Journal of Applied Mathematics and Physics, 2024, 12, 3567-3578
https://www.scirp.org/journal/jamp
ISSN Online: 2327-4379
ISSN Print: 2327-4352

Simultons in Nonstationary CARS by Polaritons: Energy and Velocity

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How to cite this paper: Feshchenko, V.P. and Feshchenko, G.P. (2024) Simultons in Nonstationary CARS by Polaritons: Energy and Velocity. *Journal of Applied Mathematics and Physics*, **12**, 3567-3578.
https://doi.org/10.4236/jamp.2024.1210212

Received: August 14, 2024
Accepted: October 27, 2024
Published: October 30, 2024

Abstract

This paper is the continuation of our previous research in which we studied such aspects of CARS spectroscopy in dipole-active crystals by polaritons as the regimes of coherent simultaneous propagation of three waves (anti-Stokes, Stokes, and the pump field) to increase the efficiency of CARS as a spectroscopic method. In our previous research, we have shown the possibility of the existence of simultons at all frequencies of interacting waves. All interacting waves were supposed to be linearly polarised and plane, the medium was assumed to be nonmagnetic, and the medium was transparent at frequencies of anti-Stokes, Stokes, and the pump field (laser). The purpose of the present paper is to consider the energy carried by electromagnetic waves and its relationship with the gain factor and velocity of the simultons.

Keywords

Simultons, Coherent Anti-Stokes Raman Scattering, Spectroscopy, Polaritons

1. Introduction

One of the areas of our scientific interest is the study of the nonstationary SRS with excitation of polar optical phonons, resulting in the formation of different types of solitons [1]-[6]. In these papers, we investigated the transition regimes of solitons in SRS [1], those asymptotic regimes of wave propagation in the case of CARS by polaritons [4], considering the possibility of existence and simultaneous propagation of several solitons (simultons) at different polarizations (polarization simultons) [5] [6], etc. Many important characteristics of solitons were found (amplitudes at frequencies of interacting waves, time durations, relation with the characteristics of the medium, etc.) However, some questions still need to be investigated. One of them is the relationship between the soliton energy and its velocity of propagation as a typical nonlinear formation. More to the point, since we specifically research the processes of SRS (which have a threshold and are characterized by a gain factor) on dipole-active phonons (which are "heavy" when compared to electrons), we can expect some process of "slowing down" since the electromagnetic waves were at the beginning involved in quasi-resonant interaction with phonons and then formed simultons. To evaluate the energy of the system, we used the Manley-Rowe relations [7]-[9] as the important relations in the theory of waves, which predict the distribution of energy at the frequencies of the interacting waves.

Those relations were used to find the distribution of energy in many systems, such as metamaterials [10]-[12], plasma [13] [14], cavities [15], nonlinear crystals [16] [17] [18], and optical fibers [19]. The new approach for the application of Manley-Rowe relations was considered in [20], where Manley-Rowe relations were formulated for a discrete Hamiltonian system with an arbitrary number of resonances. Their quantum derivation was presented in [21], in which the Ermakov-Lewis quantum invariant for the time-dependent harmonic oscillator was expressed in terms of photon number and phase operators. The identification of these variables is made under the correspondence principle and the amplitude and phase representation of the classical orthogonal function's invariant. In the specific case where the excitations represent the photon number, these relations were equivalent to the power density transport equations derived in nonlinear optical processes. The combination of the inverse-scattering method and Manley-Rowe relations was considered in [22]. Of course, those relations are also applicable in the case of propagation of the solitons. The questions related to that were considered, for instance, in [23]-[31]. Consequently, the distribution of energy of the electromagnetic wave(s) in the nonlinear substance determines some other properties of both the medium and wave(s) [32] [33].

As mentioned previously, one of the important features, especially in optical communications, is the velocity of the electromagnetic waves (in our case solitons) affected by their energy. Knowing how the speed of soliton(s) depends on the energy is an important factor in the case of high-speed optical communications [34]-[42]. For example, [40] considered the interactions of two identical, orthogonally polarized vector solitons in an optical fiber with two polarization directions. It was shown by using the numerical simulations that sufficiently fast solitons were moving by each other without much interaction, but below a critical velocity, the solitons might be captured. In certain bands of initial velocities, the solitons were initially captured, but separated after passing each other twice, a phenomenon known as the two-bounce or two-pass resonance. In this paper, the authors also derived an analytic formula for the critical velocity and determined the locations of these "resonance windows". Some interesting theoretical aspects of that problem were considered in [43]-[49]. In [44] presented a systematic study on the dynamics of an ultraslow optical soliton in a cold, highly resonant three-state atomic system under Raman excitation. Using a method of multiple scales, a modified nonlinear Schrödinger equation with high-order corrections was derived to describe the effects of linear and differential absorption, nonlinear dispersion, delay response of nonlinear refractive index, diffraction, and third-order dispersion. Taking these effects as perturbations, the evolution of the ultraslow optical soliton using a standard soliton perturbation theory was investigated in detail. It was shown that due to these high-order corrections, the ultraslow optical soliton undergoes deformation, change of propagating velocity, and shift of oscillating frequency. In [46] presented a feedback mechanism for dissipative solitons in the cubic complex Ginzburg-Landau equation with a nonlinear gradient term. It was demonstrated that, for a small magnitude of the nonlinear gradient term, simple types of scaling behavior were found for the amplitude, the full width at half maximum, the velocity, and the effective frequency of the stable pulse as a function of the magnitude of the nonlinear gradient term. However, those features of propagating simultons (the dependence of velocity from boundary conditions, etc.) were not fully covered in the case of dipole-active crystals. That is why, in this paper, we consider some additional aspects of such propagation (for instance, the relationship between velocity and the gain factor) for the simultons in the case of CARS by polaritons [4], which was investigated by the authors earlier.

2. Basic Equations

We begin with considering the nonlinear interaction of four electromagnetic waves: anti-Stokes, Stokes, pump (laser), and polariton. Those waves are suggested to be linearly polarized plane waves. Here, it is also assumed that the nonlinear medium takes the form of a layer bounded by the planes $z = 0$ and $z = L$ (and is nonmagnetic). The pump wave

$$\vec{E}_l\left(\vec{r},t\right)=\hat{e}_l A_l\left(z,t\right)\exp\left[i\left(k_l^z z-\omega_l t\right)\right]+c.c. \tag{1}$$

propagates along the z-axis. The subscripts a, l, s, and p denote the anti-Stokes, pump (laser), stokes, and polariton wave fields, $\omega_{a,l,s,p}$ are the frequencies, $n_{a,l,s,p}$ and $\vec{k}_{a,l,s,p}$ are the refractive indices, the wave vectors in the unpumped medium, and $\hat{e}_{a,l,s,p}$ the real unit vectors of electromagnetic fields. The nonlinear medium is assumed to be transparent at the frequencies $\omega_{a,l,s}$. We use the anti-Stokes, Stokes, and polariton fields in the form

$$\vec{E}_a\left(\vec{r},t\right)=\hat{e}_a A_a\left(z,t\right)\exp\left[i\left(k_a^z z-\omega_a t\right)\right]+c.c., \tag{2}$$

$$\vec{E}_s\left(\vec{r},t\right)=\hat{e}_s A_s\left(z,t\right)\exp\left[i\left(k_s^z z-\omega_s t\right)\right]+c.c., \tag{3}$$

$$\vec{E}_p\left(\vec{r},t\right)=\hat{e}_p A_p\left(z,t\right)\exp\left[i\left(W^z z-\omega_p t\right)\right]+c.c., \tag{4}$$

where $k_{a,s}=q_{a,s}n_{a,s}$; $q_{a,s}=\omega_{a,s}/c$; $W^z=k_l^z-k_s^z$; $\omega_p=\omega_l-\omega_s$.

In the process of CARS, the nonlinear interaction of two electromagnetic waves $\omega_{l,s}$ results in the generation of anti-Stokes and polariton waves. The system of shortened equations for the amplitudes $A_{a,l,s,p}$ is obtained from Maxwell's equation by using the standard approximation of slowly varying amplitudes [4] and takes the form

$$\frac{\partial A_a}{\partial z}+\frac{1}{v_a^z}\frac{\partial A_a}{\partial t}=i\frac{2\pi\omega_a}{cn_a\cos\theta_a^z}\left\{\chi_a A_l A_p \mathrm{e}^{i\Delta k^z z}+\gamma_a\left(\left|A_l\right|^2+\left|A_s\right|^2\right)A_a\right\}, \quad (5)$$

$$\frac{\partial A_l}{\partial z}+\frac{1}{v_l^z}\frac{\partial A_l}{\partial t}=i\frac{2\pi\omega_l}{cn_l\cos\theta_l^z}\left\{\chi_{l1}A_s A_p+\chi_{l2}A_a A_p^* \mathrm{e}^{-i\Delta k^z z}\right\}, \quad (6)$$

$$\frac{\partial A_s}{\partial z}+\frac{1}{v_s^z}\frac{\partial A_s}{\partial t}=i\frac{2\pi\omega_s}{cn_s\cos\theta_s^z}\left\{\chi_s A_l A_p^*+\gamma_s\left(\left|A_l\right|^2+\left|A_s\right|^2\right)A_s\right\}, \quad (7)$$

$$\frac{\partial A_p^*}{\partial z}+\frac{1}{v_p^z}\frac{\partial A_p^*}{\partial t}=i\frac{q_p^2\varepsilon_p^\infty}{2W^z}\left(\frac{W^2}{q_p^2\varepsilon_p^\infty}-1\right)A_p^*-i\frac{2\pi q_p^2}{W^z}\left\{\chi_{p1}A_l^* A_s+\chi_{p2}A_a^* A_l \mathrm{e}^{i\Delta k^z z}\right\}, \quad (8)$$

where $\chi_a, \chi_{l1,l2}, \chi_s, \chi_{p1,p2}, \gamma_{a,s}$ are the corresponding tensor contractions of non-resonance quadratic and cubic nonlinear polarizabilities with unit vectors of polarization of interacting waves; e_p^∞ is the non-resonance part of dielectric permeability at frequency ω_p; $v_{a,l,s,p}^z$ are z-components of velocities of waves on $\omega_{a,l,s,p}$; $\Delta k^z \equiv k_l^z + W^z - k_a^z$ is the wave mismatch between the pump, polariton, and anti-Stokes waves.

Given the strong polariton absorption, we have [4]:

$$\left|\frac{\partial A_p^*}{\partial z}\right|\approx\left|\frac{1}{v_p^z}\frac{\partial A_p^*}{\partial t}\right|\ll\frac{q_p^2\varepsilon_p^\infty}{2W^z}\left(\frac{W^2}{q_p^2\varepsilon_p^\infty}-1\right)A_p^*, \quad (9)$$

so that we can neglect in (8) the terms with the derivatives after which this equation yields

$$A_p^*=\frac{4\pi q_p^2}{W^2-q_p^2\varepsilon_p^\infty}\left(\chi_{p1}A_l^* A_s+\chi_{p2}A_a^* A_l \mathrm{e}^{i\Delta k^z z}\right). \quad (10)$$

If we insert the obtained expression for the amplitude of polariton wave in Equations (5)-(7), we can obtain a system of 3 differential equations $A_{a,l,s}$ as follows:

$$\frac{\partial A_a}{\partial z}+\frac{1}{v_a^z}\frac{\partial A_a}{\partial t}=i\frac{2\pi\omega_a}{cn_a\cos\theta_a^z}\left\{\frac{4\pi q_p^2\chi_a\chi_{p1}^*}{W^2-q_p^2\varepsilon_p^\infty}A_l^2 A_s^* \mathrm{e}^{i\Delta k^z z}+\gamma_{a1}\left|A_l\right|^2 A_a+\gamma_a\left|A_s\right|^2 A_a\right\}, \quad (11)$$

$$\frac{\partial A_l}{\partial z}+\frac{1}{v_l^z}\frac{\partial A_l}{\partial t}=i\frac{2\pi\omega_l}{cn_l\cos\theta_l^z}\left\{\frac{4\pi q_p^2\left(\chi_{l1}\chi_{p2}^*+\chi_{l2}\chi_{p1}\right)}{W^2-q_p^2\varepsilon_p^\infty}A_a A_l^* A_s \mathrm{e}^{-i\Delta k^z z}+\frac{4\pi q_p^2\chi_{l2}\chi_{p2}}{W^2-q_p^2\varepsilon_p^\infty}\left|A_a\right|^2 A_l+\gamma_{l1}\left|A_s\right|^2 A_l+\gamma_l\left|A_l\right|^2 A_l\right\}, \quad (12)$$

$$\frac{\partial A_s}{\partial z}+\frac{1}{v_s^z}\frac{\partial A_s}{\partial t}=i\frac{2\pi\omega_s}{cn_s\cos\theta_s^z}\left\{\frac{4\pi q_p^2\chi_s\chi_{p2}}{W^2-q_p^2\varepsilon_p^\infty}A_l^2 A_a^* \mathrm{e}^{i\Delta k^z z}+\gamma_{s1}\left|A_l\right|^2 A_s+\gamma_s\left|A_s\right|^2 A_s\right\}, \quad (13)$$

where $q_p \equiv \omega_p/c$, $\gamma_{a1}\equiv\gamma_a+\frac{4\pi q_p^2\chi_a\chi_{p2}^*}{W^2-q_p^2\varepsilon_p^\infty}$, $\gamma_{s1}\equiv\gamma_s+\frac{4\pi q_p^2\chi_s\chi_{p1}}{W^2-q_p^2\varepsilon_p^\infty}$, and

$$\gamma_{l1} \equiv \gamma_l + \frac{4\pi q_p^2 \chi_{l1} \chi_{p1}^*}{W^2 - q_p^2 \varepsilon_p^\infty}.$$

The systems (11)-(13) can be simplified if we use new variables

$$A_a' \equiv A_a e^{-\frac{i\Delta k^z z}{2}} \tag{14}$$

and

$$A_s' \equiv A_s e^{-\frac{i\Delta k^z z}{2}}. \tag{15}$$

The systems (11)-(13) in terms of $A_{a,s}'$ can be rewritten as follows:

$$\frac{\partial A_a'}{\partial z} + \frac{1}{v_a^z}\frac{\partial A_a'}{\partial t} = i\frac{2\pi\omega_a}{cn_a\cos\theta_a^z}\left\{\frac{4\pi q_p^2 \chi_a \chi_{p1}^*}{W^2 - q_p^2\varepsilon_p^\infty} A_l^2 A_s'^* + \gamma_{a1}\left|A_l\right|^2 A_a' + \gamma_a\left|A_s'\right|^2 A_a'\right\}, \tag{16}$$

$$\begin{aligned}\frac{\partial A_l}{\partial z} + \frac{1}{v_l^z}\frac{\partial A_l}{\partial t} = i\frac{2\pi\omega_l}{cn_l\cos\theta_l^z}&\left\{\frac{4\pi q_p^2\left(\chi_{l1}\chi_{p2}^* + \chi_{l2}\chi_{p1}\right)}{W^2 - q_p^2\varepsilon_p^\infty} A_a' A_s' A_l^*\right.\\ &\left. + \frac{4\pi q_p^2 \chi_{l2}\chi_{p2}}{W^2 - q_p^2\varepsilon_p^\infty}\left|A_a'\right|^2 A_l + \gamma_{l1}\left|A_s'\right|^2 A_l + \gamma_l\left|A_l\right|^2 A_l\right\},\end{aligned} \tag{17}$$

$$\frac{\partial A_s'}{\partial z} + \frac{1}{v_s^z}\frac{\partial A_s'}{\partial t} = i\frac{2\pi\omega_s}{cn_s\cos\theta_s^z}\left\{\frac{4\pi q_p^2 \chi_s \chi_{p2}}{W^2 - q_p^2\varepsilon_p^\infty} A_l^2 A_a'^* + \gamma_{s1}\left|A_l\right|^2 A_s' + \gamma_s\left|A_s\right|^2 A_s'\right\}. \tag{18}$$

where $k_{a,s} = q_{a,s} n_{a,s}$; $q_{a,s} = \omega_{a,s}/c$; $W^z = k_l^z - k_s^z$; $\omega_p = \omega_l - \omega_s$. $\chi_a, \chi_{l1,l2}, \chi_s, \chi_{p1,p2}, \gamma_{a,s}$ are the corresponding tensor contractions of non-resonance quadratic and cubic nonlinear polarizabilities with unit vectors of polarization of interacting waves; e_p^∞ is the non-resonance part of dielectric permeability at frequency ω_p; $v_{a,l,s,p}^z$ are z-components of velocities of waves on $\omega_{a,l,s,p}$; $\Delta k^z \equiv k_l^z + W^z - k_a^z$ is the wave mismatch between the pump, polariton, and anti-Stokes waves. We assumed a "weak" wave mismatch at Stokes and anti-Stokes frequencies, that is

$$\left|\frac{\partial A_{a,s}'}{\partial z} + \frac{1}{v_{a,s}^z}\frac{\partial A_{a,s}'}{\partial t}\right| \gg \frac{\Delta k^z}{2} A_{a,s}'.$$

$$q_p \equiv \omega_p/c,\quad \gamma_{a1} \equiv \gamma_a + \frac{4\pi q_p^2 \chi_a \chi_{p2}^*}{W^2 - q_p^2\varepsilon_p^\infty},\quad \gamma_{s1} \equiv \gamma_s + \frac{4\pi q_p^2 \chi_s \chi_{p1}}{W^2 - q_p^2\varepsilon_p^\infty}, \text{ and}$$

$$\gamma_{l1} \equiv \gamma_l + \frac{4\pi q_p^2 \chi_{l1}\chi_{p1}^*}{W^2 - q_p^2\varepsilon_p^\infty}.$$

3. The Manley-Rowe Relation for Simultaneously Propagating Waves at Frequencies $\omega_{a,l,s}$

To facilitate the further analysis of the systems (16)-(18), we bring it to unitless form first. To do that, we multiply both the left and right part of each equation by the factor z_0/A_0 (A_0 and τ_0 are the peak amplitude and characteristic duration of the pump, $z_0 = c\tau_0$). After that, the systems (16)-(18) can be reduced to

$$\frac{\partial \tilde{A}_a'}{\partial \tilde{z}} + \frac{1}{\tilde{v}_a^z}\frac{\partial \tilde{A}_a'}{\partial \tilde{t}} = i\left\{C_{a1}\tilde{A}_l^2\tilde{A}_s'^* + C_{a2}\left|\tilde{A}_l\right|^2\tilde{A}_a' + C_{a3}\left|\tilde{A}_s'\right|^2\tilde{A}_a'\right\}, \tag{19}$$

$$\frac{\partial \tilde{A}_l}{\partial \tilde{z}} + \frac{1}{\tilde{v}_l^z}\frac{\partial \tilde{A}_l}{\partial \tilde{t}} = i\left\{C_{l1}\tilde{A}_a'\tilde{A}_s'\tilde{A}_l^* + C_{l2}\left|\tilde{A}_s'\right|^2\tilde{A}_l + C_{l3}\left|\tilde{A}_l\right|^2\tilde{A}_l + C_{l4}\left|\tilde{A}_a'\right|^2\tilde{A}_l\right\}, \tag{20}$$

$$\frac{\partial \tilde{A}_s'}{\partial \tilde{z}} + \frac{1}{\tilde{v}_s^z}\frac{\partial \tilde{A}_s'}{\partial \tilde{t}} = i\left\{C_{s1}\tilde{A}_l^2\tilde{A}_a'^* + C_{s2}\left|\tilde{A}_l\right|^2\tilde{A}_s' + C_{s3}\left|\tilde{A}_s'\right|^2\tilde{A}_s'\right\}, \tag{21}$$

where $\tilde{A}_{a,s}' \equiv \frac{A_{a,s}'}{A_0}$, $\tilde{A}_l \equiv \frac{A_l}{A_0}$, $\tilde{t} \equiv \frac{t}{\tau_0}$, $C_{a1} \equiv \frac{2\pi\omega_a z_0}{cn_a\cos\theta_a^z}\frac{4\pi q_p^2\chi_a\chi_{p1}^* A_0^2}{W^2 - q_p^2\varepsilon_p^\infty}$,

$$C_{a2} \equiv \frac{2\pi\omega_a z_0}{cn_a\cos\theta_a^z}\gamma_{a1}A_0^2,\quad C_{a3} \equiv \frac{2\pi\omega_0 z_0}{cn_a\cos\theta_a^z}\gamma_a A_0^2,\quad C_{l2} \equiv \frac{2\pi\omega_l z_0}{cn_l\cos\theta_l^z}\gamma_{s1}A_0^2,$$

$$C_{l1} \equiv \frac{2\pi\omega_l z_0}{cn_l\cos\theta_l^z}4\pi q_p^2\left(\chi_{l1}\chi_{p2}^* + \chi_{l2}\chi_{p1}\right)A_0^2, \tag{22}$$

$$C_{l4} \equiv \frac{2\pi\omega_l z_0}{cn_l\cos\theta_l^z}\frac{4\pi q_p^2\chi_{l2}\chi_{p2}A_0^2}{W^2 - q_p^2\varepsilon_p^\infty},\quad C_{s1} \equiv \frac{2\pi\omega_s z_0}{cn_s\cos\theta_s^z}\frac{4\pi q_p^2\chi_s\chi_{p2}A_0^2}{W^2 - q_p^2\varepsilon_p^\infty},$$

$$C_{s2} \equiv \frac{2\pi\omega_s z_0}{cn_s\cos\theta_s^z}\gamma_{s1}A_0^2,\quad C_{s3} \equiv \frac{2\pi\omega_s z_0}{cn_s\cos\theta_s^z}\gamma_s A_0^2.$$

Here, we will show the simplified form proving that the total energy per area is constant during the process of wave propagation. In that simplified form, all coefficients are of the same order of magnitude:

$$C_{a1} \approx C_{a2} \approx C_{a3} \approx C_{l2} \approx C_{l3} \approx C_{l4} \approx C_{s1} \approx C_{s2} \approx C_{s3} \approx C,\ C_{l1} \approx 2C. \tag{23}$$

Then, we multiply each of the Equations (23)-(25) by the corresponding *c.c.* amplitude and add with its *c.c.* counterpart:

$$\begin{gathered} A_a^{\sim\prime *}\left(\frac{\partial \tilde{A}_a'}{\partial \tilde{z}} + \frac{1}{\tilde{v}_a^z}\frac{\partial \tilde{A}_a'}{\partial \tilde{t}}\right) = iA_a^{\sim\prime *}\left\{C\tilde{A}_l^2\tilde{A}_s'^* + C\left|\tilde{A}_l\right|^2\tilde{A}_a' + C\left|\tilde{A}_s'\right|^2\tilde{A}_a'\right\}, \\ A_a^{\sim\prime}\left(\frac{\partial \tilde{A}_a^{*\prime}}{\partial \tilde{z}} + \frac{1}{\tilde{v}_a^z}\frac{\partial \tilde{A}_a^{*\prime}}{\partial \tilde{t}}\right) = -iA_a^{\sim\prime}\left\{C\tilde{A}_l^{*2}\tilde{A}_s' + C\left|\tilde{A}_l\right|^2\tilde{A}_a^{*\prime} + C\left|\tilde{A}_s'\right|^2\tilde{A}_a^{*\prime}\right\}, \end{gathered} \tag{24}$$

$$\begin{gathered} \tilde{A}_l^*\left(\frac{\partial \tilde{A}_l}{\partial \tilde{z}} + \frac{1}{\tilde{v}_l^z}\frac{\partial \tilde{A}_l}{\partial \tilde{t}}\right) = i\tilde{A}_l^*\left\{2C\tilde{A}_a'\tilde{A}_s'\tilde{A}_l^* + C\left|\tilde{A}_s'\right|^2\tilde{A}_l + C\left|\tilde{A}_l\right|^2\tilde{A}_l + C\left|\tilde{A}_a'\right|^2\tilde{A}_l\right\}, \\ \tilde{A}_l\left(\frac{\partial \tilde{A}_l^*}{\partial \tilde{z}} + \frac{1}{\tilde{v}_l^z}\frac{\partial \tilde{A}_l^*}{\partial \tilde{t}}\right) = -i\tilde{A}_l\left\{2C\tilde{A}_a^{*\prime}\tilde{A}_s^{*\prime}\tilde{A}_l + C\left|\tilde{A}_s'\right|^2\tilde{A}_l^* + C\left|\tilde{A}_l\right|^2\tilde{A}_l^* + C\left|\tilde{A}_a'\right|^2\tilde{A}_l^*\right\}, \end{gathered} \tag{25}$$

$$\begin{gathered} A_s^{\sim *}\left(\frac{\partial \tilde{A}_s'}{\partial \tilde{z}} + \frac{1}{\tilde{v}_s^z}\frac{\partial \tilde{A}_s'}{\partial \tilde{t}}\right) = iA_s^{\sim *}\left\{C\tilde{A}_l^2\tilde{A}_a'^* + C\left|\tilde{A}_l\right|^2\tilde{A}_s' + C\left|\tilde{A}_s'\right|^2\tilde{A}_s'\right\}, \\ A_s^{\sim}\left(\frac{\partial \tilde{A}_s^{*\prime}}{\partial \tilde{z}} + \frac{1}{\tilde{v}_s^z}\frac{\partial \tilde{A}_s^{*\prime}}{\partial \tilde{t}}\right) = -iA_s^{\sim}\left\{C\tilde{A}_l^{*2}\tilde{A}_a' + C\left|\tilde{A}_l\right|^2\tilde{A}_s^{*\prime} + C\left|\tilde{A}_s'\right|^2\tilde{A}_s^{*\prime}\right\}. \end{gathered} \tag{26}$$

When we add those equations together, the right part yields 0, which means that

$$\frac{\partial\left|\tilde{A}_a'\right|^2}{\partial\tilde{z}}+\frac{1}{\tilde{v}_a^z}\frac{\partial\left|A_a^{\sim\prime}\right|^2}{\partial\tilde{t}}+\frac{\partial\left|\tilde{A}_l'\right|^2}{\partial\tilde{z}}+\frac{1}{\tilde{v}_l^z}\frac{\partial\left|A_l^{\sim\prime}\right|^2}{\partial\tilde{t}}+\frac{\partial\left|\tilde{A}_s'\right|^2}{\partial\tilde{z}}+\frac{1}{\tilde{v}_s^z}\frac{\partial\left|A_s^{\sim\prime}\right|^2}{\partial\tilde{t}}=0. \quad (27)$$

If we introduce the energy per area delivered by any wave as

$$W_{a,s}\equiv\int_{-\infty}^{\infty}\left|A_{a,s}^{\sim}{}'\right|^2\mathrm{d}\tilde{t},W_l\equiv\int_{-\infty}^{\infty}\left|A_l^{\sim}\right|^2\mathrm{d}\tilde{t}\ . \quad (28)$$

Then, it can be easily shown (after integration over time) that

$$\frac{\mathrm{d}}{\mathrm{d}\tilde{z}}\left(W_a+W_l+W_s\right)=0, \quad (29)$$

which means that electromagnetic energy is conserved when traveling in a nonlinear medium.

4. Simultons Speed in the Case of CARS by Polaritons

To do that, we will analyze the system of nonlinear equations found in [4].

$$\frac{\mathrm{d}Q}{\mathrm{d}\tilde{\xi}}=\alpha Q^2\sin\Phi, \quad (30)$$

$$\frac{\mathrm{d}\Phi}{\mathrm{d}\tilde{\xi}}=2\alpha Q\cos\Phi+\beta Q, \quad (31)$$

where

$$\lambda_a^2=-\kappa_aC_{a1},\ \ \lambda_l^2=\kappa_lC_{l1},\ \ \lambda_s^2=-\kappa_sC_{s1},\ \ \alpha\equiv2\lambda_a\lambda_s\lambda_l^2, \quad (32)$$

$$\beta\equiv2\kappa_l\left(C_{l2}\lambda_s^2+C_{l3}\lambda_l^2+C_{l4}\lambda_a^2\right)-\kappa_s\left(C_{s2}\lambda_l^2+C_{s3}\lambda_s^2\right)-\kappa_a\left(C_{a2}\lambda_l^2+C_{a3}\lambda_s^2\right).$$

$$\tilde{A}_{a,s}'\left(\tilde{z},\tilde{t}\right)\equiv B_{a,s}\left(\tilde{\xi}\right)\mathrm{e}^{i\Phi_{a,s}\left(\tilde{\xi}\right)},\ \ \tilde{A}_l\left(\tilde{z},\tilde{t}\right)\equiv B_l\left(\tilde{\xi}\right)\mathrm{e}^{i\Phi_l\left(\tilde{\xi}\right)},$$

$$Q\equiv-\frac{B_a^2}{\kappa_aC_{a1}}=-\frac{B_s^2}{\kappa_sC_{s1}}=\frac{B_l^2}{\kappa_lC_{l1}}$$

$\tilde{\xi}\equiv\tilde{t}-\tilde{z}/\tilde{v}^z$; $\tilde{v}^z$ is the velocity of simultaneously propagating waves at the frequencies $\omega_{a,l,s}$; $B_{a,l,s}$ and $\Phi_{a,l,s}$ are the real amplitudes and phases of the waves, respectively, $\kappa_{a.s.l}\equiv\tilde{v}_{a,s,l}^z\tilde{v}^z\Big/\left(\tilde{v}^z-v_{a,s,l}^z\tilde{v}_{a,s,l}^z\right)$, $\Phi\equiv2\Phi_l-\Phi_s-\Phi_a$.

We can reduce the number of equations by using the integral of motion

$$Q\left(\tilde{\xi}\right)=\frac{Const}{\sqrt{2\cos\Phi\left(\tilde{\xi}\right)+\tilde{\beta}}}, \quad (33)$$

where $Q>0$, $\tilde{\beta}>2$ ($\tilde{\beta}\equiv\beta/\alpha$).

It is easy to show (by using the system of equations with the integral of motion) that

$$\int_{-\infty}^{\infty}Q\left(\tilde{\xi}\right)\mathrm{d}\tilde{\xi}=\frac{2\pi}{\sqrt{\left(\beta+2\alpha\right)\left(\beta-2\alpha\right)}} \quad (34)$$

On the other hand, we can introduce the ratio of the energy (per unit area) to the energy (per unit area) for the laser pump as

$$\tilde{W}_{a,l,s} \equiv \int_{-\infty}^{\infty} \tilde{B}_{a,l,s}^2 \mathrm{d}\tilde{\xi} \tag{35}$$

and the energy conservation relationship as

$$\tilde{W}_a + \tilde{W}_l + \tilde{W}_s = \tilde{W}_0 , \tag{36}$$

where $\tilde{W}_0$ is the total energy per unit area of all interacting electromagnetic waves at the input to the nonlinear media.

Consequently, when we consider the left part of Equation (36), we can get

$$\begin{aligned} \tilde{W}_a + \tilde{W}_l + \tilde{W}_s &= \int_{-\infty}^{\infty} B_a^2 \mathrm{d}\tilde{\xi} + \int_{-\infty}^{\infty} B_l^2 \mathrm{d}\tilde{\xi} + \int_{-\infty}^{\infty} B_s^2 \mathrm{d}\tilde{\xi} \\ &= \lambda_a^2 \int_{-\infty}^{\infty} Q(\tilde{\xi}) \mathrm{d}\tilde{\xi} + \lambda_l^2 \int_{-\infty}^{\infty} Q(\tilde{\xi}) \mathrm{d}\tilde{\xi} + \lambda_s^2 \int_{-\infty}^{\infty} Q(\tilde{\xi}) \mathrm{d}\tilde{\xi} \\ &= \left(\lambda_a^2 + \lambda_l^2 + \lambda_s^2\right) \int_{-\infty}^{\infty} Q(\tilde{\xi}) \mathrm{d}\tilde{\xi}. \end{aligned} \tag{37}$$

Finally, when we use Equations (36) and (37), we get the relationship between the boundary conditions and the simultons speed

$$\frac{2\pi\left(\lambda_a^2 + \lambda_l^2 + \lambda_s^2\right)}{\sqrt{(\beta + 2\alpha)(\beta - 2\alpha)}} = \tilde{W}_0 \tag{38}$$

In the case of weak dispersion ($C_{a1} \approx C_{s1} \approx C_{l1} \approx C$ (in the next topic, it is shown that $g \approx C$ where g is the gain factor of Raman scattering) so that the coefficient $\alpha = \lambda_a \lambda_l^2 \lambda_s \approx C^2 \approx g^2$, $g \approx C \approx 8\pi^2 \omega z_0 \chi^2 A_0^2 / (cn)$ [4]) it can be shown that

$$\frac{1}{\tilde{v}^z} \approx \frac{1}{\tilde{v}_{em}^z} + g\tilde{W}_0 \tag{39}$$

where $\tilde{v}_{em}^z = \tilde{v}_l^z \approx \tilde{v}_a^z \approx \tilde{v}_s^z$. To do the numerical estimation we use the results of [50] [51]:

We take as a typical value of the gain factor 10^{-8} cm/W and the pump intensity of 80 MW/cm^2, which would give us an estimation of ≈1 cm^{-1}. Thus, if we take the crystal with a length of 1 cm, we could get the value for our $g \approx 1$. As for $\tilde{W}_0$, the restriction in the form of conservation of energy results in ≤ 1.

5. Conclusion

In this paper, we have found that the system of differential equations that model the process of coherent anti-Stokes Raman scattering by polaritons in crystals obeys the Manley-Rowe relation. We have also found the relationship between simultons velocity, the gain factor of Raman scattering, and the energy of the electromagnetic waves involved in the process of CARS. For instance, from the simplified Formula (39), we can conclude the processes of SRS may result in a decrease of simultons speed because of the coherent interaction between the electromagnetic fields and phonons (more inertia compared to electrons).

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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Abbreviations

SRS (Stimulated Raman Scattering);

CARS (Coherent Anti-Stokes Raman Scattering).