significant behavior and has been extensively studied and applied in many scientific areas. The general framework presented here can be further investigated using some books listed in Further Reading below.

See also

Fiber and Guided Wave Optics: Nonlinear Optics. **Nonlinear Optics, Applications:** Phase Matching. **Nonlinear Optics, Basics:** $\chi^{(3)}$–Third-Harmonic Generation. **Nonlinear Sources:** Harmonic Generation in Gases. **Optical Parametric Devices:** Optical Parametric Oscillators (Continuous Wave); Optical Parametric Oscillators (Pulsed).

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Kramers–Krönig Relations in Nonlinear Optics

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Since their introduction nearly 75 years ago, the Kramers–Krönig (KK) dispersion relations have been widely appreciated and applied in the analysis of linear optical systems. Because they are a consequence of strict causality, the KK relations apply not only to optical systems, but also to any linear, causal system such as electrical networks and particle scattering. In this article, we review the formulation and application of these relations in nonlinear optical systems. Simple logical arguments are used to derive dispersion relations that relate the nonlinear absorption coefficient to the nonlinear refraction coefficient. More general formalisms are then derived that apply to all nonlinear susceptibilities including the harmonic generating cases. Examples of recent successful application of these dispersion relations in analyzing various nonlinear materials will be presented.

The mathematical formalism of the KK dispersion relations in nonlinear optics was studied in the formative days of the field. The great usefulness of these relations was appreciated only recently, however, when they were used to derive the dispersion of the optical Kerr effect in solids from the corresponding nonlinear absorption coefficients, including two-photon absorption.

Before examining the details of KK relations in nonlinear optical systems, it is instructive to revisit the linear dispersion relations and their derivation based on the logic of causality. We will begin this task by introducing the definition of the linear as well as nonlinear susceptibilities $\chi^{(n)}$. In most nonlinear optics texts, the total material polarization (P) that drives the wave equation for the electric field (E) is expressed as

$$\begin{aligned} P_i(t) = {} & \varepsilon_0 \int_{-\infty}^{\infty} R_{ij}^{(1)}(t-t_1)E_j(t_1)\mathrm{d}t_1 + \varepsilon_0 \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} \\ & \times R_{ijk}^{(2)}(t-t_1,t-t_2)E_j(t_1)E_k(t_2)\mathrm{d}t_1\,\mathrm{d}t_2 \\ & + \varepsilon_0 \int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} R_{ijkl}^{(3)}(t-t_1,t-t_2,t-t_3) \\ & \times E_j(t_1)E_k(t_2)E_l(t_3)\mathrm{d}t_1\,\mathrm{d}t_2\,\mathrm{d}t_3 + \cdots \end{aligned} \quad [1]$$

where $R^{(n)}$ is defined as the nth-order, time-dependent response function or time-dependent susceptibility. The subscripts are polarization indices indicating, in general, the tensor nature of the interactions. The summation over the various indices $j, k, l, \ldots$ is implied for the various tensor elements of $R^{(n)}$. Upon Fourier transformation, we obtain:

$$\begin{aligned} \vec{P}_i(\omega) = {} & \varepsilon_0 \int_{\infty}^{-\infty} \mathrm{d}\omega_1\, \chi_{ij}^{(1)}(\omega_1)\vec{E}_j(\omega_1)\delta(\omega-\omega_1) \\ & + \varepsilon_0 \int_{\infty}^{-\infty} \mathrm{d}\omega_1 \int_{\infty}^{-\infty} \mathrm{d}\omega_2 \\ & \times \chi_{ijk}^{(2)}(\omega_1,\omega_2)\vec{E}_j(\omega_1)\vec{E}_k(\omega_2)\delta(\omega-\omega_1-\omega_2) \\ & + \varepsilon_0 \int_{\infty}^{-\infty} \mathrm{d}\omega_1 \int_{\infty}^{-\infty} \mathrm{d}\omega_2 \int_{\infty}^{-\infty} \mathrm{d}\omega_3 \\ & \times \chi_{ijkl}^{(3)}(\omega_1,\omega_2,\omega_3)\vec{E}_j(\omega_1)\vec{E}_k(\omega_2)\vec{E}_l(\omega_3) \\ & \times \delta(\omega-\omega_1-\omega_2-\omega_3), \cdots \end{aligned} \quad [2]$$

where δ is the Dirac delta-function. Here the $E(\omega)$ are Fourier transforms of the corresponding electric field. The nth-order susceptibility is defined as the Fourier transform of the nth-order response function:

$$\begin{aligned} \chi_{ijk\ldots n}^{(n)}(\omega_1,\omega_2,\ldots,\omega_m) = {} & \int_{-\infty}^{+\infty}\mathrm{d}\tau_1 \int_{-\infty}^{+\infty}\mathrm{d}\tau_2 \cdots \int_{-\infty}^{+\infty} \\ & \times \mathrm{d}\tau_n R_{ijk\ldots m}^{(n)}(\tau_1,\tau_2,\ldots,\tau_m)\mathrm{e}^{\mathrm{i}(\omega_1\tau_1+\omega_2\tau_2+\cdots+\omega_m\tau_m)} \end{aligned} \quad [3]$$

For simplicity, we drop the polarization indices $i, j, \ldots$, and thus ignore the tensor properties of $\chi^{(n)}$ as well as the vector nature of the electric fields.

Let us for the moment concentrate on the linear polarization alone and derive the linear KK relations for the first-order susceptibility $\chi^{(1)}(\omega)$. For this, we rewrite eqn [3] for $n = 1$:

$$\chi^{(1)}(\omega) = \int_{-\infty}^{\infty} R^{(1)}(\tau)\mathrm{e}^{-\mathrm{i}\omega\tau}\,\mathrm{d}\tau \quad [4]$$

(As defined above, $\chi^{(1)}(\omega)$ and $R^{(1)}(\tau)$ are not a strict Fourier transform pair because of a missing factor of 2π.) Causality means that the effect cannot precede the cause. This can be restated mathematically as:

$$R^{(1)}(t) = R^{(1)}(t)\Theta(t) \quad [5]$$

i.e., the response to an impulse at $t = 0$ must be zero for $t < 0$. Here $\Theta(t)$ is the Heaviside step function defined as $\Theta(t) = 1$ for $t > 0$ and $\Theta(t) = 0$ for $t < 0$. Upon Fourier transforming this equation, the product in the time domain becomes a convolution integral in frequency space

$$\begin{aligned} \chi^{(1)}(\omega) &= \chi^{(1)}(\omega)\left[\frac{\delta(\omega)}{2} + \frac{\mathrm{i}}{2\pi\omega}\right] \\ &= \frac{\chi^{(1)}(\omega)}{2} + \frac{\mathrm{i}}{2\pi}\wp\int_{-\infty}^{\infty}\frac{\chi^{(1)}(\omega')}{\omega-\omega'}\mathrm{d}\omega' \\ &= \frac{1}{\mathrm{i}\pi}\wp\int_{-\infty}^{\infty}\frac{\chi^{(1)}(\omega')}{\omega'-\omega}\mathrm{d}\omega' \end{aligned} \quad [6]$$

which is the KK relation for the linear optical susceptibility. The symbol $\wp$ stands for the Cauchy principal value of the integral. The KK relation is thus a restatement of the causality condition [5] in the frequency domain. Taking the real part we have,

$$\Re e\{\chi^{(1)}(\omega)\} = \frac{1}{\pi}\wp\int_{-\infty}^{\infty}\frac{\Im m\{\chi^{(1)}(\omega')\}}{\omega'-\omega}\mathrm{d}\omega' \quad [7]$$

Taking the imaginary part of eqn [6] leads to a similar relation relating the imaginary part to an integral involving the real part. It is conventional to write the optical dispersion relations in terms of the more familiar quantities of refractive index, $n(\omega)$, and absorption coefficient, $\alpha(\omega)$. For $|\chi^{(1)}| \ll 1$ then $n - 1 = \Re e\{\chi^{(1)}\}/2$ and $\alpha = \omega\Im m\{\chi^{(1)}\}/c$, and eqn [7] is transformed into

$$n(\omega) - 1 = \frac{c}{\pi}\wp\int_{0}^{\infty}\frac{\alpha(\omega')}{\omega' 2 - \omega^2}\mathrm{d}\omega' \quad [8]$$

where we additionally used the reality conditions of $n(\omega) = n(-\omega)$, and $\alpha(\omega) = \alpha(-\omega)$ to change the lower integral limit to 0. More rigorous analysis shows that eqn [8] is general and valid for any value of $|\chi^{(1)}|$. Although the KK dispersion relations and the extent of their applications in linear optics are well understood, some confusion sometimes exists about their applications to nonlinear optics. Causality clearly holds for both linear and nonlinear systems. The question is: what form do the resulting dispersion relations take in a nonlinear system? The linear Kramers–Krönig relations were derived from linear system theory, so it would appear to be impossible to apply the same logic to a nonlinear system. The key insight is that one can linearize the system. This is illustrated in Figure 1 where a linear

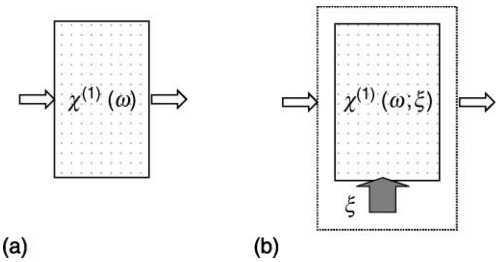

Figure 1 (a) A causal linear system obeying KK relations. (b) The system in (a) when externally perturbed by ξ. The dotted box now represents our new linear causal system whose altered $\chi^{(1)}$ obeys the KK relations.

(and of course, causal) optical material is transformed into a 'new' linear system that now contains the material and an external perturbation denoted by ξ. Although we are interested in perturbations of an optical nature, this formalism is general under any type of perturbation. It is important to appreciate the fact that our new system is causal even in the presence of the perturbation. This allows us to write down a modified form of the Kramers–Krönig relation linking the index of refraction to the absorption:

$$\begin{aligned}&[n(\omega) + \Delta n(\omega;\zeta)] - 1\\&\quad = \frac{c}{\pi}\wp\int_0^\infty \frac{\alpha(\omega') + \Delta\alpha(\omega';\zeta)}{\omega'2 - \omega^2}\mathrm{d}\omega' \qquad [9]\end{aligned}$$

which, after subtracting the linear relation between n and α leaves a relation between the changes in index and absorption:

$$\Delta n(\omega;\zeta) = \frac{c}{\pi}\wp\int_0^\infty \frac{\Delta\alpha(\omega';\zeta)}{\omega'2 - \omega^2}\mathrm{d}\omega' \qquad [10]$$

where ζ denotes a general perturbation. An equivalent relation also exists whereby the change in absorption coefficient can be calculated from the change in the refractive index. It is essential that the perturbation be independent of frequency of observation, ω', in the integral (i.e., the excitation ζ must be held constant as ω' is varied).

Equation [10] has been used to determine refractive changes due to 'real' excitations such as thermal and free-carrier nonlinearities in semiconductors. In those cases, ζ denotes either ΔT (change of temperature) or ΔN (change of free-carrier density), respectively. In the former case, one calculates the refractive index change resulting from a thermally excited electron–hole plasma and the temperature shift of the band edge. For cases where an electron–hole plasma is injected (e.g., optically), the change of absorption gives the plasma contribution to the refractive index. In this case, the ζ parameter in eqn [10] is taken as the change in plasma density regardless of the mechanism of generation or the optical frequency.

Let us now extend this formalism to the case where the perturbation is virtual, occurring at an excitation frequency Ω that is below any material resonance. To the lowest order in the excitation irradiance I_Ω, we write

$$\Delta\alpha(\omega;\zeta) = \Delta\alpha(\omega;\Omega) = 2\alpha_2(\omega;\Omega)I_\Omega \qquad [11]$$

and

$$\Delta n(\omega;\zeta) = \Delta n(\omega;\Omega) = 2n_2(\omega;\Omega)I_\Omega \qquad [12]$$

where n_2 and α_2 are the nonlinear refractive index and absorption coefficients of the material, respectively. By definition, these coefficients are related to the third-order nonlinear susceptibility $\chi^{(3)}(\omega_1,\omega_2,\omega_3)$ via (*see* **Nonlinear Optics, Basics**: Nomenclature and Units)

$$n_2(\omega;\Omega) = \frac{3}{4\varepsilon_0 n_0(\omega)n_0(\Omega)c}\Re e\{\chi^{(3)}(\omega,-\Omega,\Omega)\} \qquad [13]$$

and

$$\alpha_2(\omega;\Omega) = \frac{3\omega_\mathrm{a}}{2\varepsilon_0 n_0(\omega)n_0(\Omega)c^2}\Im m\{\chi^{(3)}(\omega,-\Omega,\Omega)\} \qquad [14]$$

We can therefore write the dispersion relations between α_2 and n_2:

$$n_2(\omega;\Omega) = \frac{c}{\pi}\wp\int_0^\infty \frac{\alpha_2(\omega';\Omega)}{\omega'2 - \Omega^2}\mathrm{d}\omega' \qquad [15]$$

Note that even when the degenerate $n_2(\omega) = n_2(\omega;\omega)$ is desired (at a given ω), the dispersion relation requires that we should know the nondegenerate absorption spectrum $\alpha_2(\omega';\omega)$ at all frequencies ω'.

Let us pause here and discuss some physical mechanisms that can be involved for a given system of interest. Consider a material characterized by an optical resonance occurring at, say ω_0 (i.e., a degenerate two-level system). For a solid, this resonance can be regarded as that of the fundamental energy gap; $\omega_0 = \omega_\mathrm{g} = E_\mathrm{g}/\hbar$ in a two-band system. Now, let us examine how the presence of an optical excitation at $\Omega < \omega_0$ can alter the absorption spectrum (at a variable probe ω'). In the quantum mechanical picture, this gives rise to a 'new' material whose perturbed wave functions are 'dressed' by the intensity and frequency of the applied optical field.

The lowest-order correction to the absorption is given by $\alpha_2(\omega';\Omega)$ which involves three major physical processes. Recalling that $\Omega < \omega_0$, these processes include (1) two-photon absorption (2PA) when $\omega' + \Omega \rightarrow \omega_0$ and (2) Raman-induced absorption when $\omega' - \Omega \rightarrow \omega_0$, both implying an absorption of a photon at the probe frequency ω' (i.e., $\alpha_2 > 0$). The third process can be identified as resulting from the blue-shift (for $\Omega < \omega_0$) of the resonance (known as the quadratic optical Stark effect) caused by the excitation field. For our two-level system, the latter results in a decrease followed by an increase in absorption in the vicinity of ω_0. An example of the overall absorption changes due to such processes is shown in Figure 2 where $\alpha_2(\omega';\Omega)$ is qualitatively plotted for a degenerate two-level system. We should note that the relative magnitude of each contribution as well as the width and shape of the resonances are chosen arbitrarily for the purpose of illustration. Using the KK relation in eqn [15], we can now arrive at the nonlinear index coefficient $n_2(\omega;\Omega)$. The result of this transformation is also given in Figure 2. The above simple example elucidates the key concepts involving the relationship between nonlinear absorption and refraction in materials for third-order processes. These concepts, when applied more rigorously to semiconductors, have been successful in predicting the sign, magnitude, and dispersion of n_2 due to the anharmonic motion of bound electrons. This will be briefly discussed later. Returning to the mathematical foundation of KK relations, we use eqns [13] and [14] to write eqn [15] in terms of the nonlinear susceptibility $\chi^{(3)}$:

$$\Re e\{\chi^{(3)}(\omega_1,\omega_2,-\omega_2)\} = \frac{1}{\pi}\wp\int_{-\infty}^{\infty}\frac{\Im m\{\chi^{(3)}(\omega',\omega_2,\omega_2)\}}{\omega'-\omega_1}\,d\omega' \qquad [16]$$

The above dispersion relation for $\chi^{(3)}$ was obtained using the physical and intuitive arguments that followed the linearization scheme depicted in Figure 1. General dispersion relations can be formulated following a mathematical procedure that is similar to the derivation of the linear KK relations. In this case we apply the causality condition directly to the nth-order nonlinear response $R^{(n)}$. For example, without loss of generality, we can write

$$R^{(n)}(\tau_1,\tau_2,\ldots,\tau_n) = R^{(n)}(\tau_1,\tau_2,\ldots,\tau_n)\Theta(\tau_j) \qquad [17]$$

and then calculate the Fourier transform of this equation. Here j can apply to any one of the indices $1, 2, \ldots, n$. Following the same procedure as for a linear response, we obtain

$$\chi^{(n)}(\omega_1,\omega_2,\ldots,\omega_j,\ldots,\omega_n) = \frac{-i}{\pi}\wp\int_{-\infty}^{\infty}\frac{\chi^{(n)}(\omega_1,\omega_2,\ldots,\omega',\ldots,\omega_n)}{\omega_j-\omega'}\,d\omega' \qquad [18]$$

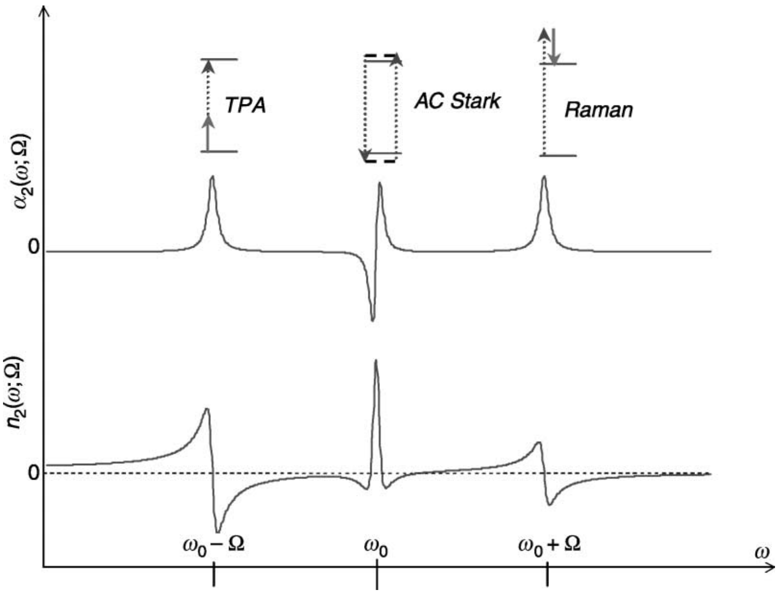

Figure 2 Upper trace: the nonlinear absorption coefficient in a fictitious 'degenerate' two-level system. Lower trace: the resulting nonlinear refractive index obtained using the KK relations. The insets show the three possible physical mechanisms involved.

By separating the real and imaginary parts of this equation, we get the generalized Kramers–Krönig relation pairs for a nondegenerate, nth-order nonlinear susceptibility:

$$\Re e\{\chi^{(n)}(\omega_1,\omega_2,\ldots,\omega_j,\ldots,\omega_n)\} = \frac{1}{\pi}\wp\int_{-\infty}^{\infty}\frac{\Im m\{\chi^{(n)}(\omega_1,\omega_2,\ldots,\omega',\ldots,\omega_n)\}}{\omega'-\omega_j}\mathrm{d}\omega' \qquad [19]$$

and

$$\Im m\{\chi^{(n)}(\omega_1,\omega_2,\ldots,\omega_j,\ldots,\omega_n)\} = -\frac{1}{\pi}\wp\int_{-\infty}^{\infty}\frac{\Re e\{\chi^{(n)}(\omega_1,\omega_2,\ldots,\omega',\ldots,\omega_n)\}}{\omega'-\omega_j}\mathrm{d}\omega' \qquad [20]$$

In particular, for $\chi^{(3)}$ processes having $\omega_1=\omega_a,\omega_2\omega_b$, and ${}_3b$, this becomes identical to eqn [16].

Note that in describing the nonlinear susceptibilities, no special attention was given to the harmonic generating susceptibility $\chi^{(N)}(N\omega)\equiv\chi^{(N)}(\omega,\omega,\ldots\omega)$, i.e., the susceptibility generating the Nth harmonic at $N\omega$. It turns out that in addition to the KK relations given by eqns [19] and [20], the real and imaginary parts of $\chi^{(N)}(N\omega)$ can also be related in different sets of dispersion integrals that involve only the degenerate forms of the susceptibilities. A more general yet simple analysis gives the most general form of KK relations for any type of $\chi^{(n)}$:

$$\chi^{(n)}(\omega_1+p_1\omega,\omega_2+p_2\omega,\ldots,\omega_m+p_m\omega) = \frac{1}{\mathrm{i}\pi}\wp\int_{-\infty}^{\infty}\frac{\chi^{(n)}(\omega_1+p_1\Omega,\omega_2+p_2\Omega,\ldots,\omega_m+p_m\Omega)}{\Omega-\omega}\mathrm{d}\Omega \qquad [21]$$

for all $p_1,p_2,\ldots,p_m\geq 0$. Setting $\omega_1=\omega_2=\cdots=\omega_m\equiv 0$, and $p_1=p_2=\cdots=p_m=1$ in eqn [21] yields an interesting form of the KK relations for the Nth-harmonic susceptibilities:

$$\Re e\{\chi^{(N)}(N\omega)\}=\frac{1}{\pi}\wp\int_{-\infty}^{\infty}\frac{\Im m\{\chi^{(N)}(N\omega')\}}{\omega'-\omega}\mathrm{d}\omega' \qquad [22]$$

These dispersion relations have allowed calculations of $\chi^{(2)}(2\omega)$ and $\chi^{(3)}(3\omega)$ in semiconductors using full band structures.

At the beginning of this article, it was noted that all the KK relations for nonlinear optics were known in the early days of the field. Their application in unifying nonlinear absorption (in particular two-photon absorption) and the optical Kerr effect (n_2) in solids came only much later. More recent work demonstrated that the KK relations are a powerful analytical tool in nonlinear optics. Following the picture of a degenerate two-level system shown in Figure 2, a simple two-band model has been used to calculate the nonlinear absorption coefficient, $\alpha_2(\omega_1;\omega_2)$, resulting from three mechanisms: 2PA, the Raman absorption process, and the ac Stark effect. The optical Kerr coefficient $n_2(\omega_1;\omega_2)$ was then calculated using eqn [15]. Of particular practical interest is the degenerate case ($\omega_1=\omega_2=\omega$), from which the 2PA coefficient $\beta(\omega)=\alpha_2(\omega;\omega)$ can be extracted. Figure 3 depicts the calculated dispersion of n_2 and β as a function of $\hbar\omega/E_g$ where E_g is the bandgap energy of the solid. The dispersion of n_2 and its sign reversal shown in Figure 3 has been observed experimentally in many optical solids.

Finally, let us discuss a related implication of causality in nonlinear optics. The KK dispersion relations are traditionally derived in terms of internal material parameters such as susceptibility, absorption coefficient, and refractive index. Similar to the case of electrical circuits, one can obtain dispersion relations that apply to an external transfer function of the system that relates an input signal to an output signal. In this case, the dispersion of the transfer function includes system structure as well as the intrinsic dispersion of the material. As an optical (and linear) example, consider a Fabry–Perot etalon. The optical transmission of this system has well-known spectral

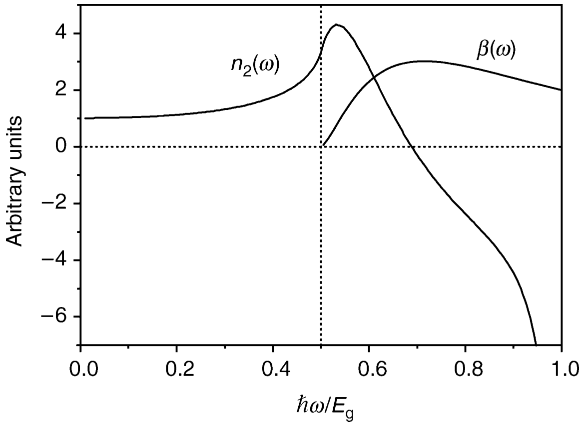

Figure 3 The two-photon absorption coefficient in semiconductors (β) calculated for a two-band model. The resultant nonlinear refractive index (n_2) obtained using a KK transformation of the calculated nondegenerate nonlinear absorption coefficient includes all major mechanisms.

features that are primarily caused by structural dispersion (i.e., interference) in addition to the intrinsic dispersion of the material. Causality still demands that the transmitted signal has a phase variation whose value and dispersion can be determined using a KK relation linking the real and imaginary parts of the transmission coefficient. In other words, the KK relations provide a spectral correlation between the real and imaginary components of the transfer function which in turn may translate to a spectral correlation between the phase and amplitude of the transmitted signal. However, the variations in phase do not necessarily imply the presence of a varying index of refraction, nor does an amplitude variation suggest the existence of material absorption (dissipation). Ultimately, this implies that any mechanism causing a variation in amplitude (including reflection, scattering, or absorption) must be accompanied by a phase variation. (One should note that the reverse of the previous statement is not necessarily true; i.e., a variation in phase does not have to be accompanied by an amplitude modulation.)

In nonlinear optics with the 'black box' approach of Figure 1, the optical perturbation ξ (with frequency Ω) can render an amplitude variation in the probe (at ω) using various frequency mixing schemes in a noncentrosymmetric material (i.e., with nonzero $\chi^{(2)}$). For instance, the probe at ω can be depleted by nonlinear conversion to $\omega_{\text{sum}} = \omega + \Omega$ via sum-frequency generation involving $\chi^{(2)}(\omega, \Omega)$ and/or to $\omega_{\text{diff}} = \omega - \Omega$ via difference-frequency generation involving $\chi^{(2)}(\omega, -\Omega)$. Such a conversion (or depletion) should be accompanied by a phase variation according to the KK dispersion relations. This type of nonlinear phase modulation is known as a $\chi^{(2)}$:$\chi^{(2)}$ cascaded nonlinearity. Such cascaded processes are routinely (and more simply) analyzed with Maxwell's equations governing the propagation of beams in a second-order nonlinear material. The KK relations, however, provide an interesting physical perspective of the process. We find that cascaded second-order nonlinearities are yet another manifestation of causality in nonlinear optics.

List of Units and Nomenclature

α	linear absorption coefficient
α_2	nonlinear absorption coefficient
β	two-photon absorption coefficient
$\chi^{(n)}$	*n*th-order nonlinear optical susceptibility
n	linear refractive index
n_2	nonlinear refractive index coefficient, coefficient of optical Kerr effect
$\Theta(t)$	step function
$\wp$	principal value
2PA	two-photon absorption
KK relations	Kramers–Krönig relations

See also

Materials for Nonlinear Optics: Liquid Crystals for NLO. **Nonlinear Optics, Basics:** Cascading; Nomenclature and Units.

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