Causality-Based Criteria for a Negative Refractive Index Must Be Used With Care

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Using the principle of causality as expressed in the Kramers-Kronig relations, we derive a generalized criterion for a negative refractive index that admits imperfect transparency at an observation frequency ω. It also allows us to relate the global properties of the loss (i.e., its frequency response) to its local behavior at ω. However, causality-based criteria rely on the group velocity, not the Poynting vector. Since the two are not equivalent, we provide some simple examples to compare the two criteria.

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Remarkable electromagnetic properties can be seen in materials engineered so that the phase velocity of electromagnetic-wave propagation opposes the electromagnetic power flow; such materials are often called "left handed" (see, e.g., [1–9]), but are more precisely described as negative phase velocity (NPV) media. As might be expected in a rapidly evolving field of research, a variety of conditions [10–12] for NPV have been proposed in the literature. The presence of a negative refractive index (NRI) allows for a number of intriguing possibilities: e.g., the creation of a "perfect lens" that produces an undistorted image without causing any surface reflections [13], the possibility of a reversed Casimir force being used to levitate ultrathin mirrors [14], the automatic compensation of dissipation or dispersion to enhance quantum interference [15], or the possibility of "trapped rainbow" light storage [16].

The dispersive nature of the effective medium parameters is exploited in metamaterials to produce a NRI, as confirmed through experimental, theoretical, and numerical studies [2–9,17]. Such metamaterials therefore inherit unavoidable losses on the grounds of causality. Since losses can cause a significant drop in performance, a key challenge is to successfully compensate for loss by adding gain, but note that care must be taken in theoretical investigations to ensure that the gain model is both stable and causal [18,19], and we also need to use the correct NPV criterion [20]. Here we specifically address the role of the losses required by causality by considering the famous Kramers-Kronig (KK) relations (see, e.g., [21]), which control the relationship between the real and imaginary parts of the electric and magnetic material responses (i.e., the permittivity ϵ and permeability μ). Such relations can also be established for the square of the refractive index $n^2 = c^2\epsilon\mu$, as this quantity inherits the analytical properties of $\epsilon(\omega)$ and $\mu(\omega)$; i.e., it lacks singularities in the upper half-plane of complex ω, and $n^2(\omega) \rightarrow 1$ as $\omega \rightarrow \infty$ (see, e.g., [18,21]). In a recent Letter, Stockman [22] adapted the KK relation on n^2 to place limits on the *minimum* losses that accompany NRI for a medium which is perfectly transparent at the observation frequency. He concluded that any significant reduction in the losses near the chosen observation frequency will also eliminate the NRI. Whether real metamaterials can *in principle* be made with low loss is a question of utmost importance in practical metamaterial design. Previous work [22] claimed that the answer is emphatically negative, but we show here that the answer is actually affirmative.

In this Letter we replace Stockman's zero-loss criterion with another causality-based criterion, one capable of giving useful answers for NPV propagation because it admits arbitrary linear optical losses both at and away from the observation frequency. Here we assume a homogeneous medium with ϵ and μ being effective parameters obtained for the composite metamaterial by, e.g., a modified S-parameter technique [23,24]. Such effective medium approaches are less reliable in the short wavelength (high frequency) regime, but existing analytic attempts only apply to (at best) thin composite layers [25]. The KK relation for n^2 can be written

$$\mathrm{Re}\,(n^2) - 1 = \frac{2}{\pi}\mathcal{P}\int_0^\infty \frac{\mathrm{Im}(n^2)}{s^2-\omega^2} s ds, \tag{1}$$

where Re() and Im() take the real and imaginary parts; thus, $\mathrm{Im}(n^2) = \epsilon'\mu'' + \epsilon''\mu'$, where ϵ' and μ' are the real parts of ϵ and μ, with the imaginary parts being ϵ'' and μ''. $\mathcal{P}$ takes the Cauchy principal value. Noting that the loss in the material is important in the calculation of $\mathrm{Im}(n^2)$, Stockman transformed Eq. (1) into one relating the material loss to the presence of NRI,

$$\frac{c^2}{v_p v_g} = 1 + \frac{2}{\pi}\int_0^\infty \frac{\mathrm{Im}(n^2(s))}{(s^2-\omega^2)^2} s^3 ds < 0, \tag{2}$$

by applying the operation $\mathcal{L} = \omega^{-1}\partial_\omega\omega^2$ to both sides of Eq. (1). The behavior of several experimental systems was claimed in [22] to be consistent with this criterion. Here the NRI condition relies on opposed (real valued) phase and group velocities, i.e., $v_p v_g < 0$. Since the usual acronym NPV is ambiguous, we refer to $v_p v_g < 0$ as NPVG (i.e., NPV with respect to group velocity); the usual case [26] is then NPVE (i.e. NPV with respect to energy velocity).

When $v_p v_g < 0$, the integral on the right-hand side (RHS) of Eq. (2) must be negative. Stockman therefore concluded that even if the losses vanish at the observation frequency, there must still be significant loss nearby, otherwise the integral will produce a positive result. Consequently, systems with imperfect tuning or an insufficiently narrow operating bandwidth would have their performance degraded.

The limitations of Eq. (2) are threefold.

(1) $\mathrm{Im}(n^2)$ and its derivative must be exactly zero at the observation frequency ω—otherwise the integral diverges and the constraint becomes uncertain.

(2) It only applies at a particular observation frequency, despite utilizing the global properties of the material response. It can be used to infer the presence of nearby loss, but does not indicate whether NRI is present there.

(3) The NPVG condition $v_p v_g < 0$ is not equivalent to the NPVE condition $\vec{P}\cdot\vec{k} < 0$.

These limitations make it hard to determine how losses might be minimized while still maintaining NPV over some frequency window.

We now replace Stockman's criterion with one that avoids divergences while allowing for nonzero loss, thus removing the first two limitations given above. All necessary convergence or limiting properties for n^2 can be satisfied if ϵ and μ are described by functions of s which are both rational and causal. If necessary, inconvenient singularities or divergences in $n^2(\omega)$ can be removed by considering $f(\omega)n^2(\omega)$, where f is some rational function of ω designed to cancel the pole or to remove the divergence [28]. The third limitation is intrinsic to the approach, but has the advantage that it also enables us to evaluate the presence of NRI (or, strictly, the presence of NPVG) using causality. Here the group velocity v_g amounts to the commonly used $\partial_\omega k(\omega)$; although imperfect in the case of loss or gain (see, e.g., [29]), it is that which follows most naturally here.

Our first step is to integrate the RHS of Eq. (1) by parts, but only after splitting it into two pieces covering the ranges $[0, \omega - \sigma)$ and $(\omega + \sigma, \infty)$, then taking the limit $\sigma \to 0$ at the end. After defining $Q_j = \partial_s^j \mathrm{Im}(n^2(s))$, and with $Q_0(s) = \mathrm{Im}(n^2(s))$ tending to zero fast enough so that the $s = \infty$ surface term vanishes, we find that

$$\int_0^\infty \frac{Q_0(s)}{s^2 - \omega^2} s ds = -\int_0^\infty \frac{Q_1(s)}{2} \ln\left|1 - \frac{s^2}{\omega^2}\right| ds. \quad (3)$$

Since the RHS is independent of Q_0, we can now obtain a criterion valid where loss is present at the observation frequency, and the better behaved integrand means it is considerably easier to make inferences about the presence of NRI. After applying $\mathcal{L}$, we find that $v_p v_g < 0$ requires

$$\pi \le \int_0^\infty Q_1(s)\left[\ln\left|1 - \frac{s^2}{\omega^2}\right| + \frac{s^2}{\omega^2 - s^2}\right] ds. \quad (4)$$

After a second integration by parts, and taking $\sigma \to 0$, the surface terms at $\omega - \sigma$ and $\omega + \sigma$ will again cancel; those at 0 and ∞ vanish. With $z = s/\omega$, we now have

$$\pi \le \omega \int_0^\infty Q_1(z\omega) \ln|1 - z^2| dz - \omega^2 \int_0^1 \left[Q_2(\omega z) + \frac{1}{z^2} Q_2\left(\frac{\omega}{z}\right)\right] \tanh^{-1}(z) dz. \quad (5)$$

The first (Q_1) term can again be integrated by parts; however, the result adds little insight.

Equation (5) is the most general causality-based criterion achievable, and is, crucially, not restricted to points of perfect transparency. It depends only on how the loss [as specified by $Q_0 = \mathrm{Im}(n^2)$] changes with frequency (i.e., on its dispersion, as given by Q_1 and Q_2), and not on its magnitude. Notably, the sign of Q_1 (i.e., whether Q_0 is increasing or decreasing with frequency) has a strong effect on the presence of NRI, as does the sign of Q_2 (crudely, whether Q_0 is near a minimum or maximum).

The non-Q_i parts of the integrands [i.e., $\ln|1 - z^2|$ and $\tanh^{-1}(z)$] are both strongly peaked at $s = \omega$, but nevertheless have finite integrals. Using the expansion $\mathrm{Im}(n^2(s)) \simeq Q_0(\omega) + xQ_1(\omega) + (x^2/2)Q_2(\omega)$, for $x = s/\omega - 1$, we can integrate Eq. (5) analytically in an attempt to obtain an approximate criterion

$$\pi \lesssim -1.34\omega Q_1(\omega)[1 - 2\omega Q_2(\omega)] - 1.39\omega^2 Q_2(\omega). \quad (6)$$

Unfortunately this fails to convincingly match Eq. (5), and the attempt only succeeds in emphasizing that it is the *global* properties of the loss which constrain the presence of NRI. Only in Stockman's zero-loss case might simple intuition be valid.

Using our causality-based criterion in Eq. (5), we can now try to infer whether the global properties of $\mathrm{Im}(n^2)$ promote (or hinder) NPVG. Since $v_p v_g$ and Eq. (5) are intimately connected by the KK relations for n^2, we used this to numerically test the examples below; nevertheless, each expression provides its own unique perspective—one local, one global. Since we may not always be able to rely on obtaining n^2 from a model (as in [23,24]), we may need to recover it from experimental data. While the standard KK relations are prone to generating inaccurate reconstructions, approaches such as the multiply subtractive KK method can resolve this for many practical applications—even nonlinear spectroscopy [30].

We now proceed to test our causality-based criterion. Since we wish to emphasize general principles, and ensure the points we make are clear, we consider simple examples with $\epsilon = \mu$, rather than more complicated systems. We also normalize with respect to some suitable reference frequency. The condition natural to the approach used here is the NPVG one (i.e., $\chi_G = v_p v_g < 0$). This means we only need to calculate (and show) one of χ_G or Eq. (5); we label the result χ_G. In contrast, the NPVE condition requires that the phase velocity is opposed to the energy velocity. This occurs if [12,20]

$$\chi_E = \epsilon' |\mu| + \mu' |\epsilon| < 0. \tag{7}$$

The two conditions ($\chi_G < 0$ and $\chi_E < 0$) will agree if the group velocity v_g and energy velocity v_E have the same sign. However this only holds in the limit of nearly undistorted pulse propagation [31], i.e., for small dispersion and loss. This is likely to be a poor approximation in NRI materials, which by their nature rely on strong dielectric or magnetic response. So although our criterion in Eq. (5) can always be used to judge the presence of NPVG, and make inferences thereon, this is not strictly equivalent to the presence of NPVE.

Our first example is a simple double-plasmon resonance, as in, e.g., [32], setting ϵ and μ according to

$$\frac{\epsilon(\omega)}{\epsilon_0} = \frac{\mu(\omega)}{\mu_0} = 1 - \frac{\omega_p^2}{\omega(\omega + \imath\gamma)}. \tag{8}$$

A simple test to evaluate the presence of NPVG [and at the same time test our generalized causality-based criterion in Eq. (5)] is to increase the losses while comparing it against the NPVE condition. The results can be seen in Fig. 1, where $\chi^{1/3}$ is plotted to accommodate the vertical range. For sufficiently weak losses ($\gamma \ll \omega$) the criteria agree, with both the χ_E and χ_G curves remaining below zero. However, as the losses get stronger, the χ_G and χ_E start to disagree. Nevertheless, we can see that in the preferred region of $\omega \simeq 1$, where $\epsilon = \mu \simeq -1$, they disagree only for very large losses. Here the χ_G criterion works relatively well because the plasmonic responses vary both smoothly and monotonically; hence, v_g does not change sign and remains in accord with v_E.

Our next example is again motivated by simplicity, but also by the possibility of creating NRI in atomic gases. In a gas, it is possible to design pumping schemes that create gain [33,34], but the freedom to manipulate the optical properties relies mainly on the dielectric response (ϵ). Here we consider two matched pairs of Lorentz resonances, so that $\epsilon(\omega) = \mu(\omega)$, and

$$\frac{\epsilon(\omega)}{\epsilon_0} = 1 + \frac{\sigma_1\omega_1^2}{\omega_1^2 - \omega^2 - \imath\omega_1\gamma_1} + \frac{\sigma_2\omega_2^2}{\omega_2^2 - \omega^2 - \imath\omega_2\gamma_2}. \tag{9}$$

We focus on a dominant lossy resonance ($\sigma_1 < 0$), with a weaker, offset, *active* resonance ($\sigma_2 > 0$) providing sufficient gain to induce near transparency at a chosen observation frequency [35]. ϵ and μ are chosen equal apart from a scale factor ϵ_0/μ_0, and are shown in Fig. 2(a), where we see that near transparency has been achieved at the cost of

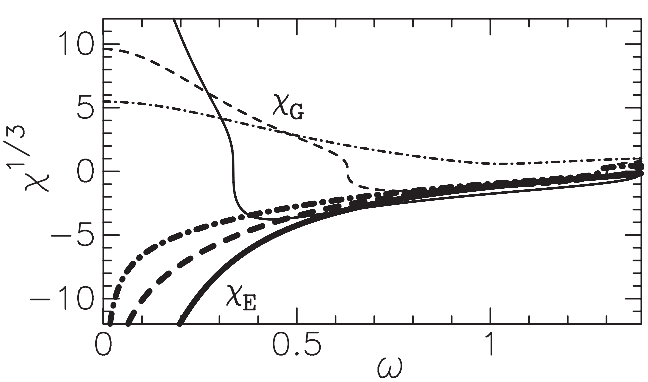

FIG. 1. A double (ϵ and μ) plasmon system exhibiting NRI, with both plasma frequencies being $\omega_p \simeq 1.4$. It compares the (χ_E) NPVE condition (thick lines) to the (χ_G) NPVG one (thin lines). The results shown are for $\gamma = 0.02$ (solid lines), $\gamma = 0.04$ (dashed lines), and $\gamma = 0.06$ (dot-dashed lines).

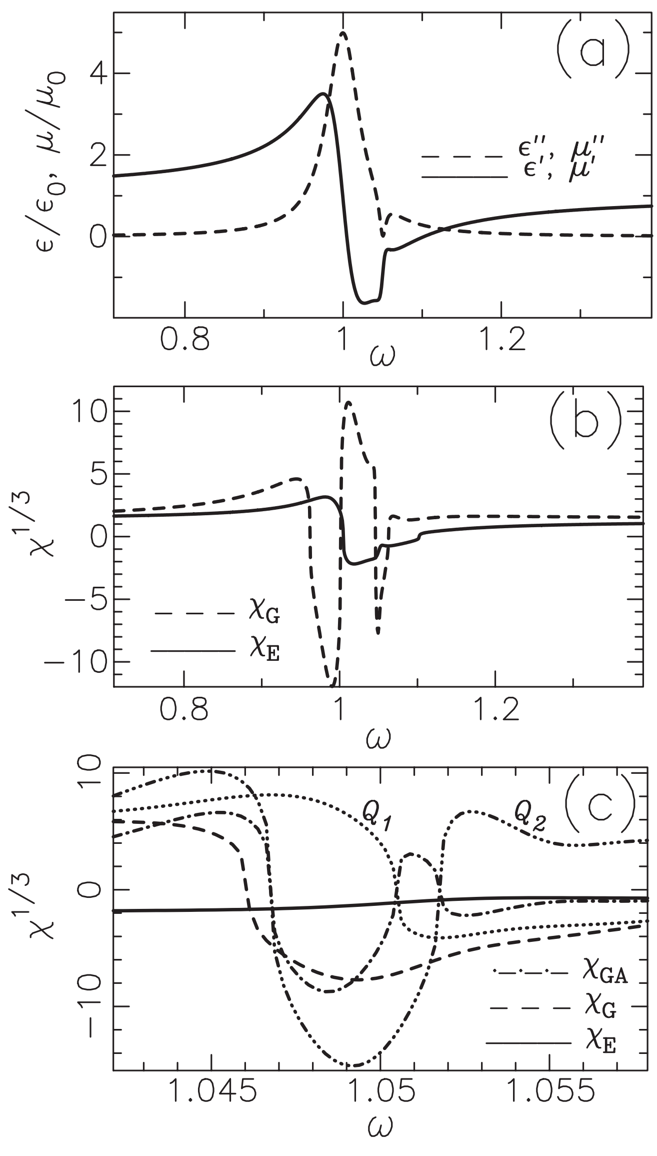

FIG. 2. A system exhibiting narrow band NRI. It combines a lossy resonance at $\omega_1 = 1$ (with $\gamma_1 = 0.05$, $\sigma_1 = -5$) and an active one at $\omega_2 = 1.05$ (with $\gamma_2 = 0.01$ and $\sigma_2 = 1.02$). (a) The real parts and imaginary parts of ϵ and μ. (b) Comparison of the NPVE (χ_E) and NPVG (χ_G) criteria. (c) Expanded view around $\omega_2 = 1.05$, showing also the NPVG approximation from Eq. (6) (labeled χ_{GA}), and Q_1 and Q_2.

increased dispersion, with ϵ' varying strongly where $\omega \simeq \omega_2$. Note how the sign of χ_G swaps back and forth according to the gradients of ϵ' and μ', even though the values of ϵ' and μ' themselves change very little: the utility of the χ_G criterion depends entirely on whether v_g has the same sign as v_E at the frequency of interest.

The narrow band region of low loss in this system makes it ideal for examining our NPVG criterion of Eq. (5) in more detail. First, note that there is an asymmetry about the loss minimum: below, the two contributions to Eq. (5) reinforce to help satisfy the criterion; above they partly cancel, making NRI less likely. This asymmetry is visible in Figs. 2(b) and 2(c) around $\omega = 1.05$. At the minimum itself, we can expect the Q_1 integral to be small since the integrand near ω will not only be small but odd; the behavior will then be dominated by that of Q_2—and indeed in Fig. 2(c) there is strong qualitative agreement between Q_2 and χ_G. The criterion therefore controls the width of allowed low-loss windows: a narrow band window will have a large Q_2, so that our criterion will be more easily satisfied. This inference is related to Stockman's—it also demands sufficient loss close to the observation frequency, but does not require $Q_0 = Q_1 = 0$.

In conclusion, we have derived a causality-based criterion for NRI allowing for frequency dependent (dispersive) losses at the observation frequency. Our new criterion is applicable to any medium with the linear response, required by the Kramers-Kronig relations. We investigated our causality-based criterion using some simple material response models, showing that since the group velocity v_g does not always match signs with the energy velocity v_E, the NPVG and NPVE forms of NRI are not equivalent. Since NPVE (i.e., $\vec{P} \cdot \vec{k} < 0$) is usually the preferred condition for NRI, this difference needs to be taken into account before causality-based NRI conditions are utilized. Nevertheless, our causality-based NPVG criterion provides unique insight into how the global response of the material affects its local performance.

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