Unified kinetic formulation of incoherent waves propagating in nonlinear media with noninstantaneous response

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This article presents a unified kinetic formulation of partially coherent nonlinear optical waves propagating in a noninstantaneous response Kerr medium. We derive a kinetic equation that combines the weak Langmuir turbulence kinetic equation and a Vlasov-like equation within a general framework: It describes the evolution of the spectrum of a random field that exhibits a quasistationary statistics in the presence of a noninstantaneous nonlinear response. The kinetic equation sheds new light on the dynamics of partially coherent nonlinear waves and allows for a qualitative interpretation of the interplay between the noninstantaneous nonlinearity and the nonstationary statistics of the incoherent field. It is shown that the incoherent modulational instability of a random nonlinear wave can be suppressed by the noninstantaneous nonlinear response. Moreover, incoherent modulational instability can prevent the generation of spectral incoherent solitons.

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I. INTRODUCTION

The nonlinear propagation of coherent optical fields has been explored in the framework of nonlinear optics [1,2], while the linear propagation of incoherent fields has been studied in the framework of statistical optics [3]. However, these two fundamental fields of optics have been mostly developed independently, so that a complete and satisfactory understanding of statistical nonlinear optics is still lacking.

The dynamics of partially coherent nonlinear optical beams has received a renewed interest since the first experimental demonstration of incoherent optical solitons in both noninstantaneous [4,5] and instantaneous [6] response nonlinear media. The remarkable simplicity of experiments performed in photorefractive media has allowed for a fruitful investigation of the dynamics of incoherent nonlinear waves [2], as witnessed by several important achievements, such as the modulational instability (MI) of incoherent optical waves [7,8]. A notable progress has been also accomplished by exploiting the analogy with nonlinear plasma phenomena, such as the Landau damping [9] or the bump-on-tail instability [10]. Actually, it is in the context of plasma physics that random phase solitons and incoherent MI were identified in the framework of pioneering studies of *Vlasov-like kinetic equations* [11–14].

Several theoretical approaches have been developed to provide a description of incoherent optical solitons [2]. The most established methods are the mutual coherence function approach [15], the self-consistent multimode theory [16], the coherent density method [17], and the Wigner transform approach [9]. It has been shown that these four methods are in fact equivalent [18] and the choice of the most suitable representation rather depends on the nature of the physical problem to be investigated. As a matter of fact, these theoretical approaches find their origins in Vlasov-like kinetic equations, whose self-consistent mathematical structure is the key property underlying the existence of incoherent soliton solutions [9,11].

More recently, an incoherent optical soliton of a fundamentally different nature has been identified in an optical fiber system by exploiting the noninstantaneous character of the nonlinear Raman effect [19,20]. This incoherent structure has been called a spectral incoherent soliton because the optical field does not exhibit a confinement in the spatiotemporal domain, but rather exclusively in the frequency domain. More specifically, the optical field exhibits a stationary statistics (i.e., the field exhibits random fluctuations that are statistically stationary in time), and the soliton behavior only manifests in the spectral domain. The analysis has revealed that the kinetic equation that describes spectral incoherent solitons has a rather simple structure, which was considered in plasma physics to study *weak Langmuir turbulence* or stimulated Compton scattering [21–24].

Our aim in this article is to provide a unified kinetic formulation of optical wave propagation in a one-dimensional nonlinear medium whose noninstantaneous response cannot be neglected. We derive a kinetic equation that combines the weak Langmuir turbulence kinetic equation and a Vlasov-like equation within a general framework; that is, it describes the propagation of an optical field that exhibits a quasistationary statistics in the presence of a noninstantaneous nonlinear response. The analysis is based on a separation of scales technique which is valid when the characteristic time of the random fluctuations of the field is much smaller than the characteristic time of variations of the averaged power of the field. The kinetic equation that we obtain sheds new light on the understanding of the dynamics of partially coherent nonlinear waves. It allows us to interpret qualitatively some remarkable results obtained by solving numerically the generalized nonlinear Schrödinger (NLS) equation. We show in particular that the incoherent MI described by the Vlasov-like term can be suppressed by the spectral red-shift induced by the Langmuir-like term. Reciprocally, the process of incoherent MI can prevent the generation of spectral incoherent Langmuir-like solitons. We show in this way that the dynamics induced by the delayed nonlinear response and the nonstationary statistics cannot be studied separately in general and that their interplay can lead to unexpected dynamical behaviors of incoherent nonlinear waves.

We remark that the weak turbulence Vlasov-Langmuir-like kinetic equation is formally reversible, a feature which is consistent with the fact that it conserves the nonequilibrium entropy. Accordingly, this kinetic equation does not describe the process of irreversible evolution toward thermodynamic equilibrium. As a matter of fact, the process of optical wave thermalization [20,25–27] is usually described in the theoretical framework of the wave turbulence theory [28], whose kinetic equation was originally derived by Hasselmann [29]. This theory implicitly assumes that the random field exhibits a *stationary or homogeneous* statistics. It turns out that the causality condition inherent to the delayed nonlinear response and, on the other hand, the nonstationary statistics of the field both prevent the process of optical wave thermalization from taking place.

Let us note that this work can also find applications in the context of supercontinuum (SC) generation in photonic crystal fibers [30], a process which is known to be characterized by a dramatic spectral broadening of the optical field during its propagation. The interpretation of the mechanisms underlying SC generation, although generally well understood, constitutes a difficult problem due to the multitude of nonlinear effects involved. We note in this respect that a kinetic description of SC generation has recently been formulated [26], and, in particular, Langmuir-like spectral incoherent solitons have been experimentally identified in the SC generation process [20].

II. MODEL EQUATION

Let us consider the NLS equation governing the evolution of an optical field $\psi(z,t)$ that propagates in a Kerr medium characterized by a nonlinear response function $\chi(t)$:

$$i\partial_z\psi = -\beta\partial_{tt}\psi + \gamma\psi\int_{-\infty}^{+\infty}\chi(\theta)\,|\psi|^2(z,t-\theta)\,d\theta. \quad (1)$$

As usual in optics, the distance z of propagation in the nonlinear medium plays the role of an evolution variable for the NLS Eq. (1), while t measures the time in a reference frame moving at the group velocity of the field [1,2]. The parameter γ denotes the nonlinear Kerr coefficient and $\beta = \frac{1}{2}\partial^2 k/\partial\omega^2$ is the dispersion parameter, k being the wave vector modulus [2]. The linear dispersion relation of the field reads $k(\omega) = \beta\omega^2$.

For the sake of generality, we consider in the following a response function that can be decomposed into the sum of an instantaneous and a delayed contribution,

$$\chi(t) = (1 - f_R)\delta(t) + f_R R(t). \quad (2)$$

The coefficient $f_R \in [0,1]$ expresses the ratio between the two contributions. The function $R(t)$ is normalized in such a way that $\int R(t)dt = 1$ (so that we have $\int \chi(t)dt = 1$ whatever f_R is) and the causality condition imposes $R(t) = 0$ for $t < 0$. According to the linear response theory, the causality condition imposes restrictions on the Fourier transform of the response function

$$\widetilde{R}(\omega) = \int R(t)\exp(-i\omega t)dt.$$

Because of the causality of $R(t)$, the function $\widetilde{R}(\omega)$ is analytic in the lower half-plane $\mathrm{Im}(\omega) < 0$, so that the real and imaginary parts of $\widetilde{R}(\omega) = \widetilde{R}_{\mathrm{r}}(\omega) + i\widetilde{R}_{\mathrm{i}}(\omega)$ turn out to be related by the Kramers-Krönig relations, $\widetilde{R}_{\mathrm{r}}(\omega) = -\frac{1}{\pi}\mathcal{P}\int\frac{\widetilde{R}_{\mathrm{i}}(\omega')}{\omega'-\omega}d\omega'$ and $\widetilde{R}_{\mathrm{i}}(\omega) = \frac{1}{\pi}\mathcal{P}\int\frac{\widetilde{R}_{\mathrm{r}}(\omega')}{\omega'-\omega}d\omega'$, where $\mathcal{P}$ denotes the principal Cauchy value [1,31]. We recall that $\widetilde{R}_{\mathrm{r}}(\omega)$ is an even function, while $\widetilde{R}_{\mathrm{i}}(\omega)$ is an odd function. The decomposition (2) finds a direct application in optical fiber systems, which are known to exhibit both an instantaneous electronic contribution and a noninstantaneous molecular Raman contribution [2]. Note however that the model (1) only describes the forward Raman scattering effect, but neglects the Raman backscattering. This assumption is justified in the so-called "pulsed regime," in which the considered optical pulses are typically shorter than a nanosecond [2]. In this regime the short duration of the pulse prevents a significant amplification of the backscattered wave. In other terms, the backward Raman effect does not have sufficient time to enter the stimulated regime and its influence is therefore negligible. Furthermore, we remark that the model (1) has a form analogous to the so-called "Zakharov equations" used to describe Langmuir waves in the context of plasma physics [32]. In the limit of a narrow resonance, one recovers the stimulated Brillouin scattering effect, in which the electric field is coupled to an ion-sound wave equation. This collective and nonlocal wave-like response cannot be described by the "local" response function $R(t)$ considered in Eq. (1). Conversely, in the opposite limit of a broad resonance, one has the so-called "induced scattering on ions," in which Langmuir waves play a role analogous to Raman molecular vibrations in the context of nonlinear optics. We also note that the NLS Eq. (1) for $f_R > 0$ only conserves the total power of the field

$$\mathcal{N} = \int|\psi|^2\,dt. \quad (3)$$

The evolution of the random field is characterized by the nonlinear length $L_{\mathrm{nl}} = 1/(\gamma\langle|\psi|^2\rangle)$ and by the linear dispersion length $L_d = t_c^2/\beta$, where t_c is the coherence time of the field and $\langle|\psi|^2\rangle$ is the typical averaged power. In the following we consider the highly incoherent regime of interaction, $\rho = L_d/L_{\mathrm{nl}} \ll 1$, where the rapid temporal fluctuations of the field make linear effects dominant with respect to nonlinear effects.

III. DERIVATION OF THE WEAK TURBULENCE VLASOV-LANGMUIR-LIKE KINETIC EQUATION

We follow the usual procedure to derive an equation describing the evolution of the autocorrelation function $C(z,t_1,t_2) = \langle\psi(z,t_1)\psi^*(z,t_2)\rangle$,

$$i\partial_z C = \beta(\partial_{t_2}^2 - \partial_{t_1}^2)C + \gamma\int\chi(\theta)[\langle\psi(t_1)\psi^*(t_2)\psi(t_1-\theta) \times\psi^*(t_1-\theta)\rangle - \langle\psi(t_1)\psi^*(t_2)\psi(t_2-\theta)\psi^*(t_2-\theta)\rangle], \quad (4)$$

where we omitted to write the z label in the integrand. Because of the nonlinear character of the NLS equation, the evolution of the second-order moment of the field depends on the fourth-order moment. In the same way, the equation for the fourth-order moment depends on the sixth-order moment, and so on. A simple way to achieve a closure of the infinite hierarchy of moment equations is to assume that the field has Gaussian statistics. This approximation is justified in the weakly nonlinear regime, $\rho = L_d/L_{\rm nl} \ll 1$ [12,21]. Under these conditions, one can exploit the property of factorizability of moments of Gaussian fields, for example, $\langle \psi(t_1)\psi^*(t_2)\psi(t_1-\theta)\psi^*(t_1-\theta)\rangle = C(t_1,t_2)C(t_1-\theta,t_1-\theta) + C(t_1,t_1-\theta)C(t_1-\theta,t_2)$.

Introducing the change of variables $t = (t_1+t_2)/2$ and $\tau = t_1 - t_2$, we obtain a closed equation for the evolution of the second-order moment

$$B(z,t,\tau) = C(z,t+\tau/2,t-\tau/2) = \langle \psi(z,t+\tau/2)\psi^*(z,t-\tau/2)\rangle$$

that has the form

$$i\partial_z B(t,\tau) = -2\beta\partial^2_{t\tau}B(t,\tau) + \gamma P(t,\tau) + \gamma Q(t,\tau), \quad (5)$$

where we have omitted the z label and we have denoted

$$P(t,\tau) = B(t,\tau)\int \chi(\theta)[N(t-\theta+\tau/2) - N(t-\theta-\tau/2)]d\theta, \quad (6)$$

$$Q(t,\tau) = \int \chi(\theta)[B(t-\theta/2+\tau/2,\theta)B(t-\theta/2,\tau-\theta) - B(t-\theta/2,\tau+\theta)B(t-\theta/2-\tau/2,-\theta)]d\theta, \quad (7)$$

and

$$N(z,t) \equiv B(z,t,0) = \langle |\psi(z,t)|^2\rangle \quad (8)$$

denotes the averaged power of the field, which depends on time t because the statistics of the field is *a priori* nonstationary.

On the one hand we can remark that in the limit of an instantaneous response, that is, $f_R = 0$, we have $P = Q$ and Eqs. (5)–(7) recover the well-known equation for the mutual coherence function [15]:

$$i\partial_z B(t,\tau) = -2\beta\partial^2_{t\tau}B(t,\tau) + 2\gamma B(t,\tau) \times [N(t+\tau/2) - N(t-\tau/2)]. \quad (9)$$

Under the assumption of a quasistationary statistics, $N(t+\tau/2) - N(t-\tau/2) \simeq \tau\partial_t N(t)$, a Fourier transform of Eq. (9) leads to the Vlasov-like kinetic equation. Note that there is a factor 2 in front of the nonlinear term in Eq. (9), a feature that is discussed in Sec. IV D (see also Ref. [8]).

On the other hand, in the limit of a stationary statistics, the instantaneous contribution of the nonlinear response no longer contributes to the kinetic equation ($P = 0$), and Eqs. (5)–(7) can be reduced to

$$i\partial_z B(\tau) = \gamma f_R \int R(\theta)[B(\theta)B(\tau-\theta) - B^*(\theta)B(\tau+\theta)]d\theta, \quad (10)$$

where the autocorrelation function B only depends on the time lag τ. A Fourier transform of Eq. (10) readily gives the weak Langmuir turbulence kinetic equation. Our aim in the following is to derive a kinetic equation that generalizes the weak Langmuir turbulence equation and the Vlasov-like kinetic equation.

Equations (5)–(7) are quite involved. To provide an insight into the physics of Eqs. (5)–(7), we shall assume that the optical field exhibits initially a quasistationary statistics. We introduce the small parameter ε, which is the ratio between the coherence time of the initial field (i.e., the time scale of the random fluctuations) and the time scale of variation of the power of the field (i.e., the duration of the incoherent pulse), $\varepsilon = t_c/t_p$. The autocorrelation function at $z = 0$ can then be written in the form

$$B(z=0,t,\tau) = B^{(0)}(z=0,\varepsilon t,\tau)$$

and we look for the solution of Eq. (5) in the form

$$B(z,t,\tau) = B^{(0)}(\varepsilon z,\varepsilon t,\tau) + \varepsilon B^{(1)}(\varepsilon z,\varepsilon t,\tau) + \cdots. \quad (11)$$

The fact that evolution variable is scaled as εz follows from the forthcoming analysis, in which it is shown that effects of order one can be observed for propagation distances z of the order of ε^{-1}. It turns out that different regimes can be obtained, depending on the ratio f_R between the delayed and the instantaneous contributions to the nonlinear response function $\chi(t)$. The most interesting regime happens when f_R is of the order of ε, since then the two contributions are of the same order in the kinetic equation. We therefore denote

$$f_R = \varepsilon f_{R0}. \quad (12)$$

We substitute the ansatz (11) into (5) and collect the terms with the same powers in ε. It proves convenient to write the kinetic equation for the local spectrum of the field, defined as a Fourier (Wigner-like) transform of the autocorrelation function,

$$n^{(0)}_\omega(Z,T) = \int B^{(0)}(Z,T,\tau)\exp(-i\omega\tau)d\tau.$$

In the Appendix we show that $n^{(0)}_\omega(Z,T)$ is ruled by the following weak turbulence Vlasov-Langmuir-like kinetic equation:

$$\partial_Z n^{(0)}_\omega(Z,T) + \partial_\omega \kappa^{(0)}_\omega(Z,T)\partial_T n^{(0)}_\omega(Z,T) - \partial_T\kappa^{(0)}_\omega(Z,T)\partial_\omega n^{(0)}_\omega(Z,T) = \frac{\gamma f_{R0}}{\pi} n^{(0)}_\omega(Z,T)\int \widetilde{R}_{\rm i}(\omega-\omega')n^{(0)}_{\omega'}(Z,T)d\omega'. \quad (13)$$

The generalized dispersion relation reads

$$\kappa^{(0)}_\omega(Z,T) = k(\omega) + V^{(0)}(Z,T), \quad (14)$$

with the effective potential

$$V^{(0)}(Z,T) = \frac{\gamma}{\pi}\int n^{(0)}_{\omega'}(Z,T)d\omega'. \quad (15)$$

Let us briefly address the degenerate cases in which f_R is not of the form (12):

If f_R is smaller than (12), that is, $f_R = \varepsilon^p f_{R0}$ with $p > 1$, then the collision term of the right side of Eq. (13) vanishes and we recover the Vlasov limit. This means that, in the first-order approximation in ε, the noninstantaneous character of the nonlinearity does not affect the evolution of the incoherent wave.

If f_R is larger than (12), that is, $f_R = \varepsilon^p f_{R0}$ with $p < 1$, then the collision term of the right side is dominant and we recover the weak Langmuir turbulence kinetic equation; that is, the nonstationary statistics does not affect the dynamics of the incoherent field. The "Vlasov" and "Langmuir" limits of Eq. (13) are discussed in Sec. IV.

In the particular case (12), if we push the expansion to the second order in ε and consider

$$n_\omega^{(\varepsilon)}(Z,T)=\int[B^{(0)}(Z,T,\tau)+\varepsilon B^{(1)}(Z,T,\tau)]\exp(-i\omega\tau)d\tau,$$

we obtain the following generalized Vlasov-Langmuir-like kinetic equation for $n_\omega^{(\varepsilon)}(Z,T)$ (see the Appendix):

$$\begin{aligned}\partial_Z n_\omega^{(\varepsilon)}(Z,T)&+\partial_\omega\kappa_\omega^{(\varepsilon)}(Z,T)\partial_T n_\omega^{(\varepsilon)}(Z,T)\\&-\partial_T\kappa_\omega^{(\varepsilon)}(Z,T)\partial_\omega n_\omega^{(\varepsilon)}(Z,T)\\&=\frac{\gamma f_{R0}}{\pi}n_\omega^{(\varepsilon)}(Z,T)\int\widetilde{R}_{\rm i}(\omega-\omega')n_{\omega'}^{(\varepsilon)}(Z,T)d\omega',\end{aligned}\tag{16}$$

with the effective dispersion relation and the effective potential

$$\kappa_\omega^{(\varepsilon)}(Z,T)=k(\omega)+V_\omega^{(\varepsilon)}(Z,T),\tag{17}$$

$$\begin{aligned}V_\omega^{(\varepsilon)}(Z,T)=&\frac{\gamma(2-f_{R0}\varepsilon)}{2\pi}\int n_{\omega'}^{(\varepsilon)}(Z,T)d\omega'\\&+\frac{\varepsilon\gamma f_{R0}}{2\pi}\int\widetilde{R}_{\rm r}(\omega-\omega')n_{\omega'}^{(\varepsilon)}(Z,T)d\omega'.\end{aligned}\tag{18}$$

Let us remark that the effective potential $V_\omega^{(\varepsilon)}(Z,T)$ now involves a convolution with the real part of the Fourier transform of the response function, $\widetilde{R}_{\rm r}(\omega)$, so that $V_\omega^{(\varepsilon)}(Z,T)$ now depends on the frequency ω. Then contrarily to the conventional Vlasov-like equation [see Eqs. (13)–(15)], the effective dispersion relation $\kappa_\omega^{(\varepsilon)}(Z,T)$ no longer splits into the sum of a t-dependent and a ω-dependent contribution. Note that the kinetic equations (13) and (16) have the same structure as the *inhomogeneous* weak Langmuir turbulence kinetic equation discussed in Refs. [12,21]. Let us remark, however, that the mean-field potential $V_\omega(z,t)$ involved in the dispersion relation considered in Refs. [12,21] differs substantially from the mean-field potentials obtained here.

IV. PROPERTIES OF THE VLASOV-LANGMUIR KINETIC EQUATION

A. Vlasov limit

As discussed previously, when the delayed response of the nonlinearity is not relevant, the kinetic equation (13) recovers the Vlasov-like equation,

$$\partial_z n_\omega(z,t)+\partial_\omega\kappa_\omega(z,t)\partial_t n_\omega(z,t)-\partial_t\kappa_\omega(z,t)\partial_\omega n_\omega(z,t)=0,\tag{19}$$

with the generalized dispersion relation

$$\kappa_\omega(t)=k(\omega)+2\gamma N(z,t),\tag{20}$$

where we recall that $N(z,t)=\frac{1}{2\pi}\int n_\omega(z,t)d\omega$.

An important phenomenon described by Eq. (19) is the MI of partially coherent waves. Incoherent MI has been the subject of a recent detailed investigation in the context of optical waves, from both the theoretical and the experimental points of view [2,7]. In the following we briefly recall some salient aspects of incoherent MI that will be used to analyze the properties of the Vlasov-Langmuir kinetic Eqs. (13) and (16) in Sec. V. In the temporal domain, an incoherent field that exhibits a stationary statistics can become modulationally unstable in the presence of anomalous dispersion, $\beta<0$. Let us recall that any statistical stationary distribution $n_0(\omega)$ is a solution of the Vlasov Eqs. (19) and (20), that is, $\partial_z n_0(\omega)=0$. Modulational instability is thus studied by perturbing such a z-invariant solution with small noise, $n_\omega(z,t)=n_0(\omega)+\delta n_\omega(z,t)$. An analytical expression of the incoherent MI gain can be obtained under the assumption of an initial Lorentzian spectrum [7,12],

$$n_0(\omega)=\frac{2\sigma N}{\sigma^2+\omega^2},$$

where σ represents the characteristic spectral width of the Lorentzian. Linearizing the Vlasov Eqs. (19) and (20) with respect to $\delta n_\omega(z,t)$ yields the MI growth rate for a modulation at the frequency α,

$$\lambda(\alpha)=-2|\beta\alpha|\sigma+2|\alpha|\sqrt{\gamma N|\beta|},\tag{21}$$

in which we implicitly assume $\beta<0$. Note that this expression of the MI gain corresponds to the low-frequency expansion of the corresponding gain derived from the equation for the mutual coherence function $B(z,t,\tau)$ [Eq. (9)], that is, $\lambda_B(\alpha)=-2|\beta\alpha|\sigma+|\alpha|\sqrt{4\gamma N|\beta|-\alpha^2\beta^2}$. This is consistent with the fact that the Vlasov Eq. (19) corresponds to the first-order correction of the nonstationary statistics of Eq. (9). Accordingly, the Vlasov MI gain (21) only provides the low-frequency slope of the whole gain curve $\lambda_B(\alpha)$, and for this reason it does not exhibits a frequency cutoff; that is, the cutoff frequency goes to infinity as the scaling parameter $\varepsilon=t_c/t_p\to 0$.

We also recall that, contrarily to the usual MI induced by a coherent field, wave incoherence can suppress MI [2,7,8,12,14]. The threshold for incoherent MI is usually defined from the MI-gain slope at the origin, $\partial_\alpha\lambda|_{\alpha=0}>0$. The Vlasov expression (21) thus correctly predicts the MI threshold, that is, $\sigma<\sqrt{\gamma N/|\beta|}$, which indicates that MI is suppressed when the bandwidth of the spectrum becomes comparable to the MI frequency. Let us note that these expressions of incoherent MI gain and MI threshold slightly differ from those usually reported in Ref. [7] for an "inertial" nonlinearity, that is, a nonlinearity whose response time is much larger than the coherence time of the optical field (see Ref. [8]). This aspect is discussed in Sec. IV D.

B. Weak Langmuir turbulence limit

We have seen in Sec. III that when the nonstationarity of the statistics is not relevant, Eq. (13) can be reduced to the weak Langmuir turbulence kinetic equation

$$\partial_z n_\omega(z)=\frac{\gamma f_{R0}}{\pi}n_\omega(z)\int\widetilde{R}_{\rm i}(\omega-\omega')n_{\omega'}(z)d\omega'.\tag{22}$$

Several simplified forms of this kinetic equation have been the subject of a detailed study in the literature. A differential ("hydrodynamic") approximation of the integrodifferential equation (22) was derived for the first time by Kompaneets [33]. This Compton Fokker-Planck equation has been subsequently analyzed by several authors [34]. The complete integral kinetic equation (22) may be derived from the Zakharov equations [32]; it can also be derived from the quantum version of the Boltzmann-like kinetic equation describing the nonlinear induced Compton scattering [35].

A peculiar property of the weak Langmuir turbulence equation (22) is that it admits solitary wave solutions [21–23]. This fact can be anticipated by remarking that, as a result of the convolution product in Eq. (22), the spectral gain curve $\widetilde{R}_{\rm i}(\omega)$ amplifies the front of the spectrum at the expense of its trailing edge, thus leading to a global red-shift of $n_\omega(z)$. The numerical simulations of the NLS Eq. (1) and of the Langmuir-like Eq. (22) reveal that, after a transient regime, the averaged spectrum of the field self-organizes in the form of a solitary wave, which propagates without distortion in the frequency domain toward the low-frequency components [19,21–23]. Because the statistics of the field is stationary, the soliton behavior manifests itself in the spectral domain, but not in the temporal domain.

A spectral soliton can be generated in the presence of a background, $n_\omega \to n_\infty > 0$ as $|\omega| \to \pm\infty$ [21–23]. The weak Langmuir turbulence Eq. (22) verifies the following conservation relations:

$$\partial_z\left[\int \ln\left(\frac{n_\omega}{n_\infty}\right)d\omega\right] = 0,$$

$$\partial_z\left[\int \left(\frac{n_\omega}{n_\infty} - 1\right)d\omega\right] = 0,$$

$$\partial_z\left[\int \omega \ln\left(\frac{n_\omega}{n_\infty}\right)d\omega\right] = \left[\frac{\gamma}{\pi}\int \omega \widetilde{R}_{\rm i}(\omega)d\omega\right]\left[\int \left(\frac{n_\omega}{n_\infty} - 1\right)d\omega\right].$$

The first and second equations express the conservation of the nonequilibrium entropy and of the total power of the field, respectively. The third relation can be used to compute the soliton velocity V. Indeed, if n_ω is a spectral soliton with center frequency $\Omega(z)$, the third relation gives the constant velocity $V \equiv \partial_z \Omega(z)$ in terms of the constant profile:

$$V = \frac{\int \left(\frac{n_\omega}{n_\infty} - 1\right)d\omega}{\int \ln\left(\frac{n_\omega}{n_\infty}\right)d\omega}\left[\frac{\gamma}{\pi}\int \omega \widetilde{R}_{\rm i}(\omega)d\omega\right]. \qquad (23)$$

Note that $\ln(n_\omega/n_\infty) \simeq (n_\omega/n_\infty - 1) - \frac{1}{2}(n_\omega/n_\infty - 1)^2 + \mathcal{O}[(n_\omega/n_\infty - 1)^3]$. Therefore, if the profile has a small amplitude, Eq. (23) reduces to $V = \frac{\gamma}{\pi}\int \omega \widetilde{R}_{\rm i}(\omega)d\omega$, which corresponds to the Korteweg–de-Vries limit of the soliton velocity [21]. Formula (23) also shows that the velocity increases with the amplitude of the wave.

It is possible to compute the width and velocity of the soliton given its peak amplitude n_m in the regime $n_m \gg n_\infty$. This was done in Ref. [23] for the particular case where the gain spectrum $\widetilde{R}_{\rm i}(\omega)$ is the derivative of a Gaussian. In the following we generalize the procedure of Ref. [23] for a generic gain spectrum $\widetilde{R}_{\rm i}(\omega)$. For this purpose, let us introduce the antiderivative of the MI spectrum:

$$G(\omega) = -\int_\omega^\infty \widetilde{R}_{\rm i}(\omega')d\omega'. \qquad (24)$$

The gain spectrum $\widetilde{R}_{\rm i}(\omega)$ is characterized by its typical gain amplitude $g_{\rm i}$ and its typical spectral width $\omega_{\rm i}$. Regardless of the details of the gain curve $\widetilde{R}_{\rm i}(\omega)$, $g_{\rm i}$ and $\omega_{\rm i}$ can be assessed by two characteristic quantities, namely, the gain slope at the origin $\partial_\omega \widetilde{R}_{\rm i}(0)$ and the total amount of gain $G(0) = -\int_0^\infty \widetilde{R}_{\rm i}(\omega)d\omega$. A dimensional analysis allows us to express $g_{\rm i}$ and $\omega_{\rm i}$ in terms of these two quantities:

$$g_{\rm i} = \frac{1}{\sqrt{2}}[-\partial_\omega \widetilde{R}_{\rm i}(0)]^{1/2}\left[-\int_0^\infty \widetilde{R}_{\rm i}(\omega)d\omega\right]^{1/2},$$
$$\omega_{\rm i} = \sqrt{2}\frac{\left[-\int_0^\infty \widetilde{R}_{\rm i}(\omega)d\omega\right]^{1/2}}{[-\partial_\omega \widetilde{R}_{\rm i}(0)]^{1/2}}. \qquad (25)$$

With these definitions, the function $G(\omega)$ can be written in the following normalized form

$$G(\omega) = g_{\rm i}\omega_{\rm i} h\left(\frac{\omega}{\omega_{\rm i}}\right),$$

where the dimensionless function $h(x)$ verifies $h(0) = 1$, $h'(0) = 0$, and $h''(0) = -2$. Proceeding as in [23], we find that the profile of the soliton in the regime $n_m \gg n_\infty$ is of the form

$$\ln\left(\frac{n_\omega(z)}{n_\infty}\right) = \ln\left(\frac{n_m}{n_\infty}\right) h\left(\frac{\omega - Vz}{\omega_{\rm i}}\right),$$
$$n_\omega(z) - n_\infty = (n_m - n_\infty)\exp\left[-\ln\left(\frac{n_m}{n_\infty}\right)\frac{(\omega - Vz)^2}{\omega_{\rm i}^2}\right], \qquad (26)$$

where the velocity of the soliton is

$$V = -\frac{n_m - n_\infty}{\ln^{3/2}\left(\frac{n_m}{n_\infty}\right)}\frac{\gamma g_{\rm i}\omega_{\rm i}^2}{\sqrt{\pi}}, \qquad (27)$$

and its full width at half maximum is

$$\omega_{\rm sol} = \frac{2\ln^{1/2} 2}{\ln^{1/2}\left(\frac{n_m}{n_\infty}\right)}\omega_{\rm i}. \qquad (28)$$

This soliton solution is valid, in principle, for any gain spectrum $\widetilde{R}_{\rm i}(\omega)$. It generalizes the solution found in Ref. [23] for the particular case where the gain spectrum $\widetilde{R}_{\rm i}(\omega)$ is the derivative of a Gaussian.

C. Reversibility of the Vlasov-Langmuir kinetic equation

The Vlasov-like equation and the weak Langmuir turbulence equation both conserve the total power (quasiparticle number) of the optical field, $\mathcal{N} = (2\pi)^{-1}\int\int n_\omega(z,t)d\omega dt$. These equations are also known to conserve the nonequilibrium entropy,

$$\mathcal{S} = \frac{1}{2\pi}\int\int \ln[n_\omega(z,t)]d\omega dt. \qquad (29)$$

Let us show that the Vlasov-Langmuir-like kinetic Eq. (13) or (16) also conserves $\mathcal{S}$. This is obvious for (13) since the dispersion relation (14) for $\kappa_\omega^{(0)}(t)$ splits into the sum of a t-dependent and an ω-dependent contribution, as it occurs for the Vlasov equation. However, this is not the case for the generalized dispersion relation $\kappa_\omega^{(\varepsilon)}(t)$ [Eq. (17)] associated to the Vlasov-Langmuir Eq. (16). To show that Eq. (16) conserves

$\mathcal{S}$, one can simply write

$$\partial_z \mathcal{S} = \frac{1}{2\pi}\iint \partial_t \kappa \, \partial_\omega \ln(n) d\omega \, dt - \frac{1}{2\pi}\iint \partial_\omega \kappa \, \partial_t \ln(n) d\omega \, dt. \tag{30}$$

Integrating by parts the first (second) term with respect to t (ω), the two terms cancel each other and $\partial_z \mathcal{S} = 0$.

The conservation of the nonequilibrium entropy (29) is consistent with the fact that the *Vlasov-Langmuir kinetic Eqs. (13) and (16) are formally reversible*; that is, they are invariant under the transformation $(z, \omega, t) \to (-z, -\omega, t)$. Note that the requirement of the sign inversion in ω can be understood by analogy with kinetic gas theory, where time reversal needs the inversion of the velocities of the particles, $(t, \boldsymbol{k}, \boldsymbol{x}) \to (-t, -\boldsymbol{k}, \boldsymbol{x})$. Accordingly, the Vlasov-Langmuir kinetic Eq. (13) or (16) does not describe an irreversible evolution of the optical field to thermodynamic equilibrium.

The essential properties of optical wave thermalization to equilibrium are described by the wave turbulence theory [28]. The derivation of the wave turbulence kinetic equation requires a second-order closure of the hierarchy of moments equations, that is, a second-order perturbation expansion in $\rho = L_d/L_{\rm nl}$ [28]. In this framework, one obtains a kinetic equation whose collision term exhibits a H theorem of entropy growth, $d_z\mathcal{S} \geqslant 0$, where $\mathcal{S}$ refers to the nonequilibrium entropy (29). This kinetic equation has a structure analogous to the Boltzmann equation and, by analogy with kinetic gas theory, it describes an irreversible evolution of the wave to the thermodynamic equilibrium distribution, that is, the Rayleigh-Jeans spectrum [28]. More precisely, the wave turbulence theory assumes that the random field exhibits a stationary (or homogeneous) statistics. But this is not sufficient, since the noninstantaneous nonlinearity leads to the weak Langmuir turbulence equation in the first-order approximation in ρ, and as discussed previously, this equation does not describe wave thermalization. Actually, it is the causality property inherent to the nonlinear response function $R(t)$ which prevents the thermalization process to occur. This becomes apparent by remarking that Eq. (1) is almost identical to the NLS equation governing wave propagation in a *nonlocal* nonlinear medium [2], provided one substitutes the response function with the nonlocal potential, that is, $i\partial_z\psi = -\alpha\partial_{xx}\psi + \psi\int_{-\infty}^{+\infty} V(x - x')|\psi|^2(x')dx'$. However, nonlocal effects are not constrained by the causality condition. Moreover, under the assumption of spatial homogeneity, $V(x)$ is an even function of x. Its Fourier transform $\widetilde{V}(k)$ is thus purely real and the weak Langmuir turbulence equation (22) reduces to the trivial equation $\partial_z n(z, \omega) = 0$. Accordingly, the kinetic description of a nonlocal interaction requires a second-order perturbation expansion in ρ, and the corresponding kinetic equation exhibits the usual H theorem of entropy growth, which describes the irreversible thermalization process. This discussion reveals that (in the first-order approximation in ρ), both the noninstantaneous nonlinear response and the nonstationary statistics prevent the thermalization of the nonlinear wave from taking place.

D. The limit of an "inertial" nonlinearity

Let us finally discuss the limit of an "inertial" nonlinearity, that is, the limit in which the response time of the nonlinearity τ_R is much larger than the coherence time of the incoherent field, $t_c \ll \tau_R$. In this limit, the convolution product into the NLS Eq. (1) can be approximated by an averaged nonlinearity, $\int \chi(\theta)\, |\psi|^2(z, t - \theta)\, d\theta \simeq \langle|\psi|^2\rangle = N(z, t)$. This means that the nonlinear medium responds to the averaged intensity, but not to the individual fluctuations (speckles) of the optical field. The evolution of the field amplitude is thus governed by an averaged NLS equation,

$$i\partial_z\psi(t) = -\beta\partial_{tt}\psi(t) + \gamma N(t)\psi(t). \tag{31}$$

Note that, because of the "inertial" character of the nonlinearity, Gaussian statistics is *preserved* under the nonlinear evolution of the field [36]. It is important to underline that, in contrast to the usual NLS equation, Eq. (1), the averaged NLS equation, Eq. (31), *does not lead to an infinite hierarchy of moment equations*. Indeed, the derivation of the second-order moment equation from (31) does not require any additional assumption on the nature of the statistics of the field. Following the same procedure as that outlined at the beginning of Sec. III, one can derive the following equation without any approximation:

$$i\partial_z B(t, \tau) = -2\beta\partial_{t\tau}^2 B(t, \tau) + \gamma B(t, \tau) \times [N(t + \tau/2) - N(t - \tau/2)]. \tag{32}$$

We remark that, except for the factor 2 in front of the nonlinear term, this equation is identical to that derived in the *opposite limit of an instantaneous nonlinearity*, $t_c \gg \tau_R$, that is, the limit $f_R = 0$ [see Eq. (9)]. The corresponding kinetic equation for an "inertial" nonlinearity is thus again the Vlasov Eq. (19) without the factor 2 in the generalized dispersion relation (20), that is $\kappa_\omega(t) = k(\omega) + \gamma N(t)$. Accordingly, the value of the MI growth rate is reduced by a factor $\sim\sqrt{2}$ with respect to the corresponding value of the instantaneous nonlinearity considered in Sec. IV A, a feature that was discussed in Ref. [8]. The limit of an "inertial" nonlinearity actually corresponds to the limit usually considered experimentally in photorefractive crystals, where the dynamics of spatial incoherent MI and spatial incoherent solitons were investigated in detail [4,5,7]. Let us note in this respect that the Vlasov-Langmuir-like kinetic equations (13) and (16) derived in the temporal domain may be extended to account for the *spatiotemporal* coherence properties of the optical field (see, e.g., [37]).

V. INTERPLAY OF THE VLASOV AND LANGMUIR TERMS IN THE KINETIC EQUATION

In this section we see that a simple analysis of the Vlasov-Langmuir-like equation derived in Sec. III provides physical insight into the dynamics of partially coherent optical waves ruled by the NLS Eq. (1). In particular, the process of incoherent MI described by the Vlasov term can be highly affected by the delayed nonlinear response described by the Langmuir term. Reciprocally, the dynamics of spectral incoherent solitons can be affected by incoherent MI. In the following we illustrate these two aspects separately.

A. Incoherent MI suppression by the noninstantaneous nonlinear response

The existence of a noninstantaneous nonlinear response can change the dynamics of incoherent MI in a significant way. This fact can be anticipated by remarking that, contrarily to what happens for the Vlasov Eq. (19) (see Sec. IV A), a statistically stationary (in time) distribution is no longer a z-invariant solution of the Vlasov-Langmuir kinetic Eq. (13) or (16), that is, $\partial_z n_0(\omega) \neq 0$. Indeed, let us refer back to the incoherent MI analysis discussed in Sec. IV A, in which the z-invariant solution is perturbed as $n_\omega(z,t) = n_0(\omega) + \delta n_\omega(z,t)$. Because of the presence of the weak Langmuir turbulence collision term in the right-hand side of Eq. (13), a statistical stationary spectrum is now governed by the weak Langmuir turbulence equation, $\partial_z n_0(z,\omega) = \frac{\gamma}{\pi} n_0(z,\omega) \int \widetilde{R}_{\rm i}(\omega-\omega') n_0(z,\omega') d\omega'$. This indicates that, in the first-order approximation in δn, the incoherent field is solely governed by the weak Langmuir turbulence term and thus exhibits a spectral red-shift instead of the expected incoherent MI process. In other terms, incoherent MI can be suppressed by the noninstantaneous nonlinear response.

We illustrate this unexpected conclusion by direct numerical simulations of the NLS Eq. (1) in the anomalous dispersion regime ($\beta < 0$). For concreteness, we consider the detailed expression of the Raman response function of silica optical fibers, $R(t) = f_a h_a(t) + f_b h_b(t)$, where $h_a(t) = \tau_1(\tau_1^{-2} + \tau_2^{-2}) \exp(-t/\tau_2) \sin(t/\tau_1)$ and $h_b(t) = (2\tau_b - t)\exp(-t/\tau_b)/\tau_b^2$, with $\tau_1 = 12$ fs, $\tau_2 = 32$ fs, $\tau_b = 96$ fs, and $f_b = 0.21$ ($f_a + f_b = 1$) [2,38]. The initial condition of the field refers to a Gaussian spectrum with δ-correlated random spectral phases, that is, $\psi(z=0,t)$ has mean zero and a stationary Gaussian statistics. The Gaussian spectrum has been superposed to a small amplitude background white noise. In order to illustrate the MI suppression, parameters have been chosen in such a way that the Raman effect and the MI processes compete with each other. The optimal MI frequency has thus been taken of the same order as the maximum gain frequency of the Raman gain curve, that is, $\omega_{\rm MI} \sim \omega_{\rm i}$ [see Eq. (25)]. Figure 1 reports the spectra of the field in the presence of an instantaneous (a) and a delayed (b) response function. This numerical simulation shows that incoherent MI can be suppressed by the noninstantaneous nonlinear response: Instead of the expected incoherent MI process, we see in Fig. 1(b) that the spectrum experiences an asymmetric deformation, which is characterized by an energy transfer toward the low-frequency components. Such asymmetry of the spectrum can be naturally ascribed to the spectral red-shift induced by the Langmuir term of the kinetic Eq. (13).

B. Incoherent MI prevents spectral incoherent soliton generation

One can wonder whether weak Langmuir turbulence incoherent solitons could be affected by the incoherent MI described by the Vlasov terms in the kinetic Eq. (13). To analyze this aspect, we remark that the characteristic width of a Langmuir incoherent soliton is of the same order as the width of the spectral gain curve $\widetilde{R}_{\rm i}(\omega)$, that is, $\omega_{\rm sol} \sim \omega_{\rm i}$ [see Eq. (28)]. We can thus expect that the Vlasov terms will not affect the generation of a spectral incoherent soliton when

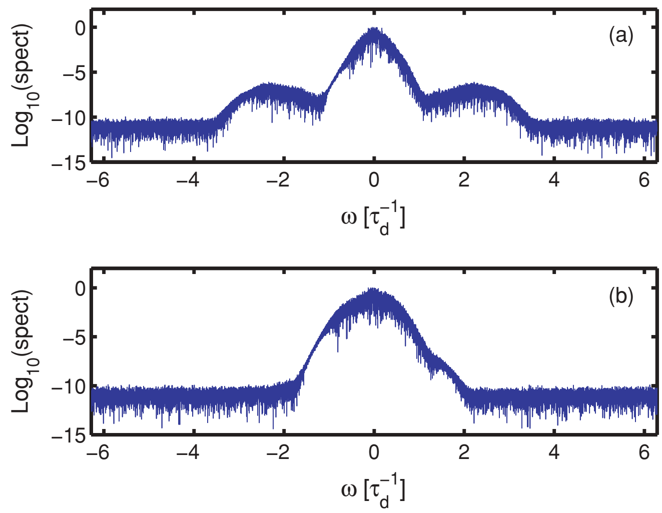

FIG. 1. (Color online) Spectrum of the field, $|\tilde{\psi}|^2(z,\omega)$ (in Log_{10} scale), obtained by integrating numerically the NLS Eq. (1) with $f_R = 0$ (a) and $f_R = 1$ (b). Panel (a) refers to $z = 1.6 L_{\rm nl}$; panel (b) refers to $z = 12.5 L_{\rm nl}$ [$\tau_d = (|\beta| L_{\rm nl})^{1/2} = 11$ fs]. The frequency ω is in units of τ_d^{-1}.

the optimal MI frequency is much smaller than the (Raman) gain frequency, $\omega_{\rm MI} \ll \omega_{\rm i} \sim \omega_{\rm sol}$, since in this case the broad spectrum of the field suppresses the MI (see Sec. IV A). Conversely, when $\omega_{\rm MI}$ becomes greater than $\omega_{\rm i}$, we can anticipate that incoherent MI will prevent the generation of the spectral incoherent soliton.

The numerical simulations of the NLS Eq. (1) confirm these expectations. A suitable initial condition to generate a spectral soliton is provided by Eq. (26). The corresponding initial spectrum of the field $\tilde{\psi}(z=0,\omega)$ thus refers to the Gaussian (26), in which we have added random spectral phases and a small-amplitude white noise. The numerical results are reported in Fig. 2, for $\omega_{\rm MI}/\omega_{\rm i} \simeq 0.015 \ll 1$ (a) and for $\omega_{\rm MI} \sim 10\omega_i$ (b).

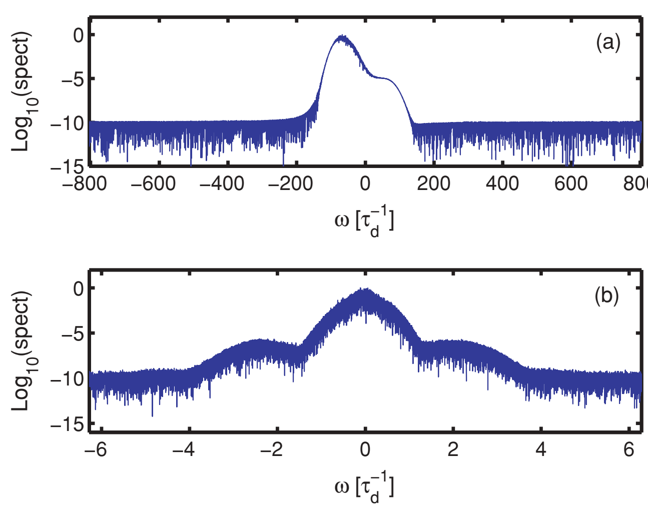

FIG. 2. (Color online) Spectrum of the field, $|\tilde{\psi}|^2(z,\omega)$ (in Log_{10} scale), obtained by integrating numerically the NLS Eq. (1) in the anomalous dispersion regime $\beta < 0$ ($f_R = 0.25$). The initial condition corresponds to Eq. (26). The spectrum of the field is plotted at $z = 25 L_{\rm nl}$ (a) ($\tau_d = 0.78$ ps) and $z = 1.7 L_{\rm nl}$ (b) ($\tau_d = 2.2$ fs). The frequency ω is in units of τ_d^{-1}.

Note that in practice one can go from configuration (a) to configuration (b) by increasing the power of the optical field. The numerical simulations reveal that, as expected, a spectral incoherent soliton is generated in Fig. 2(a), whereas in Fig. 2(b) incoherent MI prevents the generation of the soliton.

VI. CONCLUSION

In summary, we have derived a kinetic equation describing the propagation of a random field that exhibits a quasistationary statistics in the presence of a noninstantaneous nonlinear response. Note that, although the kinetic equation has been derived in one-dimension and in the temporal domain, it can easily be generalized to the spatiotemporal evolution of the field. This unified kinetic formulation combines the previously studied Vlasov-like and Langmuir-like approaches within a general framework. While the Vlasov and Langmuir dynamics have been usually studied separately, we have shown that their interplay can lead to rather unexpected results: The generation of spectral incoherent solitons can be prevented by incoherent MI, and reciprocally, incoherent MI can be suppressed by the weak Langmuir turbulence effect. A complete analysis of the Vlasov-Langmuir kinetic equation still needs to be done. In particular, it would be important to study the existence of soliton solutions of the whole Vlasov-Langmuir kinetic equations [Eqs. (13) and (16)], which would constitute a nontrivial generalization of Vlasov-like solitons [11] and weak Langmuir turbulence solitons [22]. This issue is presently under consideration.

Given the universality of the NLS equation in physics, this work can find applications in any system of random nonlinear waves, such as water, matter, or plasma waves. In the latter case, we can mention, for instance, the important issue of inertial confinement fusion for which the coherence properties of nonlinear waves are known to be essential for the ultimate control of the confinement process [39].

APPENDIX: DERIVATION OF EQS. (13) AND (16)

In this appendix we compute the expansions up to the second order in ε of the terms P and Q defined by (6) and (7) when B has the two-scale form

$$B(t,\tau) = B_0(\varepsilon t,\tau) = \frac{1}{2\pi}\int n_\omega(\varepsilon t)e^{i\omega\tau}d\tau.$$

The Fourier transform of the term P defined by (6) gives

$$\begin{aligned}\int P\left(\frac{T}{\varepsilon},\tau\right)e^{-i\omega\tau}d\tau &= \iint B_0(T,\tau)\chi(\theta)\left[B_0\left(T-\varepsilon\theta+\frac{\varepsilon\tau}{2},0\right)-B_0\left(T-\varepsilon\theta-\frac{\varepsilon\tau}{2},0\right)\right]e^{-i\omega\tau}d\theta d\tau\\ &= \frac{1}{(2\pi)^2}\iint n_{\omega_1}(T)\chi(\theta)\left[n_{\omega_2}\left(T-\varepsilon\theta+\frac{\varepsilon\tau}{2}\right)-n_{\omega_2}\left(T-\varepsilon\theta-\frac{\varepsilon\tau}{2}\right)\right]e^{-i(\omega-\omega_1)\tau}d\omega_1 d\omega_2 d\theta d\tau.\end{aligned}$$

Remember that $\chi(\theta)=(1-f_{R0}\varepsilon)\delta(\theta)+\varepsilon f_{R0}R(\theta)$. We decompose $P=P_1+P_2$, where P_1 is the contribution of the instantaneous nonlinear response and P_2 is the contribution of the delayed nonlinear response.

The contribution of the instantaneous response is

$$\int P_1\left(\frac{T}{\varepsilon},\tau\right)e^{-i\omega\tau}d\tau = \frac{(1-f_{R0}\varepsilon)}{(2\pi)^2}\iint n_{\omega_1}(T)\left[n_{\omega_2}\left(T+\frac{\varepsilon\tau}{2}\right)-n_{\omega_2}\left(T-\frac{\varepsilon\tau}{2}\right)\right]e^{-i(\omega-\omega_1)\tau}d\omega_1 d\omega_2 d\tau.$$

By expanding in ε the difference inside the integral,

$$\begin{aligned}\int P_1\left(\frac{T}{\varepsilon},\tau\right)e^{-i\omega\tau}d\tau &= \frac{\varepsilon(1-f_{R0}\varepsilon)}{(2\pi)^2}\iint n_{\omega_1}(T)\partial_T n_{\omega_2}(T)\tau e^{i(\omega_1-\omega)\tau}d\omega_1 d\omega_2 d\tau + O(\varepsilon^3)\\ &= -\frac{i\varepsilon(1-f_{R0}\varepsilon)}{(2\pi)^2}\iint n_{\omega_1}(T)\partial_T n_{\omega_2}(T)\partial_{\omega_1}[e^{i(\omega_1-\omega)\tau}]d\omega_1 d\omega_2 d\tau + O(\varepsilon^3).\end{aligned}$$

By integrating by parts in ω_1,

$$\int P_1\left(\frac{T}{\varepsilon},\tau\right)e^{-i\omega\tau}d\tau = \frac{i\varepsilon(1-f_{R0}\varepsilon)}{(2\pi)^2}\iint \partial_{\omega_1}n_{\omega_1}(T)\partial_T n_{\omega_2}(T)e^{i(\omega_1-\omega)\tau}d\omega_1 d\omega_2 d\tau + O(\varepsilon^3).$$

The integration in τ gives the Dirac distribution $\delta(\omega_1-\omega)$ and finally we obtain

$$\int P_1\left(\frac{T}{\varepsilon},\tau\right)e^{-i\omega\tau}d\tau = i\varepsilon(1-f_{R0}\varepsilon)\partial_\omega n_\omega(T)\partial_T\left[\frac{1}{2\pi}\int n_{\omega'}(T)d\omega'\right] + O(\varepsilon^3).$$

We proceed in a similar way to compute the contribution of the delayed nonlinear response:

$$\begin{aligned}\int P_2\left(\frac{T}{\varepsilon},\tau\right)e^{-i\omega\tau}d\tau &= \frac{\varepsilon^2 f_{R0}}{(2\pi)^2}\iint n_{\omega_1}(T)R(\theta)\partial_T n_{\omega_2}(T)\tau e^{i(\omega_1-\omega)\tau}d\omega_1 d\omega_2 d\theta d\tau + O(\varepsilon^3)\\ &= -\frac{\varepsilon^2 f_{R0}}{(2\pi)^2}\iint n_{\omega_1}(T)\partial_T n_{\omega_2}(T)\partial_{\omega_1}[e^{i(\omega_1-\omega)\tau}]d\omega_1 d\omega_2 d\tau + O(\varepsilon^3)\\ &= \frac{\varepsilon^2 f_{R0}}{(2\pi)^2}\iint \partial_{\omega_1}n_{\omega_1}(T)\partial_T n_{\omega_2}(T)e^{i(\omega_1-\omega)\tau}d\omega_1 d\omega_2 d\tau + O(\varepsilon^3)\\ &= i\varepsilon^2 f_{R0}\partial_\omega n_\omega(T)\partial_T\left[\frac{1}{2\pi}\int n_{\omega'}(T)d\omega'\right] + O(\varepsilon^3).\end{aligned}$$

The Fourier transform of the term Q defined by (7) gives

$$\int Q\left(\frac{T}{\varepsilon},\tau\right)e^{-i\omega\tau}d\tau = \frac{1}{(2\pi)^2}\iint \chi(\theta)\left[n_{\omega_1}\left(T-\frac{\varepsilon\theta}{2}+\frac{\varepsilon\tau}{2}\right)n_{\omega_2}\left(T-\frac{\varepsilon\theta}{2}\right)e^{i\omega_1\theta+i\omega_2(\tau-\theta)}\right.$$
$$\left.-n_{\omega_1}\left(T-\frac{\varepsilon\theta}{2}-\frac{\varepsilon\tau}{2}\right)n_{\omega_2}\left(T-\frac{\varepsilon\theta}{2}\right)e^{-i\omega_1\theta+i\omega_2(\tau+\theta)}\right]e^{-i\omega\tau}d\omega_1 d\omega_2 d\theta d\tau.$$

We decompose $Q = Q_1 + Q_2$, where Q_1 is the contribution of the instantaneous nonlinear response and Q_2 is the contribution of the delayed nonlinear response.

$$\begin{aligned}\int Q_1\left(\frac{T}{\varepsilon},\tau\right)e^{-i\omega\tau}d\tau &= \frac{(1-f_{R0}\varepsilon)}{(2\pi)^2}\iint\left[n_{\omega_1}\left(T+\frac{\varepsilon\tau}{2}\right)-n_{\omega_1}\left(T-\frac{\varepsilon\tau}{2}\right)\right]n_{\omega_2}(T)e^{i(\omega_2-\omega)\tau}d\omega_1 d\omega_2 d\tau\\
&= \frac{\varepsilon(1-f_{R0}\varepsilon)}{(2\pi)^2}\iint \partial_T n_{\omega_1}(T)n_{\omega_2}(T)\tau e^{i(\omega_2-\omega)\tau}d\omega_1 d\omega_2 d\tau + O(\varepsilon^3)\\
&= -\frac{i\varepsilon(1-f_{R0}\varepsilon)}{(2\pi)^2}\iint \partial_T n_{\omega_1}(T)n_{\omega_2}(T)\partial_{\omega_2}[e^{i(\omega_2-\omega)\tau}]d\omega_1 d\omega_2 d\tau + O(\varepsilon^3)\\
&= \frac{i\varepsilon(1-f_{R0}\varepsilon)}{(2\pi)^2}\iint \partial_T n_{\omega_1}(T)\partial_{\omega_2}n_{\omega_2}(T)e^{i(\omega_2-\omega)\tau}d\omega_1 d\omega_2 d\tau + O(\varepsilon^3)\\
&= i\varepsilon(1-f_{R0}\varepsilon)\partial_\omega n_\omega(T)\partial_T\left[\frac{1}{2\pi}\int n_{\omega'}(T)d\omega'\right] + O(\varepsilon^3).\end{aligned}$$

The contribution of the delayed nonlinear response has two nontrivial terms of order ε and ε^2, respectively,

$$\int Q_2\left(\frac{T}{\varepsilon},\tau\right)e^{-i\omega\tau}d\tau = \varepsilon\int Q_{2a}\left(\frac{T}{\varepsilon},\tau\right)e^{-i\omega\tau}d\tau + \varepsilon^2\int Q_{2b}\left(\frac{T}{\varepsilon},\tau\right)e^{-i\omega\tau}d\tau + O(\varepsilon^3),$$

$$\begin{aligned}\int Q_{2a}\left(\frac{T}{\varepsilon},\tau\right)e^{-i\omega\tau}d\tau &= \frac{f_{R0}}{(2\pi)^2}\iint R(\theta)n_{\omega_1}(T)n_{\omega_2}(T)[e^{i(\omega_1-\omega_2)\theta}-e^{i(\omega_2-\omega_1)\theta}]e^{i(\omega_2-\omega)\tau}d\omega_1 d\omega_2 d\theta d\tau\\
&= \frac{f_{R0}}{2\pi}\iint R(\theta)n_{\omega_1}(T)n_\omega(T)[e^{i(\omega_1-\omega)\theta}-e^{i(\omega-\omega_1)\theta}]d\omega_1 d\theta\\
&= \frac{f_{R0}}{2\pi}\int[\widetilde{R}(\omega-\omega_1)-\widetilde{R}(\omega_1-\omega)]n_{\omega_1}(T)n_\omega(T)d\omega_1\\
&= \frac{f_{R0}i}{\pi}\left[\int \widetilde{R}_{\mathrm{i}}(\omega-\omega_1)n_{\omega_1}(T)d\omega_1\right]n_\omega(T),\end{aligned}$$

$$\begin{aligned}\int Q_{2b}\left(\frac{T}{\varepsilon},\tau\right)e^{-i\omega\tau}d\tau &= \frac{f_{R0}}{2(2\pi)^2}\iint R(\theta)\theta(e^{i(\omega_2-\omega_1)\theta}-e^{i(\omega_1-\omega_2)\theta})\partial_T\big[n_{\omega_1}(T)n_{\omega_2}(T)\big]e^{i(\omega_2-\omega)\tau}d\omega_1 d\omega_2 d\theta d\tau\\
&\quad+\frac{f_{R0}}{2(2\pi)^2}\iint R(\theta)(e^{i(\omega_2-\omega_1)\theta}+e^{i(\omega_1-\omega_2)\theta})\partial_T n_{\omega_1}(T)n_{\omega_2}(T)\tau e^{i(\omega_2-\omega)\tau}d\omega_1 d\omega_2 d\theta d\tau\\
&= \frac{if_{R0}}{2(2\pi)^2}\iint R(\theta)\partial_{\omega_1}(e^{i(\omega_2-\omega_1)\theta}+e^{i(\omega_1-\omega_2)\theta})\partial_T\big[n_{\omega_1}(T)n_{\omega_2}(T)\big]e^{i(\omega_2-\omega)\tau}d\omega_1 d\omega_2 d\theta d\tau\\
&\quad+\frac{if_{R0}}{2(2\pi)^2}\iint R(\theta)(e^{i(\omega_2-\omega_1)\theta}+e^{i(\omega_1-\omega_2)\theta})\partial_T n_{\omega_1}(T)n_{\omega_2}(T)\partial_\omega[e^{i(\omega_2-\omega)\tau}]d\omega_1 d\omega_2 d\theta d\tau\\
&= -\frac{if_{R0}}{4\pi}\iint R(\theta)(e^{i(\omega-\omega_1)\theta}+e^{i(\omega_1-\omega)\theta})\partial_T\big[\partial_{\omega_1}n_{\omega_1}(T)n_\omega(T)\big]d\omega_1 d\theta\\
&\quad+\frac{if_{R0}}{4\pi}\partial_\omega\iint R(\theta)(e^{i(\omega-\omega_1)\theta}+e^{i(\omega_1-\omega)\theta})\partial_T n_{\omega_1}(T)n_\omega(T)d\omega_1 d\theta\\
&= -\frac{if_{R0}}{2\pi}\int\widetilde{R}_{\mathrm{r}}(\omega-\omega_1)\partial_T\big[\partial_{\omega_1}n_{\omega_1}(T)n_\omega(T)\big]d\omega_1+\frac{if_{R0}}{2\pi}\partial_\omega\int\widetilde{R}_{\mathrm{r}}(\omega-\omega_1)\partial_T n_{\omega_1}(T)n_\omega(T)d\omega_1\\
&= -\frac{if_{R0}}{2\pi}\int\partial_\omega\widetilde{R}_{\mathrm{r}}(\omega-\omega_1)\partial_T\big[n_{\omega_1}(T)n_\omega(T)\big]d\omega_1+\frac{if_{R0}}{2\pi}\partial_\omega\int\widetilde{R}_{\mathrm{r}}(\omega-\omega_1)\partial_T n_{\omega_1}(T)n_\omega(T)d\omega_1\\
&= -if_{R0}\partial_\omega\left[\frac{1}{2\pi}\int\widetilde{R}_{\mathrm{r}}(\omega-\omega_1)n_{\omega_1}(T)d\omega_1\right]\partial_T n_\omega(T)\\
&\quad+if_{R0}\partial_T\left[\frac{1}{2\pi}\int\widetilde{R}_{\mathrm{r}}(\omega-\omega_1)n_{\omega_1}(T)d\omega_1\right]\partial_\omega n_\omega(T).\end{aligned}$$

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