Light-induced nonreciprocity, field invariants, and nonlinear eigenpolarizations

A. E. Kaplan

School of Electrical Engineering, Purdue University, West Lafayette, Indiana 47907

Received June 30, 1983

It is shown that the magnitude of nonlinear nonreciprocity, self-induced by counterpropagating laser beams, is related to the anisotropic properties of the nonlinear third-order susceptibility (resulting in the light-induced birefringence and gyrotropy) and therefore is sensitive to the mutual arrangement of polarization of both of the beams. The invariants of nonlinear wave equations suggest an existence of mutual eigenarrangements of polarization with different relevant eigennonreciprocities. By using this effect, a nonreciprocal spectroscopy of the anisotropic nonlinear susceptibility may be developed.

Nonreciprocity of wave propagation (which results, e.g., in a different speed of forward- and backward-propagating light beams) is related to the fundamental issues of the electrodynamics of continuous media. In linear electrodynamics, nonreciprocity occurs in rotating or moving media (the so-called Sagnac and Fizeau effects,[1] respectively; the Sagnac effect is currently used for such applications as optical rotation sensors and laser gyroscopes[2]) or in gyrotropic materials. It was suggested recently[3,4] that counterpropagating laser beams in a Kerr nonlinear (nongyrotropic) medium can induce intensity-dependent nonreciprocity, which has been shown to be due to formation of a nonlinear index grating. This is a manifestation of the known fact that nonlinear components of susceptibility are different for each of the beams.[5–7] Under some conditions, this effect can result in a dramatic enhancement[3] of the Sagnac effect (in a rotating ring resonator) as well as in a directional bistability.[4] Subsequently, the nonlinear nonreciprocity was experimentally observed,[8,9] and different ways to eliminate its influence in the existing laser gyroscopes were discussed.[9–11]

In this Letter, we demonstrate for the first time to our knowledge that nonlinear nonreciprocity (for some specific mutual arrangements of polarization of the beams) becomes sensitive to the anisotropy of nonlinear susceptibility. This anisotropy is strongly affected by the mechanism of nonlinearity. In materials having third-order nonlinearity because of the Kerr effect, a nonlinear susceptibility is anisotropic, which provides for the light-induced birefrinence and gyrotropy.[12–15] In such a case, the nonlinear susceptibility is a tensor. On the other hand, materials with non-Kerr nonlinear mechanisms (e.g., electrostriction[13–15]) may demonstrate isotropic behavior. In the general case in which nonlinearity is due to the combination of two or more mechanisms, the anisotropic properties of the nonlinear susceptibility may vary substantially.

In this Letter, we show that the invariants of nonlinear wave equations suggest an existence of four mutual eigenarrangements of polarization of both of the beams. For each of these eigenarrangements, the type of polarization of each of the beams remains unchanged as the beams propagate through the nonlinear medium. Each of these four eigenarrangements has a different relevant eigennonreciprocity. This is caused solely by the anisotropy of nonlinear susceptibility and therefore by the physical mechanism of nonlinearity. For some configurations, the nonlinear nonreciprocity can even change sign. The set of four independent measurements of eigenvalues of nonlinear nonreciprocity (for four different mutual eigenarrangements of polarization) can provide substantial direct information about components of the tensor of nonlinear susceptibility. Therefore this effect can be used as a novel spectroscopic tool for measuring components of the nonlinear tensor. It is obvious also that the results obtained have important implications for optical gyroscope applications and for degenerate four-wave mixing.

We consider two optical beams with the same frequency, which are counterpropagating along axis z, such that the total (complex) electric field is

$$\mathbf{E} = [\mathbf{E}_1 \exp(ikz) + \mathbf{E}_2 \exp(-ikz)] \exp(-i\omega t).$$

[The total real field is $(\mathbf{E}^* + \mathbf{E})/2$, where an asterisk denotes complex conjugation.] Here $\mathbf{E}_1$ and $\mathbf{E}_2$ are envelope vectors of forward and backward waves, respectively; both of them are complex, slowly varying with respect to $\exp(ikz)$, and arbitrary polarized. Based on the results of Ref. 12, it could be shown that, in a general case of an arbitrary type of third-order nonlinear susceptibility, the nonlinear (complex) component of electric displacement $\mathbf{D}^{NL}$ can be expressed as

$$\mathbf{D}^{NL} = \epsilon_0\chi[A\mathbf{E}(\mathbf{E}\cdot\mathbf{E}^*) + B\mathbf{E}^*(\mathbf{E}\cdot\mathbf{E})], \qquad (1)$$

where ϵ_0 is the linear electric permittivity of the medium, χ is a constant of nonlinear interaction for a single linearly polarized plane wave, and A and B are dimensionless constants characteristic of the particular mechanism of nonlinearity. In a lossless medium, A and B are real quantities, and $A + B = 1$. In the case of the pure Kerr effect, in liquids[6,13] (e.g., CS_2) $B/A = 3$, i.e., $A = 1/4$, $B = 3/4$; in crystals[12] $B/A = 1/2$, i.e., A

0146-9592/83/110560-03$1.00

= 2/3, $B = 1/3$; and for electrostriction[13–15] $A = 1, B = 0$. Therefore the coefficient B may vary significantly. If $B \neq 0$, Eq. (1) suggests an anisotropy of nonlinear susceptibility χ^{NL} (which results in a light-induced birefringence[12–15]). Equation (1) could then be expressed in the form of a tensor relationship, $D_i^{NL} = \epsilon_0 \chi_{ik}^{NL} E_k$, where χ_{ik}^{NL} is a symmetrical tensor:

$$\chi_{ik}^{NL} = \chi[B(E_i E_k^* + E_i^* E_k) + (A - B) I_\Sigma \delta_{ik}]. \quad (2)$$

Here $\delta_{ik} = 1$ for $i = k$ and $\delta_{ik} = 0$ for $i \neq k$, and $I_\Sigma = |E_x|^2 + |E_y|^2 + |E_z|^2$. One can see that constant B is responsible for the induced anisotropy. Now, substituting Eq. (1) into the Maxwell equations, and taking into consideration that the field $\mathbf{E}$ consists of two counterpropagating waves, one can get wave equations for both of these waves. In doing that, we assume a conventional approximation of slowly varying envelopes as well as neglect the generation of highest-order harmonics, i.e., drop higher-order terms such as $\exp(\pm 3i\omega t \pm 3ikz)$ altogether. This is conventional in nonlinear index-related phenomena, such as self-focusing,[16] self-induced birefringence and gyrotropy,[12–15] optical bistability,[17] switching at nonlinear interfaces,[18] and four-wave mixing.[19] Finally, one gets the equations for envelope vectors $\mathbf{E}_j$ ($j = 1, 2$) of both of the counterpropagating waves:

$$2i(-1)^j \frac{d\mathbf{E}_j}{dz} + \frac{k}{\epsilon_o} \mathbf{D}_j^{NL}[\mathbf{E}_1(z); \mathbf{E}_2(z)] = 0; \quad j = 1, 2. \quad (3)$$

Here

$$\mathbf{D}_j^{NL} = \langle \mathbf{D}^{NL}(z) \exp[ikz(-1)^j] \rangle, \quad (4)$$

where angle brackets denote averaging over the distance $2\pi/k$, i.e.,

$$\mathbf{D}_j^{NL} = \epsilon_0 \chi A \{\mathbf{E}_j[(\mathbf{E}_1 \cdot \mathbf{E}_1^*) + (\mathbf{E}_2 \cdot \mathbf{E}_2^*)] + \mathbf{E}_{3-j}(\mathbf{E}_j \cdot \mathbf{E}_{3-j}^*)\} + \epsilon_0 \chi B[2\mathbf{E}_{3-j}^*(\mathbf{E}_1 \cdot \mathbf{E}_2) + \mathbf{E}_j^*(\mathbf{E}_j \cdot \mathbf{E}_j)]; \quad j = 1, 2. \quad (5)$$

The solution of Eqs. (3) must satisfy boundary conditions at both of the faces of a nonlinear layer of the thickness L: $\mathbf{E}_1(z = 0) = \mathbf{E}_{1_0}$; $\mathbf{E}_2(z = L) = \mathbf{E}_{2L}$. In the case $\mathbf{E}_2 = 0$ (or $\mathbf{E}_1 = 0$), Eqs. (3) and (5) determine one-directional self-interaction of a plane wave, in particular, light-induced gyrotropy, i.e., self-rotating of the main axis of elliptic polarization if the incident wave is slightly elliptically polarized.[12,15] In the case when both of these vectors $\mathbf{E}_1$ and $\mathbf{E}_2$ are linearly polarized and parallel to each other, Eqs. (3) and (5) provide for nonlinear nonreciprocity[3,4] with $\chi_1^{NL} - \chi_2^{NL} = \chi(I_2 - I_1)$, where $I_j = |E_j|^2$ (i.e., I_1 and I_2 are the respective intensities of both of the waves). In the case of a lossless medium (χ, A, and B are real; $A + B = 1$), our analysis of Eqs. (3) shows that they have at least four invariants:

$$[\mathbf{E}_1(z) \cdot \mathbf{E}_1^*(z)] = \text{inv} = I_1,$$
$$[\mathbf{E}_2(z) \cdot \mathbf{E}_2^*(z)] = \text{inv} = I_2, \quad (6)$$

$$(1 - 2B)|(\mathbf{E}_1 \cdot \mathbf{E}_2^*)|^2 + 3B|(\mathbf{E}_1 \cdot \mathbf{E}_2)|^2 = \text{inv}, \quad (7)$$

$$2(1 + B)|(\mathbf{E}_1 \cdot \mathbf{E}_2^*)|^2 + 3B[|(\mathbf{E}_1 \cdot \mathbf{E}_1)|^2 + |(\mathbf{E}_2 \cdot \mathbf{E}_2)|^2] = \text{inv}. \quad (8)$$

Invariants (6) describe a separate conservation of an energy flow in each of the beams (and, therefore, the absence of energy exchange between them), whereas invariants (7) and (8) are apparently related to the conservation of a momentum flux of the entire field.[6]

For arbitrary intensities and types of polarization of the beams, Eqs. (3) are far from being readily solvable. Both of the beams, if sufficiently strong, affect each other, causing mutually induced rotation of axes of polarization (which is of the same nature as induced gyrotropy[12]), transformation of linear polarization into an elliptical one and vice versa, etc. Under condition $\chi\sqrt{I_1 I_2} Lk \gtrsim 1$, the system may exhibit bistability and multistability of exiting polarization, spatial instabilities, and periodic (and even chaotic) spatial distribution of polarization. However, such does not need to be the case where the observation of the nonlinear nonreciprocity for different polarizations is concerned. Indeed, examination of invariants (6)–(8) along with Eq. (3) reveals that, for TE waves ($E_{zj} = 0$), there are at least four types of incident polarization arrangements for which the polarization of both of the waves remains unchanged during propagation as long as these arrangements exist at least at one spatial point. These arrangements are as follows: Both of the beams are linearly polarized [i.e., $|(\mathbf{E}_j \cdot \mathbf{E}_j)| = I_j$ with vectors $\mathbf{E}_1$ and $\mathbf{E}_2$ either parallel to each other[3] [i.e., $|(\mathbf{E}_1 \cdot \mathbf{E}_2)|^2 = I_1 I_2$], or orthogonal to each other [i.e., $(\mathbf{E}_1 \cdot \mathbf{E}_2) = 0$]; both of the beams are circularly polarized [i.e., $(\mathbf{E}_j \cdot \mathbf{E}_j) = 0$] with either corotating [i.e., $(\mathbf{E}_1 \cdot \mathbf{E}_2) = 0$] or counterrotating [i.e., $(\mathbf{E}_1 \cdot \mathbf{E}_2^*) = 0$] directions of circular polarization.

All these arrangements may be regarded as mutual eigenarrangements of the polarization of the counterpropagating beams for any tensor of a third-order nonlinear susceptibility. For all these eigenarrangements, the vector of nonlinear partial displacement, Eq. (5), can be expressed as $\mathbf{D}_j^{NL} = \epsilon_0 \chi_j^{NL} \mathbf{E}_j$ ($j = 1, 2$), where χ_j^{NL} are different scalars pertinent to the respective eigenarrangements. Making use of Eq. (5), one can see that in these cases the partial nonlinear susceptibilities χ_j^{NL} relevant to the respective waves $\mathbf{E}_j$ are

$$\chi_j^{NL} = \chi[\alpha I_j + (\alpha + \beta) I_{3-j}], \qquad j = 1, 2, \quad (9)$$

and thus the nonlinear nonreciprocity is

$$\chi^{NR} = \chi_1^{NL} - \chi_2^{NL} = \chi\beta(I_2 - I_1), \quad (10)$$

where $I_j = |\mathbf{E}_j|^2$ and

(1) $\alpha = 1$ and $\beta = 1$ if the beams are linearly polarized with $\mathbf{E}_1$ and $\mathbf{E}_2$ parallel to each other,

(2) $\alpha = 1$ and $\beta = -B$ if the beams are linearly polarized with $\mathbf{E}_1$ and $\mathbf{E}_2$ orthogonal to each other,

(3) $\alpha = (1 - B)$ and $\beta = (1 - B)$ if the beams are circularly polarized with $\mathbf{E}_1$ and $\mathbf{E}_2$ corotating, and

(4) $\alpha = (1 - B)$ and $\beta = 2B$ if the beams are circularly polarized with $\mathbf{E}_1$ and $\mathbf{E}_2$ counterrotating.

In the case of the pure Kerr effect in liquids, for instance, $B = 3/4$, the ratio of magnitudes β of nonlinear nonreciprocity for these respective eigenarrangements is 1:(−3/4):(1/4):(3/2), which therefore should be valid, e.g., for CS_2 and some other organic liquids.[13] For crystals ($B = 1/3$) the respective ratio is 1:(−1/3):(2/3):(2/3), whereas for electrostriction it is 1:0:1:0.

Clearly, these relationships indicate potentials for novel, nonreciprocal spectroscopy of nonlinear mechanisms in different materials. Independent measurements of nonlinear nonreciprocity, Eq. (10), with fixed intensities I_1 and I_2 for four different eigenarrangements of polarization provide four numbers, which allow one not only to calculate nonlinear constant χ and nonlinear anisotropical parameter B but also to test the validity of the general model related to Eq. (1). The new measurements of parameter B may justify the dominance of one or another nonlinear mechanism in different materials (such as liquid CS_2; liquid crystals; semiconductors, e.g., InSb; and metallic vapors, e.g., Na) by measuring nonlinear nonreciprocity, which can be sufficiently large in the above-mentioned material to allow for exploiting the cw laser regime. It should be pointed out that measurement of nonlinear nonreciprocity requires much less power than the observation of such an effect as self-focusing[16] because the required nonreciprocal phase difference $\Delta\phi^{NR} = \chi^{NR} Lk$ could be less than 10^{-3}–10^{-4}. In the experiments,[8,9] the power difference $I_2 - I_1$ used to observe the effect in silicon fibers was of the order of a few microwatts.

One may note also that the new facts about nonlinear nonreciprocity must be taken into consideration in the theory of the nonlinear Sagnac effect,[3] directional bistability,[4] and the influence of the Kerr effect in the laser gyroscopes.[9–11] For instance, in order to eliminate nonlinear nonreciprocity in laser gyroscopes in the case of differently polarized counterpropagating beams by using a periodic sequence of light pulses,[9–11] one has to choose a temporal period between the pulses different from that of the case in which the beams are linearly polarized with their fields parallel to each other [see Eq. (9)]. In the latter case the required ratio[9] of the pulses' duration to the period between them is 1:1 regardless of the magnitude of B. It can be readily shown that in the general case this ratio is $\alpha:\beta$. Thus it remains the same (1:1) in the case of circularly polarized beams with the corotating fields, whereas it becomes $(1 - B):2B$ for circularly polarized counterrotating fields. For linearly polarized beams with orthogonal fields this method of elimination of nonlinear nonreciprocities apparently does not work at all if $B > 0$ and must be modified. The existence of the field invariants and mutual eigenarrangements of polarizations of beams also has important implications in the theory of degenerate collinear four-wave mixing,[19] indicating new opportunities for exact analysis of the pertinent equations and suggesting new polarization arrangements for experiment.

I appreciate useful discussion with R. A. Bergh concerning the magnitude of B for different materials. This research was supported by the U.S. Air Force Office of Scientific Research.

References

1. For a comprehensive review, see E. Post, Rev. Mod. Phys. **39,** 475 (1967); J. Anandan, Phys. Rev. D. **24,** 338 (1981).
2. See, e.g., "Laser inertial rotation sensors," Proc. Soc. Photo-Opt. Instrum. Eng. **157,** 1978; S. Esekiel and H. J. Arditty, eds. *Fiber-Optic Rotation Sensors and Related Technology* (Springer-Verlag, Berlin, 1982).
3. A. E. Kaplan and P. Meystre, Opt. Lett. **6,** 590 (1981).
4. A. E. Kaplan and P. Meystre, Opt. Commun. **40,** 229 (1982).
5. R. Y. Chiao, P. L. Kelley, and E. Garmire, Phys. Rev. Lett. **17,** 1158 (1966).
6. B. Ya. Zeldovich, Brief Commun. Physics Lebedev Inst. **5,** 20 (1970) (in Russian).
7. A. E. Kaplan, Opt. Lett. **6,** 361 (1981).
8. S. Ezekiel, J. L. Davis, and R. W. Hellwarth, Opt. Lett. **7,** 457 (1982).
9. R. A. Bergh, H. C. Lefevre, and H. J. Shaw, Opt. Lett. **7,** 282 (1982).
10. K. Peterman, Opt. Lett. **7,** 623 (1982).
11. N. J. Frigo, H. F. Taylor, L. Goldberg, J. Weller, and S. C. Rashleigh, Opt. Lett. **8,** 119 (1983).
12. P. D. Maker, R. W. Terhune, and C. M. Savage, Phys. Rev. Lett. **12,** 507 (1964).
13. Y. R. Shen, Phys. Lett. **20,** 378 (1966).
14. G. Mayer and F. Gires, Compt. Rend. **258,** 2039 (1964).
15. P. D. McWane and D. A. Sealer, Appl. Phys. Lett. **8,** 278 (1966).
16. For a review on self-focusing, see S. A. Akhmanov, R. V. Khokhlov, and A. P. Sukhorukov in *Laser Handbook*, F. T. Arecchi and E. O. Schulz-Due Bois, eds. (North-Holland, Amsterdam, 1972), p. 1150.
17. For the most recent review on optical bistability, see E. Abraham and D. Smith, Rep. Prog. Phys. **45,** 815 (1982).
18. A. E. Kaplan, JETP Lett. **24,** 114 (1976); A. E. Kaplan, Sov. Phys. JETP **72,** 896 (1977); P. W. Smith, J.-P. Hermann, W. J. Tomlinson, and P. J. Maloney, Appl. Phys. Lett. **35,** 846 (1979); W. J. Tomlinson, J. P. Gordon, P. W. Smith, and A. E. Kaplan, Appl. Opt. **21,** 2041 (1982).
19. J. H. Marburger and J. F. Lam, Appl. Phys. Lett. **34,** 389 (1978); **35,** 249 (1979).