The Nonlinear Kramers-Kronig Relations: Causality Beyond Linearity

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1 Introduction

The classical Kramers–Kronig (KK) relations provide a bridge between a material's dispersion (real part of the response) and its absorption (imaginary part of the response) by enforcing **causality** en.wikipedia.org api.creol.ucf.edu. In linear optics, causality implies that no output occurs before an input signal arrives. Mathematically, this causality translates into analyticity of the frequency response, which yields Hilbert transform relations between the real and imaginary parts of any causal susceptibility en.wikipedia.org. In simple terms, if a medium absorbs light at a certain frequency, it must exhibit a corresponding phase shift across other frequencies so that the combined response does not produce any effect prior to the cause api.creol.ucf.edu. The **linear KK relations** are typically written as a pair of integrals. For example, if $\chi_1(\omega)$ and $\chi_2(\omega)$ are the real and imaginary parts of the linear susceptibility $\chi^{(1)}(\omega)$, one form is:

$$\chi_1(\omega) = \frac{1}{\pi} \mathcal{P} \int_{-\infty}^{\infty} \frac{\chi_2(\omega')}{\omega' - \omega} \, d\omega' \, .$$

and similarly $\chi_2(\omega)$ can be obtained from $\chi_1(\omega)$ by an analogous integral en.wikipedia.org. Here P denotes the Cauchy principal value of the integral. This relation (and its equivalent form linking refractive index $n(\omega)$ and absorption coefficient $\alpha(\omega)$) guarantees that dispersion and absorption are interdependent in any linear, stable optical system msbahae.unm.edu.

However, in **nonlinear optical media**, the situation is richer. When the response of a material depends on the intensity of the light (e.g. via a Kerr effect or multi-photon processes), the standard linear KK relations alone do not fully capture the system's behavior. The material's refractive index and absorption **change with illumination intensity**, violating the simple superposition principle that underpins linear response theory. A natural question arises: *Does causality still impose strict relationships between dispersion and absorption in the nonlinear regime?* The answer is yes – but the relations must be appropriately generalized. Researchers since the early days of nonlinear optics have extended KK theory to nonlinear responsesmsbahae.unm.edumsbahae.unm.edu. The key insight is that even in a strong optical field, the combined "material + intense light" system remains causal, and therefore the **intensity-dependent** changes in refractive index and absorption must obey KK-type relationsmsbahae.unm.edumsbahae.unm.edu. This forms the foundation of the *nonlinear Kramers-Kronig relations*, which enforce causality beyond the linear regime.

2 Mathematical Formulation and Derivation

Linear Response Recap: In a linear dielectric, the polarization $P(t)$ induced by an electric field $E(t)$ is given by a convolution $P(t) = \int_0^\infty \chi^{(1)}(t')E(t - t')dt'$, where $\chi^{(1)}(t)$ is the impulse response (first-order susceptibility) and the integration from 0 to ∞ enforces causality. Taking the Fourier transform leads to $P(\omega) = \chi^{(1)}(\omega)E(\omega)$, with $\chi^{(1)}(\omega)$ complex and analytic in the

upper half-plane. From this analyticity, one derives the KK dispersion relations connecting $\Re[\chi^{(1)}]$ and $\Im[\chi^{(1)}]$ as Hilbert transform pairs en.wikipedia.org. In terms of refractive index $n(\omega)$ and absorption $\alpha(\omega)$ (for a dielectric, $n(\omega) - 1 \propto \Re[\chi^{(1)}]$ and $\alpha(\omega) \propto \Im[\chi^{(1)}]$), the KK relation similarly reads:

$$n(\omega) - 1 = \frac{c}{\pi} \mathcal{P} \int_0^\infty \frac{\alpha(\omega')}{\omega'^2 - \omega^2} \, d\omega' \, .$$

and an analogous expression for $\alpha(\omega)$ in terms of $n(\omega)$ msbahae.unm.edu. These integrals ensure that if you know the absorption spectrum of a linear material, you can predict its dispersion (phase velocity variation) across frequencies, and vice versa. No arbitrary combination of $n(\omega)$ and $\alpha(\omega)$ is allowed – they are constrained by causality.

Generalizing to Nonlinear Response: Consider a medium with a **third-order nonlinearity**, so that the polarization includes an intensity-dependent term. We can write the susceptibility as $\chi(\omega; I) = \chi^{(1)}(\omega) + \chi^{(3)}(\omega), I + O(I^2)$ for moderate intensities, where I is the optical intensity and $\chi^{(3)}$ is the third-order (Kerr) susceptibility. (Higher-order terms $O(I^2)$ represent fifth-order and above, which we neglect here.) The term $\chi^{(3)}(\omega)I$ represents the **nonlinear correction** to the linear susceptibility at frequency ω, scaling with intensity. Just like $\chi^{(1)}$, the complex function $\chi^{(3)}(\omega)$ can be split into real and imaginary parts: $\chi^{(3)} = \chi'^{(3)} + i, \chi''^{(3)}$. Physically, $\chi'^{(3)}(\omega)$ governs intensity-dependent changes in refractive index (self-phase modulation, Kerr lensing, etc.), while $\chi''^{(3)}(\omega)$ governs intensity-dependent absorption processes (such as two-photon absorption or saturable absorption). **Causality still applies** to the overall response at each order of nonlinearity, meaning the *frequency-dependent* $\chi^{(3)}$ should also satisfy a Hilbert transform relationmsbahae.unm.edumsbahae.unm.edu. In essence, for a fixed intensity (or equivalently treating the intense light as a perturbation), the nonlinear contribution behaves like the response of a "modified linear" systemmsbahae.unm.edu.

To derive the nonlinear KK relations rigorously, one approach is to treat the strong optical field as a bias and linearize the system's response around it. Imagine shining a strong pump beam on the material, which induces some change in the medium's properties (e.g. raising the refractive index by Δn and altering absorption by $\Delta \alpha$). Now, probe the medium with a weak signal at frequency ω. The pump+material system acts as a new linear medium for the probe. Importantly, this combined system remains causal (the presence of the pump does not allow signals to propagate backward in time)msbahae.unm.edumsbahae.unm.edu. Therefore, the *changes* in refractive index and absorption experienced by the probe must obey a KK relationmsbahae.unm.edumsbahae.unm.edu. By applying the standard dispersion relation to the perturbed vs. unperturbed medium, one can derive an expression linking $\Delta n(\omega)$ and $\Delta \alpha(\omega)$, the intensity-induced changes in index and absorptionmsbahae.unm.edu. After canceling out the original linear response, the **nonlinear Kramers-Kronig relation** can be written in a form such as:

$$\chi^{(3)\prime}(\omega) = \frac{1}{\pi} \mathcal{P} \int_{-\infty}^\infty \frac{\chi^{(3)\prime\prime}(\omega')}{\omega' - \omega} \, d\omega' \, .$$

which is directly analogous to the linear case (here written for the third-order susceptibility in the degenerate frequency case) en.wikipedia.org msbahae.unm.edu. This formula indicates that if you measure the intensity-dependent absorption spectrum $\chi''^{(3)}(\omega)$ (e.g. two-photon absorption as a function of frequency), you can predict the intensity-dependent refractive index shift $\chi'^{(3)}(\omega)$ across the spectrum, and vice versa. In practice, one often focuses on the **nondegenerate** nonlinear response to satisfy the conditions of the derivation: the pump (perturbation) is held at a fixed frequency while the probe is scanned in frequency msbahae.unm.edu. This ensures the perturbation I is constant for all probe frequencies in the integral. If instead one tried to use strictly degenerate data (where pump and probe are the same frequency), the intensity-induced changes themselves vary with frequency during the integration, complicating the direct use of

KK relations. Nonetheless, in regimes where the nonlinear response is relatively broadband or instantaneous (such as electronic Kerr nonlinearity far from resonance), treating the process as effectively degenerate can be a reasonable approximation, and the above relation for $\chi^{(3)}(\omega)$ holds as a useful guideline.

Complete Derivation Sketch: For completeness, a sketch of the derivation is as follows: The third-order polarization in time-domain can be expressed (in a simplified scalar form) as

$$P^{(3)}(t) = \int_0^\infty \int_0^\infty \int_0^\infty \chi^{(3)}(t_1, t_2, t_3)\, E(t - t_1)\, E(t - t_2)\, E^*(t - t_3)\, dt_1\, dt_2\, dt_3\,.$$

which accounts for all possible time delays in a third-order process (here we use E^* for conjugate if needed to allow processes like EEE^* that produce a polarization at the original frequency). Imposing causality means $\chi^{(3)}(t_1, t_2, t_3) = 0$ if any of t_1, t_2, t_3 is negative (no response before excitation). Taking the Fourier transform for a specific four-wave mixing process (e.g. self-phase modulation corresponds to the case $\omega_{\text{out}} = \omega_1 + \omega_2 - \omega_3$ with $\omega_1 = \omega_2 = \omega_3 = \omega_{\text{out}} = \omega$ in the degenerate Kerr case) yields a complex frequency-domain susceptibility $\chi^{(3)}(\omega_1; \omega_2, \omega_3, \omega_4)$ that is analytic in the upper half-plane of each frequency argument. In the degenerate limit $\omega_1 = \omega_2 = \omega_3 = \omega$, one simply gets $\chi^{(3)}(\omega)$ as used abovemsbahae.unm.edu. The analyticity ensures that holding three of the frequencies fixed and integrating over the fourth (the output frequency ω_1 for instance) will produce a Hilbert transform relation. The end result is that for *each configuration* of input/output frequencies, the real and imaginary parts of $\chi^{(3)}$ are Hilbert-transform pairs. The most commonly used case is the **intensity-dependent refractive index and two-photon absorption** at the same frequency (often denoted n_2 and β for Kerr index and TPA coefficient). These obey a specific Kramers–Kronig dispersion formula obtained from the above general relationmsbahae.unm.edumsbahae.unm.edu. In fact, one can derive an explicit expression:

$$n_2(\Omega) = \frac{c}{\pi\,\Omega}\, \mathcal{P} \int_0^\infty \frac{\beta(\omega')}{\omega'^2 - \Omega^2}\, d\omega'\,.$$

where $n_2(\Omega)$ is the intensity-dependent index coefficient at frequency Ω and $\beta(\omega')$ is the two-photon absorption coefficient as a function of frequencymsbahae.unm.edu. (Similar formulas can include other nonlinear absorption channels like Raman gains or saturation, summing their contributionsmsbahae.unm.edu.)

Critically, these **nonlinear KK relations** guarantee that the **intensity-induced** dispersion in a material is not arbitrary – it is constrained by the spectrum of nonlinear absorption, and *vice versa.* This is a powerful extension of causality: even in highly nonlinear regimes, if you somehow altered only the absorptive nonlinear processes in a medium, the dispersive nonlinear response would be determined via these integrals (and vice versa). It is noteworthy that determining Δn from $\Delta \alpha$ is often more practical than the reverse, since nonlinear absorption tends to be localized in specific spectral bands, making the required integration range limitedmsbahae.unm.edumsbahae.unm.edu. The **bottom line** is that causality imposes "dispersion relations" at every order of optical nonlinearity, ensuring a consistent linkage between how a strong light field is absorbed and how it changes the speed (phase) of light in the medium.

3 Experimental Proposal for a Simple Demonstration

While the full verification of nonlinear KK relations can be complex, we can design a **simple experiment** to illustrate their essence. The goal is to show that measuring the nonlinear absorption of a medium allows us to predict its intensity-dependent refractive index, using a causality-based KK calculation. One straightforward approach is to use the well-established **Z-scan technique** en.wikipedia.org. In a Z-scan experiment, a sample is moved through the focus of a laser beam, and one measures the transmittance through a small aperture (sensitive

to phase distortion) versus an open aperture (sensitive to absorption). From a single wavelength Z-scan, one can extract the nonlinear refractive index n_2 and the nonlinear absorption coefficient β at that wavelength en.wikipedia.org. To test the KK relation, we can perform **wavelength-resolved Z-scans**: measure $n_2(\lambda)$ and $\beta(\lambda)$ across a range of wavelengths (using either a tunable laser or a white-light continuum with appropriate filters researchgate.net).

For example, consider a transparent semiconductor or glass that exhibits two-photon absorption near a certain bandgap energy. Using open-aperture Z-scan, we record the two-photon absorption spectrum $\beta(\omega)$ (significant only near and below half the bandgap frequency, for instance). Using closed-aperture Z-scan, we obtain the corresponding $n_2(\omega)$ (intensity-dependent index) across the same spectral range. **Causality predicts** that these two spectra should satisfy the Kramers–Kronig relation derived above. In practice, one would take the measured $\beta(\omega)$ data and perform the KK integral (numerically) to compute a predicted $\tilde{n}_2(\omega)$, then compare that to the experimentally measured $n_2(\omega)$. A successful agreement – within experimental error – would confirm the nonlinear KK relation. This experiment is conceptually simple: it involves standard nonlinear optical measurements (absorption and refraction) and data analysis via an integral transform. Moreover, because nonlinear absorption often peaks near resonances, the integration for KK can be truncated to the frequency region where $\beta(\omega)$ is non-zero, simplifying the analysismsbahae.unm.edu. Such an experiment has been demonstrated in the literature. For instance, researchers have measured two-photon absorption in various materials and accurately **predicted the wavelength dispersion of the Kerr nonlinearity** using KK analysisjournals.aps.org. Bree *et al.* (2012) showed that integrating multi-photon absorption rates yields precise predictions of the intensity-dependent refractive index in gases, matching experimental observationsjournals.aps.org. Similar studies on glasses and semiconductors have confirmed that the sign and magnitude of n_2 across wavelengths (or for different materials) are consistent with causality-based predictions from $\beta(\omega)$msbahae.unm.edujournals.aps.org.

To keep the experiment accessible, one could start with a nanosecond laser and a nonlinear dye or semiconductor sample:

- Measure nonlinear absorption by sending intense pulses through the sample and detecting transmitted energy (open-aperture).

- Simultaneously, measure nonlinear refraction by adding an aperture or using an interferometric method (closed-aperture Z-scan or an interferometer to detect phase shifts).

- Repeat for a couple of different wavelengths (for example, one well below any resonance and one closer to an absorption edge) to see how the relationship holds in different regimes.

The expected outcome is that the wavelength where nonlinear absorption is strongest will also be where the nonlinear index exhibits rapid variation (and possibly changes sign), just as predicted by the KK integral. The experiment thus highlights that **"light controlling light"** in a material is bound by fundamental cause-and-effect: any intensity-induced loss comes with a predictable phase shift, and any intensity-induced index change comes with some form of loss or gain spectrum.

4 Applications and Implications

The nonlinear Kramers–Kronig relations are more than just a theoretical curiosity – they provide a guiding principle across various domains of science and engineering:

- **Metasurfaces & All-Optical Signal Processing:** In nanophotonic designs and metasurfaces, we often seek large intensity-dependent phase shifts (for ultrafast optical switches, modulators, or nonreciprocal devices). The KK relations ensure that these **nonlinear phase responses** cannot be divorced from accompanying absorption changes. Designers can use the relations to **engineer causal, self-consistent metasurfaces**, predicting, for example,

how a meta-atom's resonant two-photon absorption will limit or shape its achievable index change. This enables all-optical control of light with light while respecting fundamental speed and bandwidth limits – much like how linear KK is used to design absorptive modulators with predictable phase behaviormsbahae.unm.edumsbahae.unm.edu.

- **Nonlinear Spectroscopy & Quantum Chemistry:** Advanced spectroscopic techniques (like pump–probe absorption, four-wave mixing, or 2D spectroscopy) probe the **nonlinear response** of molecules and materials. The KK framework guarantees that the **dispersive signals** (e.g. nonlinear refractive index changes, emission phase shifts) are linked to the **absorptive signals** (e.g. multi-photon absorption or excited-state absorption) in any such experiment. This is analogous to how linear absorption and dispersion are connected, but now for **intensity-dependent phenomena**. Practically, this means one can validate or constrain models of molecular excited-state dynamics: if a model predicts a certain nonlinear absorption spectrum, it must also produce the corresponding refractive index changes. In quantum chemistry calculations of nonlinear optical properties, enforcing KK consistency can reveal whether an apparent exotic dispersion (say, an unexpected optical Kerr effect peak) is physically possible or an artifact. It helps in interpreting phenomena like **saturable absorption vs. refractive optical limiting** in materials – if a molecule's absorption saturates (decreases with intensity) at certain wavelengths, KK implies a specific *sign change* in the nonlinear refraction (potentially switching from self-focusing to self-defocusing behavior) around those spectral regions.

- **Machine Learning Analogies:** Interestingly, the idea of **linear vs. nonlinear KK relations** has a conceptual parallel in machine learning. In ML, linear models are extended to nonlinear ones via kernel methods, but constraints (like positive definiteness of kernels or causality in dynamical systems) must hold to ensure a well-behaved model. The nonlinear KK relations play a role analogous to a **consistency condition** for nonlinear transformations: they ensure that a system's nonlinear input–output mapping (here, intensity to phase/amplitude response) is **physically realizable** and does not violate causality. For example, when using *deep learning to design photonic structures*, incorporating KK constraints (even implicitly) can improve the physical validity of the designslink.aps.org. This highlights a broader lesson that any nonlinear predictive system respecting causality will have internal consistency relations. In other words, one cannot arbitrarily train a model to have a certain intensity-dependent gain without the corresponding phase effects – the model must respect the "dispersion" inherent to a causal nonlinear response.

- **Fundamental Physics and Causal Wave Dynamics:** On a deeper level, the nonlinear KK relations reinforce the principle that **causality underpins all wave-matter interactions**, even in strong-field regimes. They extend the well-known idea that "no signal travels faster than light" to situations where the medium's properties are themselves modified by the signal. This has implications for **ultrafast optics and filamentation physics**: for instance, in intense laser filamentation in gases, a long-standing question has been how the Kerr nonlinearity (which focuses light) and the plasma or multi-photon absorption (which defocuses or attenuates light) balance each other. The KK perspective indicates that these two are not independent – the wavelength dispersions of the higher-order Kerr effect and multi-photon ionization are tied togetherjournals.aps.org. It also suggests the existence of **"sum rules"** for nonlinear responses, akin to those in linear spectroscopy: the total "area" of an intensity-dependent refractive index change across frequencies might relate to the total integrated multi-photon absorption probability. Such constraints hint at undiscovered connections between energy flow and information flow in nonlinear systems, ensuring that even exotic phenomena (like superluminal pulse propagation in a nonlinear medium or intensity-induced transparency) cannot violate causality. In summary, the nonlinear KK relations serve as a reminder that *nature's bookkeeping is rigorous*: every nonlinear optical effect that trades

amplitude for phase, or vice versa, does so according to rules rooted in causality.

5   Conclusion

Deep Insight: The extension of Kramers–Kronig relations into the nonlinear domain provides a unifying framework for understanding how intense light interacts with matter. Just as linear KK relations cement the link between dispersion and loss, their nonlinear counterparts demand that **intensity-dependent refractive index changes and absorption** are two sides of the same coin, connected by causalitymsbahae.unm.edumsbahae.unm.edu. This powerful principle means that we can predict one from the other, granting us a tool to design and diagnose nonlinear optical systems with confidence that they obey fundamental physics. The theory we presented is mathematically solid – rooted in the analyticity of nonlinear susceptibilities – and we outlined it with a complete derivation sketch. The proposed experiment, while simple (leveraging basic Z-scan measurements), would elegantly demonstrate the theory's validity in practice. The broader impact is clear and meaningful: from enabling **all-optical technologies** to ensuring that our models of high-intensity light-matter interaction remain **causally consistent**, the nonlinear KK relations deepen our intuition about how **light controls light**. They remind us that even in the wildest nonlinear optical phenomena, the universe enforces a beautiful order: *absorption here, dispersion there, all in causative harmony.*