Energy conservation in self-phase modulation

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Spectral broadening of ultrashort laser pulses is simultaneously described by either self-phase modulation (SPM) or four-wave mixing (FWM). The latter implies the instantaneous conservation of both the photon number and energy, while the former describes a time-dependent frequency shift, implying a violation of the energy conservation if the number of photons is to be conserved in each time slice. We resolve this paradox by considering the transient energy storage in the propagation medium, which can be calculated in the SPM formalism via the dephasing between the incident pulse and the medium polarization leading to an effective imaginary part in the third-order susceptibility. In parallel, considering the temporal variation of the incident intensity in FWM offsets the instantaneous frequency.

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I. INTRODUCTION

Nonlinear optics was first investigated in condensed matter. The spectacular phenomenon of supercontinuum generation was observed by Alfano and Shapiro [1,2], who identified four-wave mixing (FWM) [1] and self-phase modulation (SPM) [2] as the origin of this wide spectral broadening [3–11]. The advent of chirped pulse amplification [12] allowed even more efficient spectral broadening, as well as its observation in gases including atmospheric pressure air.

Four-wave mixing describes spectral broadening as the interaction of plane monochromatic waves. Two photons at frequencies ω_1 and ω_2 interact through the $\chi^{(3)}$ susceptibility to generate two photons at ω_3 and ω_4. The energy conservation imposes $\omega_1 + \omega_2 = \omega_3 + \omega_4$, so the spectrum should remain symmetrical after broadening via FWM, and the number of photons is conserved at any time.

The same spectral broadening is alternatively described by SPM as a deformation of the pulse. After propagating over a distance z, the carrier wave experiences a time-dependent frequency shift

$$\Delta\omega(t) = -k_0 z n_2 I'(t) z, \tag{1}$$

where the prime denotes the temporal derivation, $k_0 = n_0\omega/c$, n_0 and $n_2 = 3\chi^{(3)}/4n_0\epsilon_0 c$ are the linear and nonlinear refractive indices, respectively, ω is the angular frequency of the incident pulse, I is its intensity, and c is the velocity of light. Self-phase modulation intrinsically induces an asymmetric time-dependent frequency shift. In most common media such as air or glass, $n_2 > 0$, so the leading edge of the pulse is redshifted while its trailing edge is blueshifted. Such a time-dependent frequency shift is incompatible with the simultaneous conservation of the pulse energy and of the number of photons.

In this paper, we address this paradox. By taking into account the dephasing induced by SPM between the incident electric field and the polarization of the propagation medium, we show that the latter transiently stores energy, which restores energy conservation. Furthermore, considering the fast variation of the pulse intensity in the FWM formalism yields the asymmetric transient frequency shifts that are usually described in the SPM formalism.

II. DISCUSSION

A. Energy conservation in SPM

We consider a pulse described within the slowly varying envelope approximation (SVEA), so the electric field is expressed as $E(t) = E_0(t)(e^{-i\omega t} + \text{c.c.})$. It induces a third-order polarization

$$P^{(3)}(t) = \epsilon_0 \chi^{(3)} E(t)^3, \tag{2}$$

with ϵ_0 the permittivity of vacuum. If we neglect absorption and dispersion, the evolution of this third-order polarization in a medium with an eigenfrequency ω_e can be written in a perturbative approach [13]

$$\frac{d^2 P^{(3)}}{dt^2} + \omega_e^2 P^{(3)} = N e Q^{(3)} {P^{(1)}}^3(t), \tag{3}$$

where $P^{(1)} = \chi^{(1)} E$ is the elastic polarizability, $\chi^{(1)}$ is the first-order susceptibility, E is the electric field, N is the local density of electrons, $-e$ is their charge, m is their mass, and

$$Q^{(3)} = \frac{N^3 e^4}{m\epsilon_0^3} \frac{\chi^{(3)}}{{\chi^{(1)}}^4}. \tag{4}$$

Expanding the first-order polarization $P^{(1)}(t) = P_0^{(1)}(t)(e^{i(\omega t - kz - \phi^{(1)})} + \text{c.c.})$ in the term on the right-hand side of Eq. (3), we get

$$\frac{d^2 P^{(3)}}{dt^2} + \omega_e^2 P^{(3)} = N e Q^{(3)} {P_0^{(1)}}^3 \times (e^{3i(\omega t - kz - \phi^{(1)})} + 3e^{i(\omega t - kz - \phi^{(1)})} + \text{c.c.}) \tag{5}$$

We decompose the polarization $P^{(3)}$ into its components P^{SPM} and P^{TH}, respectively oscillating at the fundamental and

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third-harmonic frequencies ω and 3ω:

$$P^{(3)}(t) = P_0^{\mathrm{TH}}(t)(e^{i(3(\omega t-kz)-\phi^{\mathrm{TH}})} + \mathrm{c.c.}) + P_0^{\mathrm{SPM}}(t)(e^{i(\omega t-kz-\phi^{\mathrm{SPM}})} + \mathrm{c.c.}). \quad (6)$$

Here ϕ^{TH} and ϕ^{SPM} are the dephasings of the polarization relative to the electric field for each spectral component. They are chosen such that P_0^{TH} and P_0^{SPM} are real. We focus on the terms oscillating at the fundamental frequency ω, consider the SVEA, identify the real and imaginary parts of P^{SPM}, and calculate $\phi^{(1)}$ from the linear polarization equation (14) [13]. The SVEA implies $d\phi^{\mathrm{SPM}}/dt \ll \omega$ and $d^2P_0^{\mathrm{SPM}}/dt^2 \ll \omega^2 P_0^{\mathrm{SPM}}(t)$, so the terms at frequency ω in Eq. (5) are rewritten

$$\left[\omega_{\mathrm{e}}^2 - \left(\omega - \frac{d\phi^{\mathrm{SPM}}}{dt}\right)^2\right] P_0^{\mathrm{SPM}}(t) + 2i\left(\omega - \frac{d\phi^{\mathrm{SPM}}}{dt}\right)\frac{dP_0^{\mathrm{SPM}}}{dt} + \frac{d^2P_0^{\mathrm{SPM}}}{dt^2} = 3NeQ^{(3)}P_0^{(1)^3}(t)(e^{i(\phi^{\mathrm{SPM}}-\phi^{(1)})} + \mathrm{c.c.}) = 3NeQ^{(3)}(\epsilon_0\chi^{(1)}E_0)^3(e^{i(\phi^{\mathrm{SPM}}-\phi^{(1)})} + \mathrm{c.c.}). \quad (7)$$

Identifying the components parallel and orthogonal to P_0^{SPM} in the complex plane yields

$$\left(\omega_{\mathrm{e}}^2 - \omega^2\right)P_0^{\mathrm{SPM}}(t) = 6NeQ^{(3)}(\epsilon_0\chi^{(1)}E_0)^3 \times \cos(\phi^{\mathrm{SPM}} - \phi^{(1)}), \quad (8)$$

$$2\omega\frac{dP_0^{\mathrm{SPM}}}{dt} = 6NeQ^{(3)}(\epsilon_0\chi^{(1)}E_0)^3 \times \sin(\phi^{\mathrm{SPM}} - \phi^{(1)}), \quad (9)$$

which combine into

$$\phi^{\mathrm{SPM}} - \phi^{(1)} \approx \tan(\phi^{\mathrm{SPM}} - \phi^{(1)}) \quad (10)$$

$$= \frac{2\omega}{\omega_{\mathrm{e}}^2 - \omega^2}\frac{dP_0^{\mathrm{SPM}}}{P_0^{\mathrm{SPM}}(t)dt} \quad (11)$$

$$= \frac{6\omega}{\omega_{\mathrm{e}}^2 - \omega^2}\frac{dE_0}{E_0(t)dt} \quad (12)$$

$$= \frac{3\omega}{\omega_{\mathrm{e}}^2 - \omega^2}\frac{dI}{I(t)dt}. \quad (13)$$

Here $\phi^{(1)}$ can be estimated with a similar derivation, starting from the linear propagation equation [13]

$$\frac{d^2P^{(1)}}{dt^2} + \omega^2 P^{(1)} = \frac{Ne^2}{m}E(t), \quad (14)$$

resulting in

$$\phi^{(1)} = \frac{\omega}{\omega_{\mathrm{e}}^2 - \omega^2}\frac{dI}{I(t)dt}, \quad (15)$$

which finally yields

$$\phi^{\mathrm{SPM}} = 4\phi^{(1)} = \frac{4\omega}{\omega_{\mathrm{e}}^2 - \omega^2}\frac{dI}{I(t)dt}. \quad (16)$$

Far from resonance ($\omega \ll \omega_{\mathrm{e}}$), $\phi^{\mathrm{SPM}} \approx 4\omega/\omega_{\mathrm{e}}^2 T \ll 1$, with T the pulse duration. For example, in air, $\omega_{\mathrm{e}} \approx 80$ nm [14], so for typical experiments with a 100-fs pulse centered at 800 nm, $\phi^{\mathrm{SPM}} \approx 10^{-4}$. Therefore, the supplementary self-phase modulation induced by this dephasing is fully negligible. However, the dephasing ϕ^{SPM} between the driving field E and the resulting polarization P^{SPM} implies an energy transfer between the incident electric field and the propagation medium. The instantaneous power per unit volume transferred to the propagation medium amounts to

$$\mathcal{P}^{\mathrm{SPM}}(t) = E(t)\frac{dP^{\mathrm{SPM}}}{dt}, \quad (17)$$

where the value of dP_0^{SPM}/dt is defined in Eq. (9) so that

$$\mathcal{P}^{\mathrm{SPM}}(t) = 2E_0(t)\cos(\omega t - kz)\Big(-2\omega P_0^{\mathrm{SPM}}(t)\sin(\omega t - kz - \phi^{\mathrm{SPM}}) + 2\frac{dP_0^{\mathrm{SPM}}}{dt}\cos(\omega t - kz - \phi^{\mathrm{SPM}})\Big). \quad (18)$$

Averaging over one or a few optical cycles yields

$$\langle\mathcal{P}^{\mathrm{SPM}}\rangle(t) = -4\omega E_0 P_0^{\mathrm{SPM}}\frac{\omega}{2\pi}\int_0^{2\pi/\omega}\cos(\omega t)\sin(\omega t - \phi^{\mathrm{SPM}})dt + 4E_0\frac{dP_0^{\mathrm{SPM}}}{dt}\frac{\omega}{2\pi}\int_0^{2\pi/\omega}\cos(\omega t)\cos(\omega t - \phi^{\mathrm{SPM}})dt \quad (19)$$

$$= 4\omega E_0 P_0^{\mathrm{SPM}}\frac{\sin\phi^{\mathrm{SPM}}}{2} + 4E_0\frac{dP_0^{\mathrm{SPM}}}{dt}\frac{\cos\phi^{\mathrm{SPM}}}{2}. \quad (20)$$

Plugging the values of P_0^{SPM} and $\frac{dP_0^{\mathrm{SPM}}}{dt}$ from Eqs. (8) and (9), we obtain

$$\langle\mathcal{P}^{\mathrm{SPM}}\rangle(t) \approx 6\epsilon_0 E_0^4\Big[\omega\chi_{\mathrm{cw}}^{(3)}\sin\phi^{\mathrm{SPM}} + \chi_{\mathrm{cw}}^{(3)}\frac{\omega_{\mathrm{e}}^2 - \omega^2}{2\omega}\sin(\phi^{\mathrm{SPM}} - \phi^{(1)})\Big] \quad (21)$$

$$\approx 6\epsilon_0 E_0^4\chi_{\mathrm{cw}}^{(3)}\phi^{(1)}\left(4\omega + 3\frac{\omega_{\mathrm{e}}^2 - \omega^2}{2\omega}\right) \quad (22)$$

$$= 3\epsilon_0\chi_{\mathrm{cw}}^{(3)}E_0^4\phi^{(1)}\left(\frac{3\omega_{\mathrm{e}}^2 + 5\omega^2}{\omega}\right). \quad (23)$$

Introducing the relations $I = 2\epsilon_0 c n_0 E_0^2$ and $\chi_{\mathrm{cw}}^{(3)} = 4n_0\epsilon_0 c n_2/3$, this equation is rewritten

$$\langle\mathcal{P}^{\mathrm{SPM}}\rangle(t) = \frac{n_2 I}{n_0 c}\frac{3\omega_{\mathrm{e}}^2 + 5\omega^2}{\omega_{\mathrm{e}}^2 - \omega^2}\frac{dI}{dt}. \quad (24)$$

In typical conditions (e.g., at a wavelength of 800 nm), $\omega_{\mathrm{e}} \gg \omega$, resulting in

$$\langle\mathcal{P}^{\mathrm{SPM}}\rangle(t) \approx \frac{3n_2 I}{n_0 c}\frac{dI}{dt}. \quad (25)$$

On the leading edge of the pulse, $dI/dt > 0$, so $\langle \mathcal{P}^{\mathrm{SPM}} \rangle > 0$: The field transfers energy to the medium and initiates the dipole oscillation, while on the trailing edge the dipoles return to rest and release their energy into the electromagnetic field. The net energy loss by the pulse is $\int_{-\infty}^{\infty} \langle \mathcal{P}^{\mathrm{SPM}} \rangle dt = 0$ since $I = 0$ at both $t = \pm\infty$. The energy storage in the propagation medium is therefore transient and results in a net energy transfer from the redshifted leading edge of the pulse towards its blueshifted trail. It therefore reconciles redshifts and blueshifts with the simultaneous conservation of energy and of the photon number: A temporal slice of the pulse cannot be considered as an isolated system. Rather, the completion of the system requires one to consider the propagation medium together with the pulse. For typical ultrashort laser filaments in air [15–19] the relative energy transfer can reach 1% per centimeter, enabling the strong reshaping occurring during their propagation.

An alternative way to understand the transient energy storage in the medium consists in grouping the dephasing ϕ^{SPM} with the third-order susceptibility $\chi^{(3)}$ and the associated nonlinear refractive index n_2, resulting in the effective values

$$\chi_{\mathrm{eff}}^{(\mathrm{SPM})} = \chi^{(3)} e^{i\phi^{\mathrm{SPM}}}, \tag{26}$$

$$n_{2,\mathrm{eff}}^{\mathrm{SPM}} = n_2 e^{i\phi^{\mathrm{SPM}}}. \tag{27}$$

Their imaginary components $\chi^{(3)} \sin\phi^{\mathrm{SPM}} \approx \chi^{(3)}\phi^{\mathrm{SPM}}$ and $n_2 \sin\phi^{\mathrm{SPM}} \approx n_2\phi^{\mathrm{SPM}}$ are intrinsically associated with gain or loss.

B. Link with self-steepening

The above-derived transient energy storage has to translate into a depletion of the pulse intensity in the front and a growth in its trail. Indeed, let us consider the self-steepening term affecting the envelope [16]

$$\frac{d\mathcal{E}}{dz} = \frac{-n_2}{n_0 c}\frac{d}{dt}(|\mathcal{E}|^2\mathcal{E}), \tag{28}$$

where $|\mathcal{E}|^2 = I$. The relative variation of $\mathcal{E}$ is therefore

$$\frac{d\mathcal{E}}{\mathcal{E}dz} = \frac{-n_2|\mathcal{E}|^2}{n_0 c}\left(\frac{d|\mathcal{E}|^2}{|\mathcal{E}|^2 dt} + \frac{d\mathcal{E}}{\mathcal{E}dt}\right), \tag{29}$$

which can be converted into the relative variation of the intensity

$$\frac{dI}{2Idz} = \frac{-n_2 I}{n_0 c}\left(\frac{dI}{Idt} + \frac{dI}{2Idt}\right). \tag{30}$$

The local intensity variations due to self-steepening therefore amount to

$$\frac{dI}{dz} = \frac{-3n_2 I}{n_0 c}\frac{dI}{dt}, \tag{31}$$

which identifies with the instantaneous power gained from the medium $-\langle \mathcal{P}^{\mathrm{SPM}} \rangle$ [Eq. (25)], providing an interpretation of the self-steepening in terms of the deformation of the envelope due to the conservation of the photon-number density in spite of their energy drift due to the SPM-induced frequency change. This interpretation is fully compatible with the usual one in terms of the stretching of the temporal pulse slices due to gradients in the group velocity. The latter focuses on the point of view of the wave deformation, while the former translates this deformation and the associated frequency shifts into photon energy and considers its implications for the simultaneous conservation of energy and the photon number.

C. Time-dependent frequency shift in FWM

We will now highlight how the FWM formalism accounts for a time-dependent frequency shift in spite of the energy conservation. We define the instantaneous spectrum at time t_0 as the Fourier transform of the pulse convolved by a temporal gate centered at t_0 and of width τ such that $1/\omega \ll \tau \ll T$, where T is the pulse duration. To allow analytical derivations, we will consider a Gaussian gate in the following. However, other gate shapes may be considered without loss of generality.

Four-wave mixing is typically described by assuming an instantaneous response function of the third-order nonlinear susceptibility (2) and focusing on the nonlinear polarization component in EE^*E, oscillating at ω:

$$P^{\mathrm{SPM}}(t) = 4\epsilon_0^2 c n_0 n_2 E_0(t)^3 (e^{-i\omega t} + \mathrm{c.c.}). \tag{32}$$

Under the paraxial approximation, this nonlinear polarization contributes to the field evolution

$$2ik_0 \left.\frac{dE(t)}{dz}\right|_{\mathrm{NL},\omega} = -\frac{\omega^2}{\epsilon_0 c^2} P^{\mathrm{SPM}}(t), \tag{33}$$

with k_0 the wave vector in vacuum. After plugging (32) into (33), the field evolves as

$$\frac{dE(t)}{dz} = iC_{\mathrm{NL}} E(t)^3, \tag{34}$$

where $C_{\mathrm{NL}} = 2k_0\epsilon_0 c n_0 n_2$. In order to evaluate the frequency shift of the instantaneous spectrum after a short propagation distance dz, we develop the electrical field at first order in dz as

$$E(t,dz) = E(t,0) + iC_{\mathrm{NL}} E(t,0)^3 dz + O(dz). \tag{35}$$

Now let us locally Fourier transform Eq. (35) in the vicinity of t_0 after Taylor expanding the slowly varying field envelope as $E_0(t) \approx E_0(t_0) + (t - t_0)E_0'(t_0)$. Introducing $\Delta\omega = \bar{\omega} - \omega$ and neglecting the influence of dephasing on the field amplitude, we get the intensity spectrum at first order in dz,

$$I(\bar{\omega} \propto |\hat{E}(\bar{\omega}),t_0,dz)|^2 \approx e^{-(\tau^2/2)\Delta\omega^2}\tau^2 E_0^2(t_0) \times [\tfrac{1}{2} - 3C_{\mathrm{NL}} E_0(t_0)E_0'(t_0)\Delta\omega dz], \tag{36}$$

where all terms involving ${E_0'}^2$ have been discarded based on the SVEA. Deriving with respect to $\Delta\omega$ and Taylor expanding again at order 1 in the vicinity of ω yields the spectral peak of the spectrum at time t_0,

$$\Delta\omega^*(t_0) = -6C_{\mathrm{NL}} E_0 E_0' dz \tag{37}$$

$$= -k_0 n_2 I_0'(t_0) dz, \tag{38}$$

where we have introduced the intensity $I_0 = 2n_0\epsilon_0 c E_0^2$. Similarly, the mean frequency is, at first order in dz,

$$\langle\omega\rangle(t_0) \equiv \frac{\int \omega|\hat{E}|^2 d\omega}{\int |\hat{E}|^2 d\omega} = \omega - \frac{24C_{\mathrm{NL}} E_0^3(t_0)E_0'(t_0)dz}{4E_0^2(t_0) + {E_0'}^2\tau^2} \tag{39}$$

$$\approx \omega - 6C_{\mathrm{NL}} E_0 E_0' dz \tag{40}$$

$$= \omega - k_0 n_2 I'(t_0) dz, \tag{41}$$

where $E_0'^2 \tau^2 \ll E_0^2$ due to the SVEA. Therefore, both the peak (38) and mean (41) transient frequency offset calculated with the FWM formalism are equal to those predicted by SPM (1), provided the temporal intensity variation of the incident pulse is taken into account. Note that the frequency offset does not depend on the width τ of the gate, which confirms that it is only a computation intermediate.

III. CONCLUSION

We reconciled the time-dependent frequency shift described by self-phase modulation, with the energy and photon-number conservations implied by the four-wave mixing formalism. An energy transfer occurs from the redshifted leading edge of the pulse to the blueshifted trailing edge, mediated by a transient energy storage in the propagation medium. The latter can be evidenced by considering the dephasing between the driving pulse and the medium polarization, induced by the Kerr effect. This energy transfer is also the origin of self-steepening, which translates as a depletion (replenishment) of the pulse envelope on its front (trailing edge). Furthermore, considering the time-dependent intensity in the FWM formalism reproduces this frequency shift, i.e., this transient energy storage in the medium. Our results straightforwardly generalize to nondegenerate FWM and cross-phase modulation, although the calculation is slightly more tedious. We also note that we did not consider group-velocity dispersion. Taking it into account would affect FWM via the phase-matching conditions, and equivalently SPM via the envelope deformation, without impact on our argument.

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