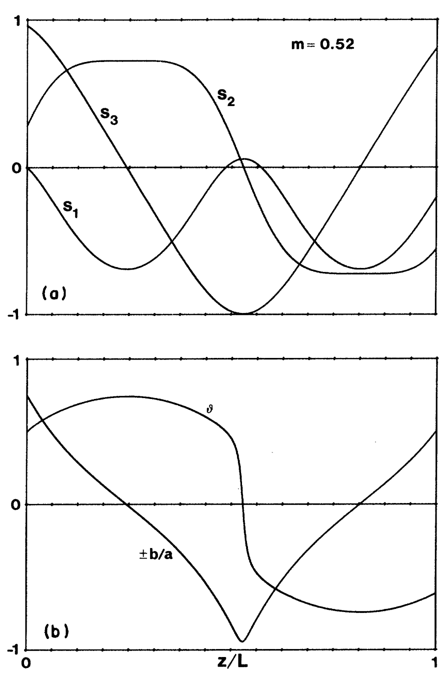

FIG. 4. The same as Fig. 3 but with $r=1.5$ and $R_p=1.41$.

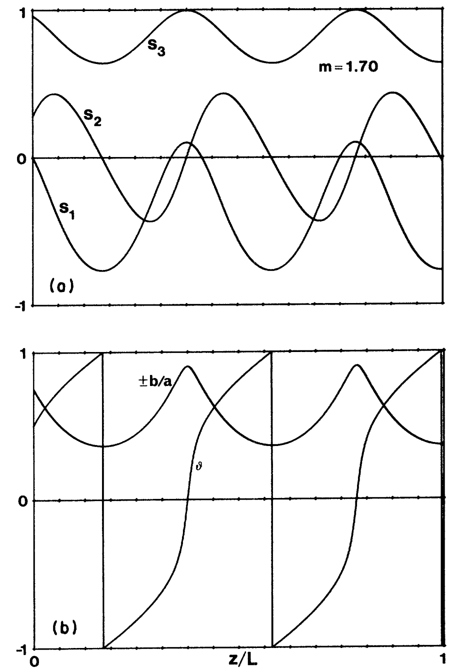

FIG. 5. The same as Fig. 3 but with $r=3$ and $R_p=0.41$.

of the polarization ellipse. All solutions were calculated from the set of coupled first-order differential equations (33) using a Hamming modified predictor-corrector algorithm and then independently verified in all cases by the direct calculation of the Jacobian elliptic functions using the method of the arithmetic-geometric mean.[41,42] With the exception of the special case where $m=1$ [for $r=2(1+s_{10})/s_{30}^2$ when $s_{30}\neq 0$ or for $r>1$ when $s_{10}=-1$], all of the solutions are periodic and may be conveniently divided into two classes according to whether $m<1$ or $m>1$.

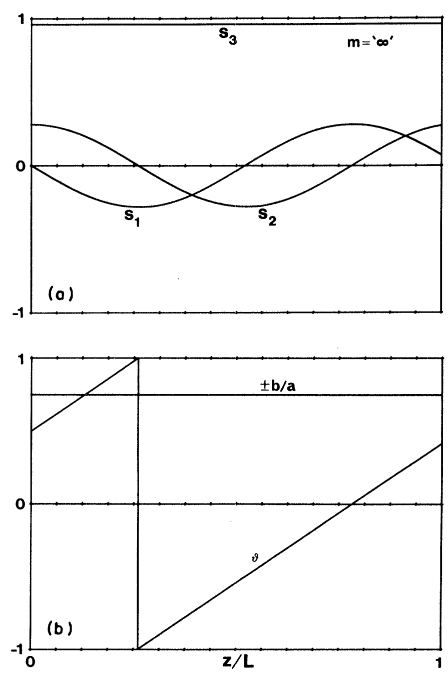

FIG. 6. The same as Fig. 3 but with $r=\infty$ (SIER only) and $R_1L=2\pi$.

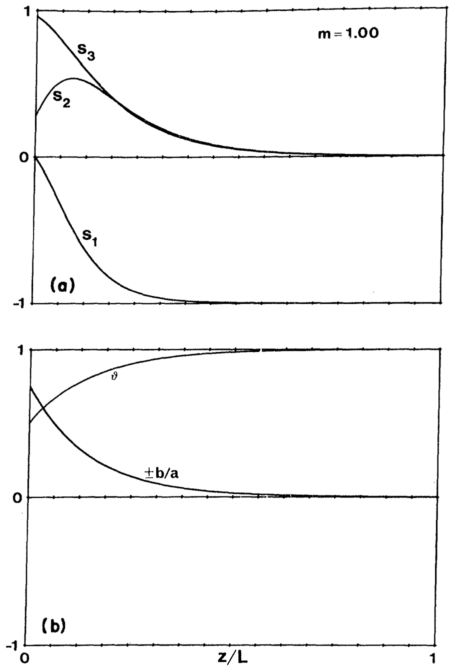

FIG. 7. The same as Fig. 3 but with $r=2(1+s_{10})/s_{30}^2$ and $m=1$.

Those solutions having $0 \le m < 1$ may be qualitatively interpreted as the "dc Kerr effect dominated" solutions in that, in addition to a varying shape and orientation for the polarization ellipse, the handedness will also vary between right- and left-handed states. Furthermore, values of $m<1$ are generally ($s_{30} \sim 0$ is the exception) the result of relatively small values of r. Figure 3 illustrates the case of the dc Kerr effect only, $r=m=0$, which is described by Eqs. (56). A "period cell length" $L(0)$ for the dc Kerr effect only is given by $L(0)=2\pi/R_0$, i.e., a retardation of

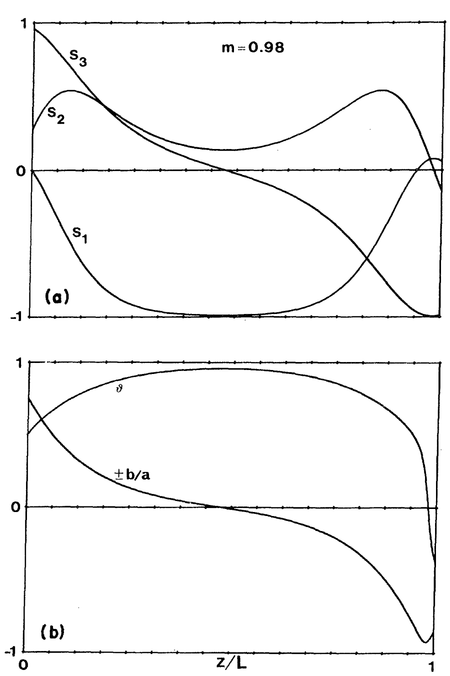

FIG. 8. The same as Fig. 3 but with $r=2.15$ and $R_p=2.04$.

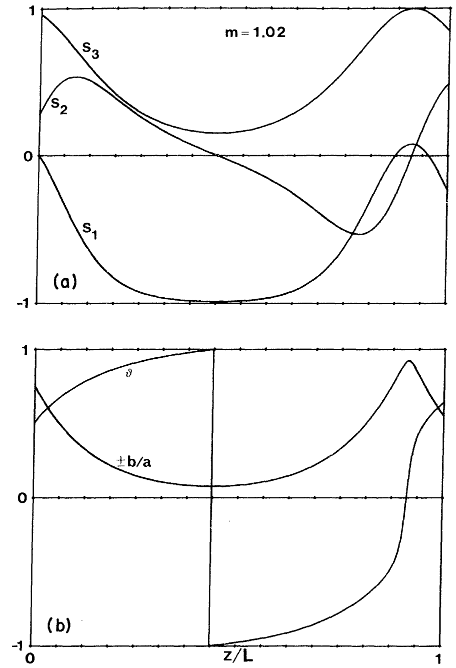

FIG. 9. The same as Fig. 3 but with $r=2.20$ and $R_p=0.95$.

one wavelength. When $0 \le m < 1$, then $L(m)=4K(m)/(fR_0)$ so that the ratio of these period cell lengths is

$$R_p(m)=L(m)/L(0)=2K(m)/\pi f, \quad 0 \le m < 1 \, . \tag{58}$$

Generally, R_p is >1 when $m<1$. Figure 4 illustrates the propagation characteristics in the case where $r=1.5$ for which $m=0.52$ and $R_p=1.14$.

The solutions for which $m>1$ may be qualitatively referred to as the "SIER dominated" solutions in that the handedness is constant when $m>1$. An example is illustrated in Fig. 5 for $r=3$ with $m=1.70$ and $R_p=0.41$. The expression for $R_p(m)$ when $m>1$ becomes

$$R_p(m)=L(m)/L(0)=K(1/m)/\pi kf, \quad m>1 \, . \tag{59}$$

In the limit of SIER only, as described by Eqs. (55), the shape as well as the handedness are constants and the orientation angle varies linearly with distance as illustrated in Fig. 6. In general, $R_p<1$ when $m>1$ and note, as Eqs. (54) indicate, that R_p is effectively halved when $m>1$ (the cn function with $m>1$ transforms[39,41] to the dn function with a parameter <1).

The special case of $m=1$ is of particular interest in that the polarization state tends asymptotically to a final state rather than varying periodically as in the cases where $m \ne 1$. Figure 7 depicts this aperiodic case $m=1$ for the same initial polarization state as the previous figures. The final state in this example corresponds to an optical beam linearly polarized and orthogonal to the dc electric field.

The solutions having $m \sim 1$ are also of particular interest since very small changes in the value of r result in pronounced differences in the propagation features. Figures 8 and 9 illustrate the solutions for values of $r=2.15$ and 2.20, respectively, with corresponding parameter values of $m=0.98$ and 1.02. The solution "switches" from a dc Kerr effect dominated one with $R_p=2.04$ to a

SIER dominated solution with $R_p=0.95$ for a change in the pump intensity (taking the dc field constant) of only 2.3%. Note that, in both cases, the polarization state is approximately constant over a substantial fraction of the cell length.

All of the examples given thus far have shown the solutions as functions of the distance z. It is also important to consider the solutions as functions of r, i.e., to examine the output polarization state at $z=L$ as the beam intensity is varied. This dependence is considerably more complicated since, in Eqs. (45)–(48), the argument and parameter of the Jacobian elliptic functions as well as the multiplying factors in the solutions all depend upon the ratio r. To illustrate, Fig. 10 plots the Stokes parameters and the polarization ellipse descriptors as functions of r for the flip-flop case [Eq. (44)] of $s_{10}=-0.8$, $s_{20}=0$, and $s_{30}=0.6$. The corresponding variation of $m(r)$ for this initial state is shown by curve 5 in Fig. 2. Note in particular the rapid changes which occur in the region $2(1+s_{10})/s_{30}^2<r<-1/s_{10}$.

IV. PROBE-BEAM PROPAGATION AND dc-INDUCED EFFECTS

A. Propagation of the probe beam

As Eqs. (25) indicate, the general solution for the propagation characteristics of the probe beam is considerably more complex than that for the pump beam, even under the assumption of a strictly real nonlinear susceptibility, and will not be treated in the present work. However, the effect on the probe due to the intense pump wave in the

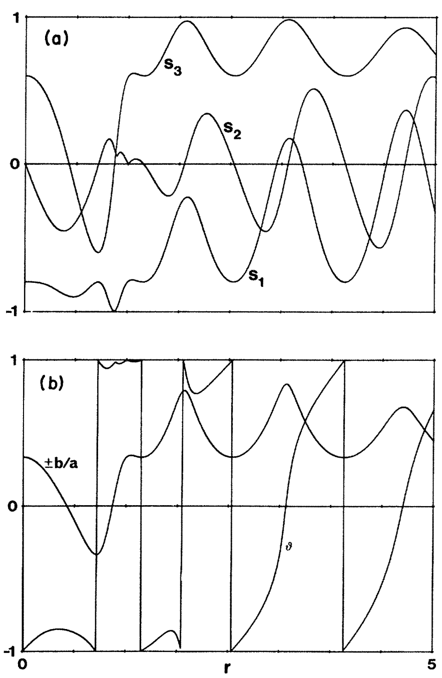

FIG. 10. Stokes parameters and the polarization ellipse descriptors as functions of r for the initial polarization state $s_{10}=-0.8$, $s_{20}=0$, and $s_{30}=0.6$ (right-hand elliptical with $b/a=\frac{1}{3}$ and $\vartheta=\pi/2$). The dependence of $m(r)$ for this initial state is given by curve 5 in Fig. 2.

absence of a dc field is included within Eqs. (25) and is here briefly considered.

Taking the nonlinear susceptibility to be strictly real (thus $s_0'=1$) and $E_0=0$, the Eqs. (25) describing the Stokes parameters of the probe beam simplify to

$$\begin{aligned}
\frac{d}{dz}s_1' &= -R_+' s_2 s_3' - R_-' s_3 s_2' , \\
\frac{d}{dz}s_2' &= R_+' s_1 s_3' + R_-' s_3 s_1' , \\
\frac{d}{dz}s_3' &= -R_+' (s_1 s_2' - s_2 s_1') ,
\end{aligned} \tag{60}$$

where $R_+'=\frac{1}{2}(R_1'+R_3')$, $R_-'=\frac{1}{2}(R_1'-R_3')$, and s_1, s_2, and s_3 are the SIER solutions given by Eqs. (55). By introducing the new variables

$$\begin{aligned}
w_1' &= 2a_r' a_l' \cos(\delta'-\delta) = s_1'\cos\delta + s_2'\sin\delta , \\
w_2' &= 2a_r' a_l' \sin(\delta'-\delta) = s_2'\cos\delta - s_1'\sin\delta
\end{aligned} \tag{61}$$

which are the "phase-relative Stokes parameters" analogous to s_1' and s_2', Eqs. (60) may be reexpressed in the form

$$\begin{aligned}
\frac{d}{dz}w_1' &= -\left[R_-' s_{30} - \frac{d\delta}{dz}\right] w_2' , \\
\frac{d}{dz}w_2' &= 2R_+' a_{r0} a_{l0} s_3' + \left[R_-' s_{30} - \frac{d\delta}{dz}\right] w_1' , \\
\frac{d}{dz}s_3' &= -2R_+' a_{r0} a_{l0} w_2' .
\end{aligned} \tag{62}$$

The shape and handedness of the probe polarization ellipse will be constants (i.e., polarization rotation only as with SIER) if and only if $ds_3'/dz=0$ for all values of z. This will be the case if the pump beam is purely circularly polarized with either a_{r0} or $a_{l0}=0$ (if the pump is linearly polarized then s_3' is constant only if the probe is also linearly polarized and either parallel or perpendicular to the pump). However, for a pump beam which is strictly elliptically polarized, the probe will, in general, experience change in the shape and handedness as well as the orientation of its polarization ellipse, a consequence of the fact that an elliptically polarized pump induces elliptical as opposed to circular birefringence. However, there exists two eigenstates for the initial polarization of the probe[8] for which it will propagate parallel to the pump and, like the pump, experience pure ellipse rotation. These two eigenstates are given by the relations

$$\begin{aligned}
w_2' &= 0 , \\
2R_+' a_{r0} a_{l0} s_{30}' &= (R_1 - R_-') s_{30} w_{10}' .
\end{aligned} \tag{63}$$

Note that although the pump induces elliptical birefringence, the nature of this birefringence is very different from the normal situation of a passive, linear elliptical retarder. In the case of SIER, the two elliptical eigenpolarizations are not fixed but rather rotate uniformly (Fig. 6) throughout the cell length. This is formally equivalent to a passive elliptical retarder which is physically twisted about its optic axis by an amount $d\vartheta/dz$ per unit length.

B. dc-induced second-harmonic generation

Denoting the integral of the functions F_i of Eqs. (29) as

$$f_i(z)=\int_0^z F_i(z)dz,\quad i=1\text{--}4 \tag{64}$$

then the intensity of the second-harmonic wave is given as

$$S_0^2=A_r^2(2\omega,z)+A_i^2(2\omega,z)=f_1^2+f_2^2+f_3^2+f_4^2\ . \tag{65}$$

Thus obtaining appreciable DCISHG requires that at least one of the terms f_i be large. However, since the value of Δk will be considerable in most isotropic media, e.g., $\Delta k\sim-5120\ \text{cm}^{-1}$ in liquid CS_2 for a pump wavelength of 1060 nm, all of the phase terms within the F_i will be rapidly varying so that the integrals f_i will be ~ 0 unless one or more of these phase terms can be made constant. Using the expressions for $d\delta/dz$ and $d\sigma/dz$ given by Eqs. (21c) and (21d), one finds that it is not possible to make either of the phases $\sigma+\delta+\Delta kz$ or $\sigma-\delta+\Delta kz$ constant except in the trivial limit of $r=0$ (for which the coefficients V_1 and V_2 vanish). However, it is possible to make $d(\sigma+\Delta kz)/dz$ vanish by satisfying the conditions

$$\begin{aligned}-2\Delta k&=2R_0+R_1+4R_3+2R_4\ ,\\ 2s_{10}&=r(s_{30}^2-1)\ .\end{aligned} \tag{66}$$

Although the DCISHG will be a maximum when these two conditions are met, in practice they are disadvantageous since they can be satisfied only for one particular optical intensity. Ensuring that $d(\sigma+\Delta kz)/dz=0$ is simply the condition for achieving phase-matched DCISHG, in this case principally by using a strong dc electric field to largely cancel the dispersion Δk and by selecting the polarization to exploit the combination of dc- and optically induced birefringence in the medium. This is not, however, the only possible approach to realizing efficient DCISHG. Alternate approaches include that based upon phase matching due to anomalous dispersion and/or suitable mixtures of different gases[44,45] as well as the concept of "periodic phase matching" using a sequence of dc electrodes having alternate polarities.[27,28]

C. dc-induced optical rectification

Assuming an incident linearly polarized pump beam which has the solution given by Eqs. (57), and denoting $a_x=\cos\phi$ and $a_y=\sin\phi$, then the induced dc voltage given by Eq. (32) takes the form, using $s_{10}=2\cos^2\phi-1$ and $s_{20}=\sin(2\phi)$,

$$V_{dc}(\phi)=6LE_0A_0^2g\left[\chi_3^{1122}+(\chi_3^{1111}-\chi_3^{1122})\cos^2\phi+(\chi_3^{1111}-\chi_3^{1122})\frac{r\sin^2(2\phi)}{4fR_0Lf^2(1-m)}\times\left[\int_0^{fR_0L}\text{cn}^2(x;m)dx-\text{sn}(fR_0L;m)\text{cd}(fR_0L;m)\right]\right]\ . \tag{67}$$

Since the last term on the right-hand side of this equation vanishes as r approaches 0, the first two terms constitute the zero-order approximation by effectively treating s_{10} as constant throughout the cell [i.e., the dc Kerr effect only solution given by Eqs. (56)]. Two important parameters which were recently measured by Ward and Guha[20] are

$$\begin{aligned}V_{dc}(0)-V_{dc}(\pi/2)&=6LE_0A_0^2g(\chi_3^{1111}-\chi_3^{1122})\ ,\\ V_{dc}(0)/V_{dc}(\pi/2)&=\chi_3^{1111}/\chi_3^{1122}\ .\end{aligned} \tag{68}$$

Most significantly, these values are independent of the value of m. However, in general, measurements of $V_{dc}(\phi)$ for angles other than 0 or $\pi/2$ must be interpreted with the aid of Eq. (67) whenever m is non-negligible. For the results reported by Ward and Guha,[20] particularly since the liquid used was nitrobenzene with its extremely large dc Kerr coefficient, the value of r was almost certainly ~ 0 and their assumption of the zero-order approximation accordingly correct. If r is small (and hence m is small since $s_{30}=0$), then the last term in Eq. (67) may be expanded in ascending orders of r with the first-order approximation taking the form

$$V_{dc}(\phi)\simeq 6LE_0A_0^2g\left[\chi_3^{1122}+(\chi_3^{1111}-\chi_3^{1122})\cos^2\phi+\tfrac{1}{8}(\chi_3^{1111}-\chi_3^{1122})r\sin^2(2\phi)\times\left[1-\frac{\sin(2R_0L)}{2R_0L}\right]\right]\ . \tag{69}$$

V. DISCUSSION

In defining the ratio r in Eq. (42), it was stated that r could be assumed ≥ 0 without loss of generality. However, the reader familiar with the dc Kerr effect will be aware that, in some media, the dc Kerr coefficient and hence the denominator in Eq. (42) can be negative. This would appear to demand that negative values of r be considered as well. A careful examination of the solutions described by Eqs. (45)–(50), however, reveals that the only odd power of r which occurs is the first, and only then either as rs_{10} or a simple factor for each of the s_i. Thus a negative value of r yields exactly the same solution found by using $|r|$ and negating the functions s_1, s_2, and s_3 (including their initial values) so that, as stated, r may be taken as positive with no loss of generality.

The principal thrust of this paper has been the formulation and the presentation of an exact, closed-form solution describing the propagation characteristics of an intense, arbitrarily polarized optical beam within a lossless, isotropic medium subjected to a dc electric field. The nature of the propagation, as illustrated in Figs. 3–10, is highly diverse in character, exhibiting a mixture of those properties associated with the dc Kerr effect on one hand and with the phenomenon of SIER on the other. The precise form of the solution depends upon both the ratio r defined in Eq. (42) as well as upon the initial state of polarization. This general solution is the prerequisite for exact studies of several related nonlinear optical effects and, in particular, the phenomena of dc-induced optical rectification and second-harmonic generation as well as the nature of the propagation of a weak probe beam have been described in light of this solution, albeit in somewhat less detail than that accorded to describing the propagation of the pump beam. It should be noted and emphasized that the nature of the pump propagation problem considered here and hence of its solution has immediate applicability to other, very similar propagation problems with a principal example being that of pulse transmission in an optical fiber exhibiting, e.g., strain-induced birefringence.

A basic and important feature of the analysis and resulting solutions given in this paper has been the use of the Stokes parameters as the fundamental variables rather than, as is more customary, the complex electric field amplitudes. This approach in turn has led directly to the concept of the Chebyshev-like elliptic functions con and son defined succinctly in Sec. III in terms of which the solutions are expressible in a compact and logically ordered manner as particular members of this eigenfunction sequence. While it cannot be expected that all other nonlinear optical phenomena will so readily admit exact solutions merely by reformulating in terms of the Stokes parameters (or some other, equally suitable set of real, direct observables), this approach is attractive in that, whenever intractability forces the necessity for some degree of approximation, such approximations are applied directly to final, real observables and not to inherently unobservable electric field quantities from which one must then derive the desired set of measurable parameters by quadrature and/or algebraic manipulation. Furthermore, as mentioned briefly in Sec. III, there is some reason to suggest that it may be possible, for certain classes of nonlinear problems which appear unamenable to exact solution, to arrive at accurate and compact analytic approximations through the combination of formulating the problem entirely in terms of some set of direct observables such as the Stokes parameters and the use of the generalized Jacobian functions of Eq. (53) as an eigenfunction basis in terms of which the real solution is expressed either as a generalized Fourier series or transform.

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