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Tunable self-action of light in optical rectification

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Abstract

We analyze the self-action of light waves mediated by cascaded optical rectification in a quadratic nonlinear crystal in the framework of the full local-field equations and show that the process can lead to a rich variety of self-effects. We put forward a general scheme to calculate the full nonlinear response mediated by the self-generated rectified fields and thus show that acting on the shape, the polarization of the light beam and the geometric arrangement of the nonlinear crystal allows tuning the sign, the strength, and the type of the induced nonlinearities, opening the door to the exploration of a variety of self- and cross-phase modulations, and solitary-waves. We also show configurations where even though the macroscopic rectified field vanishes, the macroscopic self-effects do not.

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Elucidation of new physical settings where wave excitations act on themselves is one of the ultimate goals of nonlinear optics, hence the self-action of light mediated by quadratic and cubic nonlinearities is a subject of intense research. In this context, systems supporting solitons, or self-sustained energy packets, occupy a special position [1–3]. The types of possible solitary-wave structures depends on the variety of self-effects that can be generated with each type of nonlinear material response.

Such variety can be routed back to the detailed process that induces the nonlinear response and it manifests itself on the richness of the equations that describe the light evolution. Therefore, identification of new nonlinear mechanisms is of paramount importance. Here we address the nonlinear interaction of optical waves with static fields, a process which contains contributions from different origins whose effect on soliton formation has never been explored to date in its full potential.

Gustafson and co-workers [4] noticed long ago that the combination of optical rectification (OR) and the electro-optic effect can give rise to self-phase modulation. More recently, Bosshard et al. [5] showed that the effect gives rise to an effective Kerr-like nonlinear refractive index. However, in this work the macroscopic rectified field that arises

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because of the spatial modulation of the rectified polarization was not taken into account. The same assumption was made by Steblina and co-workers [6] to examine soliton formation in systems where an optical beam is coupled to a microwave signal through OR and the electro-optic effect. On the other hand, the propagation equations for optical fields coupled to macroscopic rectified fields were derived by Ablowitz et al. [7] using a multiple scale expansion approach. However, in such a work the contribution of the rectified nonlinear response to the rectified microscopic field was neglected. Nevertheless, as it was recognized in the early days of nonlinear optics, in such processes the actual macroscopic nonlinear response can depend strongly on the difference between the macroscopic fields and polarization and the microscopic fields, the field acting on each individual molecule (termed the *local field*) [8–11]. Evidence of such effects was revealed only very recently in the investigation of degenerate four-wave mixing [12–14]. The important point for our present purposes is that taking into full account both the rectified polarization and the macroscopic field it induces creates combinations of nonlinearities which can importantly impact the detailed mechanisms that lead to soliton formation.

In this paper we put forward the general scheme that can lead to the formation of solitary waves through the interaction of light signals with self-generated static fields in noncentrosymmetric crystals, by considering the full nonlinear response due to optical rectification and the electro-optic effect. We derive general evolution equations and illustrate their richness with specific examples. From an experimental viewpoint, the central result reported is that for a given illuminating wavelength, a rich variety of different types of self-effects are possible. We show that the strength of the different contributions can be controlled by acting on the polarization and shape of the input beam. Hence, a variety of solitary-wave structures, of different features and characteristics, can be potentially implemented using this mechanism, opening the door to new opportunities.

Consider the propagation of a light beam in noncentrosymmetric crystals with a large quadratic nonlinearity, and let the coordinate axes

(x, y, z) coincide with the crystal optical axes (a, b, c) . We assume that the electric field is perpendicular to the wave vector, so that the macroscopic electric field at frequency ω writes $\vec{\mathcal{E}}(\vec{r}, t) = 1/2\vec{A}(\vec{r})\exp(-i\omega t) + \text{c.c.}$, where the phasor \vec{A} is given by $\vec{A}(\vec{r}) = \hat{x}A_x(x, y, z)\exp(ik_xz) + \hat{y}A_y(x, y, z)\exp(ik_yz)$. Here $k_j = k_0n_j$ ($j = x, y$), where k_0 is the wavenumber in vacuum and n_j are the linear refractive indices. The evolution equation for the j th component of \vec{A} in the slowly varying approximation is

$$2ik_j \frac{\partial A_j}{\partial z} + \nabla_{\perp}^2 A_j + \frac{k_0^2}{\epsilon_0} P_j^{\text{NL}}(\omega) \exp(-ik_jz) = 0, \quad (1)$$

where ϵ_0 is the vacuum permittivity and $\vec{P}^{\text{NL}}(\omega)$ is the nonlinear polarization at frequency ω . The nonlinear polarization contains the three contributions

$$\vec{P}^{\text{NL}}(\omega) = \vec{P}^{(3)}(\omega) + \vec{P}^{(\text{OR})}(\omega) + \vec{P}^{(\text{SHG})}(\omega). \quad (2)$$

In a centrosymmetric material, the only contribution to $\vec{P}^{\text{NL}}(\omega)$ comes from third-order nonlinear optical effects [$\vec{P}^{(3)}(\omega)$]. In a noncentrosymmetric material, there is an additional “cascading” contribution arising from second-order nonlinear optical effects. These are the combination of optical rectification and the linear electro-optic effect [$\vec{P}^{(\text{OR})}(\omega)$], and second harmonic generation [$\vec{P}^{(\text{SHG})}(\omega)$] [15]. In a crystal configuration where frequency generation processes are largely phase-mismatched, the corresponding contributions to $\vec{P}^{\text{NL}}(\omega)$ are negligible. In what follows we consider only the contribution from optical rectification, because it is the one that yields combinations of nonlinearities, and thus a new way to control the properties of optical solitons.

The electric field $\vec{\mathcal{E}}$ induces a real macroscopic static nonlinear polarization given by

$$P_i^{(2)}(0) = \frac{\epsilon_0}{2} \sum_{p,q} \chi_{ipq}^{(2)}(0, -\omega, \omega) A_p^* A_q \exp(i\Delta k_{qp}z), \quad (3)$$

where $\chi_{ipq}^{(2)}(0, -\omega, \omega)$ is the second-order nonlinear susceptibility for optical rectification and $\Delta k_{ij} = k_i - k_j$. Such macroscopic nonlinear polarization induces a complex microscopic nonlinear polarization $p_i(\omega)$ due to the electro-optic effect given by

$$p_i(\omega) = 2\epsilon_0 \sum_{j,k} \alpha_{ijk}^{(2)}(-\omega, 0, \omega) \exp(ik_j z), \quad (4)$$

where $\alpha_{ijk}^{(2)}(-\omega, 0, \omega)$ is the second-order nonlinear molecular polarizability, e_i is the static local field, and a_i is the slowly varying part of the local field at frequency ω . The static local field e_i can be calculated using the Lorentz approximation to get [16],

$$e_i = \frac{\epsilon_{ii}(0) + 2}{3} E_i + \frac{P_i^{(2)}(0)}{3\epsilon_0}, \quad (5)$$

where E_i is the macroscopic static electric field induced by the macroscopic nonlinear polarization $\vec{P}_i^{(2)}(0)$, and $\epsilon_{ii}(0)$ are the diagonal elements of the dielectric relative permittivity tensor at zero frequency. The local field a_i and the macroscopic fields at frequency ω are related by the expression

$$a_i = \frac{\epsilon_{ii}(\omega) + 2}{3} A_i \quad (6)$$

with $\epsilon_{ii}(\omega)$ being the diagonal elements of the dielectric permittivity tensor at frequency ω . Taking into account the relationship between the macroscopic nonlinear polarization $P_i^{NL}(\omega)$ and the microscopic nonlinear polarization $p(\omega)$, and the relationship between $\alpha_{ijk}^{(2)}(-\omega, 0, \omega)$ and $\chi_{ijk}^{(2)}(-\omega, 0, \omega)$ [8–11], one finds that the sought-after macroscopic nonlinear polarization at frequency ω can be written as

$$P_i^{NL}(\omega) = \epsilon_0 \sum_j \mathcal{G}_{ij} A_j \exp(ik_j z), \quad (7)$$

where $\mathcal{G}_{ij} = \mathcal{G}_{ij}^{(3)} + \mathcal{G}_{ij}^{(OR)}(A_x, A_y)$. The intrinsic third-order contribution is given by

$$\mathcal{G}_{ij}^{(3)} = \frac{3}{4} \sum_{pq} \chi_{iqpj}^{(3)}(-\omega, -\omega, \omega, \omega) [A_q(\omega)]^* A_p(\omega). \quad (8)$$

The matrix function $\mathcal{G}_{ij}^{(OR)}(A_x, A_y)$, hereafter to be referred to as *nonlinear structure factor*, is given by the expression (see [11,13] for further details)

$$\begin{aligned} \mathcal{G}_{ij}^{(OR)} &= \frac{n_i^2 n_j^2}{4} \sum_{pqrs} \frac{n_p^2 n_q^2 r_{ips} r_{qjs}}{\epsilon_{ss}(0) + 2} A_p(\omega) [A_q(\omega)]^* \\ &\times \exp(i\Delta k_{pq} z) - n_i^2 n_j^2 \sum_p r_{ijp} E_p, \end{aligned} \quad (9)$$

where E_i is the i th component of the rectified macroscopic field, and $\epsilon_{ii}(0)$ is the diagonal element of the electric permittivity at zero frequency. To derive this expression, use has been made of the definition $\chi_{ijk}^{(2)}(-\omega, \omega, 0) = -1/2 n_i^2 n_j^2 r_{ijk}$, and we assumed that Kleimann symmetry holds. The macroscopic static electric field \vec{E} appearing in (9) has to fulfill the equation $\nabla \times \vec{E} = 0$, together with

$$\sum_i \epsilon_{ii}(0) \partial_i E_i = -\frac{1}{\epsilon_0} \sum_i \partial_i P_i^{(2)}(0), \quad (10)$$

where ∂_i stands for the derivative with respect to the i th coordinate. This equation comes from $\nabla \cdot \vec{D} = 0$, where \vec{D} is the electric displacement vector.

Throughout this paper we consider settings in which the scale of variation of the nonlinear polarization source over the longitudinal coordinate z is much larger than that over the transverse axes x, y . Then, the longitudinal component of the static electric field, as well as all its derivatives can be neglected [17]. Under such conditions we arrive at the general evolution equation for A_j , which can be written as

$$\begin{aligned} 2ik_j \frac{\partial A_j}{\partial z} + \nabla_{\perp}^2 A_j + k_0^2 \sum_{jm} \mathcal{G}_{jm}(A_x, A_y) A_m \exp(i\Delta k_{jm} z) \\ = 0, \end{aligned} \quad (11)$$

which is coupled to the equation for the two components of the rectified electric field

$$\sum_i \epsilon_{ii}(0) \partial_i^2 E_j = \partial_j \mathcal{H}(A_x, A_y), \quad (12)$$

where

$$\mathcal{H} = \frac{1}{4} \sum_{pqm} n_p^2 n_q^2 r_{pqm} \partial_m [A_p^* A_q \exp(i\Delta k_{qp} z)]. \quad (13)$$

Eqs. (11)–(13) are one of the central results of this paper. They reveal that the type and strength of the nonlinearity that rules the evolution of the light beam \vec{A} depends critically on the shape and polarization of the beam, as well as on the crystallographic structure and orientation of the material relative to the light propagation direction. Eq. (12) describes the material response, similarly to the Bloch equations that describe the noninstantaneous response of a nonlinear material [18],

or to the description of carrier dynamics in photorefractive media [19,20]. It can be formally solved in the spatial frequency domain, to obtain

$$E_j(x, y) = \mathcal{F}^{-1} \left\{ \frac{-ik_j \mathcal{F}[\mathcal{H}(A_x, A_y)]}{\epsilon_{xx}(0)k_x^2 + \epsilon_{yy}(0)k_y^2} \right\}. \quad (14)$$

Here \mathcal{F} and \mathcal{F}^{-1} stand for the direct and the inverse Fourier transforms, respectively, and (k_x, k_y) are the coordinates in the spatial frequency domain. The concrete form of (11)–(14) strongly depends on the crystal symmetry and light pumping conditions. Inspection of (14) shows that, by and large, the contribution of the macroscopic rectified field to the nonlinear response is to introduce non-local effects, in the sense that the value of the rectified field \vec{E} depends on the whole structure of the optical beam shape. Therefore, only with special crystal geometries, illuminating configurations, and soliton dimensionality it is possible to obtain the reduced evolution discussed in [5–7]. To illustrate this point and the different existing possibilities, in what follows we discuss a few concrete examples.

Example I. A linearly polarized beam that propagates in a material with point group symmetry $mm2$. Let the crystal be oriented relative to the direction of light propagation so that $\{x||c, y||a, z||b\}$, and $n_x \equiv n_3$, $n_y \equiv n_2$, $n_z \equiv n_1$, $\epsilon_{xx}(0) \equiv \epsilon_{33}$, $\epsilon_{yy}(0) \equiv \epsilon_{22}$ and $\epsilon_{zz}(0) \equiv \epsilon_{11}$. Under such conditions, (11) and (12) yield

$$2ik_3 \frac{\partial A_x}{\partial z} + \nabla_{\perp}^2 A_x + k_0^2 [(\gamma + \alpha)|A_x|^2 - \mu E_x] A_x = 0, \quad (15)$$

$$\epsilon_{33} \frac{\partial^2 E_x}{\partial x^2} + \epsilon_{11} \frac{\partial^2 E_x}{\partial y^2} = \frac{\mu}{4} \frac{\partial^2 |A_x|^2}{\partial x^2}, \quad (16)$$

where $\alpha = (n_3^8 r_{333}^2/4)/(\epsilon_{33} + 2)$, $\mu = n_3^4 r_{333}$, $k_3 = k_0 n_3$ and $\gamma = 3/4 \chi_{3333}^{(3)}(-\omega, -\omega, \omega, \omega)$. The first contribution to the nonlinear term in Eq. (15) corresponds to a self-focusing Kerr-like nonlinearity. Except for the factor $\epsilon_{33} + 2$, the contribution from second-order cascading corresponds to the contribution reported by Bosshard and co-workers [5]. The additional new contribution is proportional to the rectified electric field $E_x(x, y; z)$, and introduces non-local (i.e., in general E_x is not

simply proportional to $|A_x|^2$) nonlinear self-guiding effects. Their strength depends on the magnitude and spatial shape of the static electric field induced by the optical beam, and can be important as shown in Fig. 1. We plotted the scaled nonlinear contribution $g_{xx} = \mathcal{G}_{xx}^{(OR)}/|A_0|^2$ computed at the center of a beam with a transverse Gaussian shape (i.e., $A_x = A_0 \exp(-x^2/\omega_x^2 - y^2/\omega_y^2)$), as a function of the ellipticity of the beam $\zeta = \omega_x/\omega_y$. For $\zeta = 1$ and an isotropic crystal, one recovers the limit case analyzed in [21]. The important result uncovered by the plot is that one can tune the value of g_{xx} by varying the beam ellipticity.

The limit cases corresponding to highly elliptical beams can be solved analytically. When $\zeta \gg 1$, the intensity of the optical beam in the spatial frequency domain is negligible for $k_x \neq 0$, thus (16) yields a vanishing macroscopic static electric field (i.e., $E_x = 0$). Hence, $g_{xx} = (n_3^8 r_{333}^2/4)/(\epsilon_{33} + 2)$. This is always a positive value. When $\zeta \ll 1$, the intensity of the optical beam in the spatial frequency domain is negligible for $k_y \neq 0$, so from (16) one obtains $E_x = \mu |A_x|^2/(4\epsilon_{33})$, and $g_{xx} = -(2/\epsilon_{33})(n_3^8 r_{333}^2/4)/(\epsilon_{33} + 2)$. Notice that the negative value of g_{xx} is $2/\epsilon_{33}$ times smaller than the positive one, and that in most materials, at low frequencies $\epsilon_{33} \gg 1$. For example, in potassium

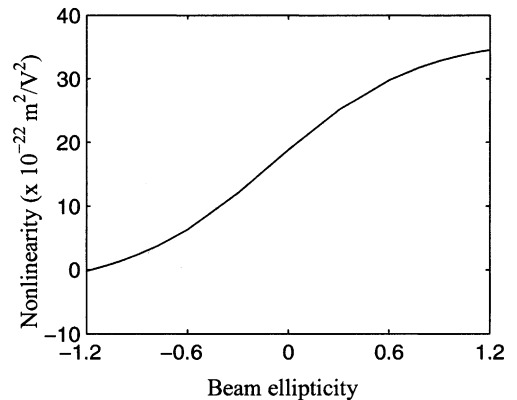


Fig. 1. Scaled nonlinear structure factor g_{xx} computed at the center of a beam with a Gaussian shape, as a function of the beam ellipticity ζ . The beam ellipticity is given in logarithmic scale. To be specific, the scaled nonlinear structure factor was calculated for the actual parameters of potassium niobate pumped at $\lambda \sim 1 \mu\text{m}$.

niobate (KNbO₃) at $\lambda \sim 1 \mu\text{m}$, one obtains $g_{xx} \simeq 36 \times 10^{-22} \text{ m}^2/\text{V}^2$ for $\zeta \gg 1$, whereas $g_{xx} \simeq -3 \times 10^{-22} \text{ m}^2/\text{V}^2$ for $\zeta \ll 1$. For comparison, the intrinsic third-order nonlinearity amounts to about $45 \times 10^{-22} \text{ m}^2/\text{V}^2$ [13].

Example II. Consider a beam that propagates along the optic axis of a crystal with point group symmetry $3m$. Let $\hat{z} = \hat{c}$, thus $n_x = n_y \equiv n_1$, $\epsilon_{xx}(0) = \epsilon_{yy}(0) \equiv \epsilon_{11}$, and $\epsilon_{zz}(0) \equiv \epsilon_{33}$. Substitution in (11) and (14) gives

$$2ik_1 \frac{\partial A_x}{\partial z} + \nabla_{\perp}^2 A_x + k_0^2 [a|A_x|^2 A_x + b|A_y|^2 A_x + cA_y^2 A_x^* + \mu(A_y E_x + A_x E_y)] = 0, \quad (17)$$

$$2ik_1 \frac{\partial A_y}{\partial z} + \nabla_{\perp}^2 A_y + k_0^2 [a|A_y|^2 A_y + b|A_x|^2 A_y + cA_x^2 A_y^* + \mu(A_x E_x + A_y E_y)] = 0. \quad (18)$$

Here $a = \alpha + v + \gamma$, $b = \alpha + 2\gamma/3$, $c = v + 2\gamma/3$, and $\mu = n_1^4 r_{222}$, where $\alpha = (n_1^8 r_{113}^2/4)/(\epsilon_{33} + 2)$, $v = (n_1^8 r_{222}^2/4)/(\epsilon_{11} + 2)$, and $\gamma = 3/4 \chi_{1111}^{(3)}(-\omega, -\omega, \omega, \omega)$. The macroscopic static field is given by

$$\frac{\partial^2 E_x}{\partial x^2} + \frac{\partial^2 E_x}{\partial y^2} = \frac{\mu}{4\epsilon_{11}} \left\{ \frac{\partial^2 f_1}{\partial x \partial y} - \frac{\partial^2 f_2}{\partial x^2} \right\}, \quad (19)$$

$$\frac{\partial^2 E_y}{\partial x^2} + \frac{\partial^2 E_y}{\partial y^2} = \frac{\mu}{4\epsilon_{11}} \left\{ \frac{\partial^2 f_1}{\partial y^2} - \frac{\partial^2 f_2}{\partial x \partial y} \right\}, \quad (20)$$

where $f_1 = |A_y|^2 - |A_x|^2$, and $f_2 = A_x^* A_y + A_x A_y^*$. Therefore, in general one can generate light evolutions dictated by a whole family of vector Schrödinger equations with tunable nonlinearities. In the limit case of a highly elliptical beam ($\zeta \ll 1$), (20) and (21) can be solved analytically to obtain $E_y \simeq 0$, and $E_x \simeq -\mu f_2/4\epsilon_{11}$. Substitution into (17) and (18), and using that $\epsilon_{11} \gg 1$ (as it is the case in most materials), yields self-phase modulation, cross-phase modulation, and three-wave-mixing coefficients given by a , $(3b + c - 2a)$, and $(2b + 2c - 2a)$, respectively.

Example III. Consider a beam that propagates in a material with point group symmetry $42m$. We let $\hat{z} = \hat{c}$, thus $n_x = n_y \equiv n_1$, $\epsilon_{xx}(0) = \epsilon_{yy}(0) \equiv \epsilon_{11}$, and $\epsilon_{zz}(0) \equiv \epsilon_{33}$. Then, one finds that the macroscopic rectified field vanishes ($E_x = E_y = 0$), but the macroscopic nonlinearities induced by the rectified fields do not. Instead, (11) yields

$$2ik_1 \frac{\partial A_x}{\partial z} + \nabla_{\perp}^2 A_x + k_0^2 \{ \alpha |A_x|^2 A_x + (v + \gamma) [|A_y|^2 A_x + A_y^2 A_x^*] \} = 0, \quad (21)$$

$$2ik_1 \frac{\partial A_y}{\partial z} + \nabla_{\perp}^2 A_y + k_0^2 \{ \alpha |A_y|^2 A_y + (v + \gamma) [|A_x|^2 A_y + A_x^2 A_y^*] \} = 0, \quad (22)$$

where $v = n_1^8 r_{123}^2/4(\epsilon_{33} + 2)$, $\alpha = 3/4 \chi_{1111}^{(3)}(-\omega, -\omega, \omega, \omega)$, and $\gamma = 3/2 \chi_{1111}^{(3)}(-\omega, -\omega, \omega, \omega)$. Truncated versions of these equations, supplemented by Ginzburg–Landau nonlinearities, describe standing waves in water channels [22], and are known to have solitary-wave solutions at some suitable limit [23]. The existence of families of solitons generated by (21) and (22) is one of the programs we put forward here for future investigation.

Settings where the macroscopic rectified field vanishes but the macroscopic nonlinearity does not are also found, e.g., with similar conditions as above but in materials with point group symmetry $4mm$. In such cases, one readily finds that the nonlinear polarization arising from optical rectification yields evolutions equations for A_x, A_y along the diffracting beam axis given by the Manakov equations [24], which gives a detailed setting where experimentally explore the properties of Manakov-like solitons when induced by the combination of OR and the electro-optic effect [6]. Manakov-like solitons has been demonstrated experimentally in other configurations [25].

The central result put forward in this paper is that taking into account the full self-rectified effects and beam shape when calculating the interaction of optical waves with static fields through optical rectification introduces a combination of nonlinearities that can manifest themselves in a rich variety of different ways. Here we considered a few illustrative examples of the potential of the new scheme, but many other possibilities contained in (11)–(14) remain open to future exploration. In particular, the mechanism uncovered here can be also implemented in geometries with large photorefractive or frequency-mixing quadratic nonlinearities, correspondingly enhancing the variety of solitary-wave structures sustained by competing nonlinearities. For example, a direct

application of this scheme is the generation of engineerable competing quadratic and cubic nonlinearities in systems where second-harmonic generation is accompanied by optical rectification, by exploiting the geometrical conditions that determine the magnitude of the rectified fields [28].

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