Nonlinear Optics Basics: Kramers-Krönig Relations in Nonlinear Optics

Mansoor Sheik-Bahae

Department of Physics and Astronomy and Department of Electrical and Computer Engineering University of New Mexico, Albuquerque, NM 87131

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 chapter, 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 ralations 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

$$\vec{P}_{i}(t) = \varepsilon_{0} \int_{-\infty}^{\infty} R_{ij}^{(1)}(t-t_{1})\vec{E}_{j}(t_{1})dt_{1} + \varepsilon_{0} \int_{-\infty-\infty}^{\infty} R_{ijk}^{(2)}(t-t_{1},t-t_{2})\vec{E}_{j}(t_{1})\vec{E}_{k}(t_{2})dt_{1}dt_{2} + \varepsilon_{0} \int_{-\infty-\infty-\infty}^{\infty} R_{ijkl}^{(3)}(t-t_{1},t-t_{2},t-t_{3})\vec{E}_{j}(t_{1})\vec{E}_{k}(t_{2})\vec{E}_{l}(t_{3})dt_{1}dt_{2}dt_{3} + \dots$$
(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*,... is implied for the various tensor elements of $R^{(n)}$. Upon Fourier transformation, we obtain:

$$\vec{P}_{i}(\omega) = \varepsilon_{0} \int_{-\infty}^{\infty} d\omega_{1} \chi_{ij}^{(1)}(\omega_{1}) \vec{E}_{j}(\omega_{1}) \delta(\omega - \omega_{1})$$

$$+\varepsilon_{0} \int_{-\infty}^{\infty} d\omega_{1} \int_{-\infty}^{\infty} d\omega_{2} \chi_{ijk}^{(2)}(\omega_{1}, \omega_{2}) \vec{E}_{j}(\omega_{1}) \vec{E}_{k}(\omega_{2}) \delta(\omega - \omega_{1} - \omega_{2})$$

$$+\varepsilon_{0} \int_{-\infty}^{\infty} d\omega_{1} \int_{-\infty}^{\infty} d\omega_{2} \int_{-\infty}^{\infty} d\omega_{3} \chi_{ijkl}^{(3)}(\omega_{1}, \omega_{2}, \omega_{3}) \vec{E}_{j}(\omega_{1}) \vec{E}_{k}(\omega_{2}) \vec{E}_{l}(\omega_{3}) \delta(\omega - \omega_{1} - \omega_{2} - \omega_{3}), \dots$$
(2)

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

$$\chi_{ijk...n}^{(n)}(\omega_1,\omega_2,...,\omega_m) = \int_{-\infty}^{+\infty} d\tau_1 \int_{-\infty}^{+\infty} d\tau_2 .. \int_{-\infty}^{+\infty} d\tau_n R_{ijk...m}^{(n)}(\tau_1,\tau_2,..\tau_m) e^{i(\omega_1\tau_1+\omega_2\tau_2+...\omega_m\tau_m)}.$$
 (3)

For simplicity, we drop the polarization indices i,j,..., 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 Eq. (3) for n=1:

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

(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:

$$R^{(1)}(t) = R^{(1)}(t)\Theta(t), \qquad (5)$$

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

$$\chi^{(1)}(\omega) = \chi^{(1)}(\omega) * \left[\frac{\delta(\omega)}{2} + \frac{i}{2\pi\omega}\right] = \frac{\chi^{(1)}(\omega)}{2} + \frac{i}{2\pi} \wp \int_{-\infty}^{\infty} \frac{\chi^{(1)}(\omega')}{\omega - \omega'} d\omega' = \frac{1}{i\pi} \wp \int_{-\infty}^{\infty} \frac{\chi^{(1)}(\omega')}{\omega' - \omega} d\omega', (6)$$

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 (Eq. (5)) in the frequency domain. Taking the real part we have,

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

Taking the imaginary part of Eq. (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 $|\chi^{(1)}| <<1$ then $n-1 = \Re e\{\chi^{(1)}\}/2$ and $\alpha = \omega \Im m\{\chi^{(1)}\}/c$, and Eq. (7) is transformed into

$$n(\omega) - 1 = \frac{c}{\pi} \wp_0^{\infty} \frac{\alpha(\omega')}{{\omega'}^2 - \omega^2} d\omega', \qquad (8)$$

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 show that Eq. (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 impossible to apply the same logic to a nonlinear system. The key insight is that one can linearize the system. This is illustrated in Fig. 1 where a linear (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: <Figure 1 near here>

$$[n(\omega) + \Delta n(\omega; \zeta)] - 1 = \frac{c}{\pi} \bigotimes_{0}^{\infty} \frac{\alpha(\omega') + \Delta \alpha(\omega'; \zeta)}{\omega'^{2} - \omega^{2}} d\omega'$$
(9)

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

$$\Delta n(\omega;\zeta) = \frac{c}{\pi} \wp_0^{\tilde{\omega}} \frac{\Delta \alpha(\omega';\zeta)}{\omega'^2 - \omega^2} d\omega'$$
(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 Eq. (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

$$\Delta \alpha(\omega; \zeta) = \Delta \alpha(\omega; \Omega) = 2\alpha_2(\omega; \Omega)I_{\Omega} \quad (11)$$

and

$$\Delta n(\omega; \zeta) = \Delta n(\omega; \Omega) = 2n_2(\omega; \Omega) I_{\Omega} , (12)$$

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 MS 753, chapter by Hasselbeck)

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

and

$$\alpha_2(\omega;\Omega) = \frac{3\omega_a}{2\varepsilon_0 n_0(\omega) n_0(\Omega)c^2} \Im m\{\chi^{(3)}(\omega,-\Omega,\Omega)\}.(14)$$

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

$$n_2(\omega;\Omega) = \frac{c}{\pi} \wp \int_0^\infty \frac{\alpha_2(\omega';\Omega)}{\omega'^2 - \Omega^2} d\omega'.$$
(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; $\omega_0=\omega_g=E_g/\hbar$ 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 a 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 (a) two-photon absorption (2PA) when $\omega' + \Omega \rightarrow \omega_0$ and (b) Raman-induced absorption when $\omega' \cdot \Omega \rightarrow \omega_0$, 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 Fig. 2 where $\alpha_2(\omega';\Omega)$ is qualitatively plotted for a degenerate two-level system. <Figure 2 near here> 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 Eq. (15), we can now arrive at the nonlinear index coefficient $n_2(\omega; \Omega)$. The result of this transformation is also given in Fig. 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 n₂ due to the anharmonic motion of bound-electrons. This will be briefly discussed later. Returning to the mathematical foundation of KK relations, we use Eqs. (13) and (14) to write Eq. (15) in terms of the nonlinear susceptibility $\chi^{(3)}$:

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

The above dispersion relation for $\chi^{(3)}$ was obtained using the physical and intuitive arguments that followed the linearization scheme depicted in **Fig. 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 n-th order nonlinear response $R^{(n)}$. For example, without loss of generality, we can write

$$R^{(n)}(\tau_1, \tau_2, ..., \tau_n) = R^{(n)}(\tau_1, \tau_2, ..., \tau_n) \Theta(\tau_j)$$
(17)

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

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

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

$$\Re e \left\{ \chi^{(n)}(\omega_1, \omega_2, ..., \omega_j, ..., \omega_n) \right\} = \frac{1}{\pi} \wp \int_{-\infty}^{\infty} \frac{\Im m \left\{ \chi^{(n)}(\omega_1, \omega_2, ..., \omega', ..., \omega_n) \right\}}{\omega' - \omega_j} d\omega', \quad (19)$$

and

$$\Im m \left\{ \chi^{(n)}(\omega_1, \omega_2, ..., \omega_j, ..., \omega_n) \right\} = -\frac{1}{\pi} \wp \int_{-\infty}^{\infty} \frac{\Re e \left\{ \chi^{(n)}(\omega_1, \omega_2, ..., \omega', ..., \omega_n) \right\}}{\omega' - \omega_j} d\omega'. (20)$$

In particular, for $\chi^{(3)}$ processes having $\omega_1 = \omega_a$, $\omega_2 = \omega_b$ and $\omega_3 = -\omega_b$, this becomes identical to Eq. (16).

Note that in describing the nonlinear susceptibilities, no special attention was given to the harmonic generating susceptibility $\chi^{(N)}(N\omega) \equiv \chi^{(N)}(\omega,\omega,...\omega)$, i.e. the susceptibility generating the Nth harmonic at N ω . It turns out that in addition to the KK relations given by Eqs. (19) and (20), the real and imaginary parts of $\chi^{(N)}(N\omega)$ can also be related in a 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 $\chi^{(n)}$:

$$\chi^{(n)}(\omega_{1}+p_{1}\omega,\omega_{2}+p_{2}\omega,...,\omega_{m}+p_{m}\omega) = \frac{1}{i\pi} \wp \int_{-\infty}^{\infty} \frac{\chi^{(n)}(\omega_{1}+p_{1}\Omega,\omega_{2}+p_{2}\Omega,...,\omega_{m}+p_{m}\Omega)}{\Omega-\omega} d\Omega$$
(21)

for all $p_1, p_2, \dots, p_m \ge 0$. Setting $\omega_1 = \omega_2, = \dots, \omega_m \equiv 0$, and $p_1 = p_2 \dots p_m = 1$ in Equation (21) yields an interesting form of the KK relations for the Nth-harmonic susceptibilities:

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

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 chapter, 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 (n₂) in solids only came much later. The 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 **Fig. 2**, a simple two-band model has been used to calculate the nonlinear absorption coefficient, $\alpha_2(\omega_1;\omega_2)$, resulting from the three mechanisms: 2PA, the Raman absorption process, and the A.C. Stark effect. The optical Kerr coefficient $n_2(\omega_1;\omega_2)$ was then calculated using Eq. (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. **Fig. 3** depicts the calculated dispersion of n_2 and β as a function of $\hbar\omega/E_g$ where E_g is the band-gap energy of the solid. The dispersion of n_2 and its sign reversal shown in **Fig. 3** has been observed experimentally in many optical solids. <Figure 3 near here>

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 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 have 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 **Fig. 1**, the optical perturbation ξ (with frequency Ω) can render an amplitude variation in the probe (at ω) using various

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frequency mixing schemes in a non-centrosymmetric material (i.e. with nonzero $\chi^{(2)}$). For instance, the probe at ω can be *depleted* by nonlinear conversion to $\omega_{sun}=\omega+\Omega$ via sum frequency generation involving $\chi^{(2)}(\omega,\Omega)$ and/or to $\omega_{diff}=\omega-\Omega$ 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 *(see the chapter "Nonlinear Optics Basics: Cascading" by G. Assanto and G. Stegeman in this encyclopedia)*. 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.

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Figure Captions

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.

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.

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 nondegenrate nonlinear absorption coefficient including all major mechanisms.



(a)

(b)





Figure 2



Figure 3



Key Words:

Kramers-Krönig, Nonlinear Optics, Nonlinear Susceptibility, Causality, Dispersion Relations, Nonlinear Absorption, Nonlinear Refraction, Two-Photon Absorption, Optical Kerr Effect.

Nomenclature:

KK relations	Kramers-Krönig relations
χ ⁽ⁿ⁾	n-th order nonlinear optical susceptibility
α	linear absorption coefficient
n	linear refractive index
α_2	nonlinear absorption coefficient
n ₂	nonlinear refractive index coefficient, coefficient of optical Kerr effect
β	two-photon absorption coefficient
2PA	two-photon absorption
<i>f</i> D	principal value
$\Theta(t)$	step-function