Causality, Nonlocality, and Negative Refraction

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The importance of spatial nonlocality in the description of negative refraction in electromagnetic materials has been put forward recently. We develop a theory of negative refraction in homogeneous and isotropic media, based on first principles, and that includes nonlocality in its full generality. The theory shows that both dissipation and spatial nonlocality are necessary conditions for the existence of negative refraction. It also provides a sufficient condition in materials with weak spatial nonlocality. These fundamental results should have broad implications in the theoretical and practical analyses of negative refraction of electromagnetic and other kinds of waves.

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The study and design of artificial materials (metamaterials) with exotic electromagnetic properties has attracted a lot of attention at both the theoretical and experimental levels. Negative refraction has remained one of the most intriguing properties: In a certain range of frequencies, the energy flow is opposite to the direction of the phase velocity [1–5]. Local electrodynamics in continuous media, in terms of the electric permittivity $\epsilon(\omega)$ and magnetic permeability $\mu(\omega)$, has been the privileged framework to investigate the conditions for negative refraction [5]. The first metamaterials exhibiting negative refraction were severely limited by absorption, and various strategies have been explored to design almost transparent media. Nevertheless, the basic principles of electrodynamics impose general constraints on the response functions. In Ref. [6], the principle of causality has been used to argue that, for spatially local materials, dissipation is a necessary condition for negative refraction. Recently, it has been observed that local effective $\epsilon(\omega)$ and $\mu(\omega)$, obtained fitting reflectivity or transmissivity curves, usually exhibit physical inconsistencies, implying intrinsic flaws in the use of local electrodynamics to analyze negative refraction in certain artificial materials and the necessity to include spatial nonlocal corrections to overcome such inconsistencies [7–12]. Independently, it has been shown that spatial nonlocality is a fundamental ingredient for negative refraction in some specific materials [13,14].

Despite this progress, a theory providing a general understanding of the conditions for negative refraction, based on first principles and including spatial nonlocality, is still missing. The purpose of this Letter is to develop such a theory and to analyze its consequences in terms of necessary and sufficient conditions for the observation of negative refraction in natural and artificial electromagnetic materials. The analysis leads to the intriguing conclusion that negative refraction for electromagnetic waves is not possible in the absence of spatial nonlocality and dissipation. It also gives some insight into a sufficient condition to observe negative refraction in media with weak nonlocality. These results provide a sound basis both for the theoretical investigation of negative refraction in wave physics and for the design and analysis of real materials.

Maxwell's equations for electrodynamics in media can be written in full generality as [15–19]

$$\nabla\times\mathbf{B}=\mu_0\mathbf{j}_{\rm ext}+\mu_0\partial_t\mathbf{D},\qquad \nabla\cdot\mathbf{D}=\rho_{\rm ext},$$
$$\nabla\times\mathbf{E}=-\partial_t\mathbf{B},\qquad \nabla\cdot\mathbf{B}=0, \tag{1}$$

where $\mathbf{j}_{\rm ext}$ and $\rho_{\rm ext}$ are external current and charge densities, respectively, and μ_0 is the vacuum permeability. In the framework of the linear response theory, the field $\mathbf{D}$ is defined as [16–19]

$$D_i(t,\mathbf{r})=\epsilon_0\int_{-\infty}^{+t}\int_V\epsilon_{ij}(t,t',\mathbf{r},\mathbf{r}')E_j(t',\mathbf{r}')dt'd^3r' \tag{2}$$

with V the volume of the medium and ϵ_0 the vacuum permittivity. Implicit summation over repeated indices is assumed. The dielectric function $\epsilon_{ij}(t,t',\mathbf{r},\mathbf{r}')$ is proportional to the retarded correlator of the current density and encodes both the electric and magnetic response of the medium [15–19]. In this formalism, there is no need to introduce an additional field $\mathbf{H}$, provided that the full dispersive and nonlocal behavior of the dielectric function is taken into account [20]. For simplicity, we restrict the analysis to linear, isotropic, nongyrotropic, and homogeneous materials, and leave generalizations for further studies. Apart from this restriction, all results are derived from first principles, without any specification of a particular medium [21]. For translation-invariant media, $\epsilon_{ij}(t,t',\mathbf{r},\mathbf{r}')=\epsilon_{ij}(t-t',\mathbf{r}-\mathbf{r}')$, and in Fourier space Eq. (2) reads as

$$D_i(\omega,\boldsymbol{k})=\epsilon_0\epsilon_{ij}(\omega,\boldsymbol{k})E_j(\omega,\boldsymbol{k}), \tag{3}$$

where ω is the frequency and $\boldsymbol{k}$ is the wave vector that describes spatial nonlocality. All magnetic effects, such as the magnetic response usually described in terms of a local permeability $\mu(\omega)$, or even the magnetoelectric activity, can be encoded into specific spatial nonlocal properties of the dielectric function [15,17–19]. A discussion on this formalism, its relevance for a first-principles analysis and its relation with the local formalism are provided in the Supplemental Material [20].

$\epsilon_{ij}(\omega,\boldsymbol{k})$ has to satisfy various properties [16,18,22,23]. Causality implies that $\epsilon_{ij}(t-t',\mathbf{r}-\mathbf{r}')$ vanishes for $t<t'$. In the frequency domain, this means that $\epsilon_{ij}(\omega,\boldsymbol{k})$ is an analytic function of ω in the positive half part of the complex plane for which $\mathrm{Im}\omega>0$. This property leads to the Kramers-Kronig relations

$$\mathrm{Re}\epsilon_{ij}(\omega,\boldsymbol{k})-\delta_{ij}=\frac{2}{\pi}P\int_0^{+\infty}\frac{\omega'\mathrm{Im}\epsilon_{ij}(\omega',\boldsymbol{k})}{\omega'^2-\omega^2}d\omega',$$
$$\mathrm{Im}\epsilon_{ij}(\omega,\boldsymbol{k})=-\frac{2\omega}{\pi}P\int_0^{+\infty}\frac{\mathrm{Re}\epsilon_{ij}(\omega',\boldsymbol{k})-\delta_{ij}}{\omega'^2-\omega^2}d\omega',\tag{4}$$

where P stands for the principal value. It is important to highlight that the Kramers-Kronig relations are valid for all values of $\boldsymbol{k}$, that essentially plays the role of a parameter in the equations and is, at this stage, independent on ω. In isotropic media, the dielectric function takes the form

$$\epsilon_{ij}(\omega,\boldsymbol{k})=\epsilon_T(\omega,k)\left(\delta_{ij}-\frac{k_ik_j}{k^2}\right)+\epsilon_L(\omega,k)\frac{k_ik_j}{k^2},\tag{5}$$

where k_i are the components of $\boldsymbol{k}$ and $k=|\boldsymbol{k}|$ [24]. The response of the material is expressed in term of two scalar functions only, the transverse and the longitudinal dielectric functions $\epsilon_T(\omega,k)$ and $\epsilon_L(\omega,k)$, that both satisfy the Kramers-Kronig relations (4).

From the Kramers-Kronig relations and minor assumptions on the asymptotic behavior of $\epsilon_{T,L}(\omega,k)$ when $\omega\to\infty$, various sum rules can be derived [23,25] that encode important information and constraints on $\epsilon_{T,L}(\omega,k)$. Indeed, for a generic medium, for $\omega\to\infty$, $\epsilon_{T,L}(\omega,k)\to 1-\omega_p^2/\omega^2$ [18], where $\omega_p^2=ne^2/(m\epsilon_0)$ is the square of the plasma frequency, as if the electrons were free, n being the electron density of the material and e and m the charge and the mass, respectively, of the electron, while the imaginary part of $\epsilon_{T,L}(\omega,k)$ goes to zero faster than $1/\omega^2$ [25]; see [26]. An important sum rule is [25]

$$\int_0^{+\infty}\mathrm{Im}\epsilon_{T,L}(\omega',k)\omega'd\omega'=\frac{\pi\omega_p^2}{2}.\tag{6}$$

Note that Eq. (6) is valid for all values of k. Finally, for any passive medium and for real positive values of k and ω [27], the dielectric function satisfies [22,23]

$$\mathrm{Im}\epsilon_{T,L}(\omega,k)\geq 0.\tag{7}$$

Equations (4), (6), and (7) are fundamental relations that constrain the actual electrodynamic properties of any causal and passive medium.

The frequency ω and the wave vector k have been treated so far as independent variables; i.e., the system is considered "off-shell," as usual in the linear response theory [22,23] (due to the presence of external sources, ω and k do not have to satisfy any relation [16]). When considering the propagation of electromagnetic waves in a medium without external charges or currents, Maxwell's equations impose the dispersion relations

$$\epsilon_T(\omega,k)=\frac{c^2k^2}{\omega^2},\qquad \epsilon_L(\omega,k)=0\tag{8}$$

for the transverse and longitudinal components of the electric field $\mathbf{E}$, respectively, with c the speed of light in a vacuum. Equations (8) define a set of relations between the wave vector k and the frequency ω. These relations provide further restrictions to the electrodynamics in the medium. In the presence of spatial nonlocality, both the transverse and longitudinal components of the electric field can propagate. In this Letter, we focus on the response to transverse electromagnetic waves.

We shall first discuss the particular case of transparent media. In this case, negative refraction means that, in a certain range of frequencies, phase and group velocities have opposite directions. Indeed, both the electromagnetic energy density and the Poynting vector are well defined, the latter being simply the product of the electromagnetic energy density and the group velocity (the group velocity equals the energy velocity, at least for media with weak spatial nonlocality [28]). For local media, it has already been argued that negative refraction cannot be observed in the absence of dissipation [6,16]. We will now show that the same conclusion holds in the presence of spatial nonlocality, namely, that, for transverse waves, negative refraction is not possible in the frequency region of transparency of a medium.

In the transparency region $\mathrm{Im}\epsilon_{T,L}(\omega,k)=0$, and the Kramers-Kronig relations involve standard integrations. Deriving Eq. (4) with respect to the frequency [16,18] and using Eq. (7), we obtain

$$\frac{d[\mathrm{Re}\epsilon_{T,L}(\omega,k)]}{d\omega}\geq 0\tag{9}$$

for all frequencies in the transparency region (this is not the case in regions in which absorption cannot be disregarded, where the dielectric function can satisfy $d[\mathrm{Re}\epsilon_{T,L}(\omega,k)]/d\omega<0$, known as anomalous dispersion). For transverse waves, Eq. (8) implies $n_T^2[\omega,k(\omega)]=k^2c^2/\omega^2=\epsilon_T[\omega,k(\omega)]$, with n_T the transverse refractive index, and hence the dispersion relation $k(\omega)$. The transverse group velocity is defined as $v_g^T=d\omega(k)/dk=c[d(\omega n_T)/d\omega]^{-1}=c[n_T+\omega dn_T/d\omega]^{-1}$, while the phase velocity is $v_p^T=c/n_T$ [29]. Using Eq. (9), noticing that k and n_T are real functions of the frequency, we obtain $n_Tdn_T/d\omega>0$ or, equivalently,

$$v_g^T v_p^T > 0. \tag{10}$$

This implies that, as a consequence of causality, negative refraction for transverse waves is not possible in the transparency spectral region of a medium.

We shall now perform a general analysis of the conditions for negative refraction, including both dissipation and spatial nonlocality. For nontransparent media, a consistent definition of concepts such as energy density, energy flux, or heat dissipation rate is out of reach [16]. In particular, the group velocity does not define the direction of the energy flow. We have to find an alternative way to assess the direction of the energy flow based on the wave vector. To proceed, we use Eq. (8) to obtain the dispersion relation $k(\omega)$. Since we consider passive media, the direction of the energy flux is dictated by the direction of the dissipation of energy or, equivalently, of the damping of the wave amplitude, the latter being given by the sign of $\mathrm{Im}k$. The direction of the phase velocity is given by $\mathrm{Re}k$. As a result, a general condition for negative refraction is the existence of frequencies ω such that [30]

$$\mathrm{Re}[k(\omega)]\mathrm{Im}[k(\omega)] < 0, \tag{11}$$

meaning that the damping of the wave amplitude occurs in a direction opposite to the phase velocity.

We have demonstrated previously that negative refraction is not possible without dissipation. We shall now show that spatial nonlocality itself is a necessary condition for negative refraction. Let us consider a dissipative medium for which spatial nonlocality is assumed to be negligible. For transverse waves, we have $\epsilon_T(\omega,k) \equiv \epsilon^0(\omega)$, and the dispersion relation Eq. (8) simplifies into

$$k(\omega) = \frac{\omega}{c}\sqrt{\epsilon^0(\omega)}. \tag{12}$$

We choose to represent any complex number z as $z = \rho_z e^{i\phi_z}$, with $\rho_z \geq 0$ and $2\pi > \phi_z \geq 0$. Because of Eq. (7), $\mathrm{Im}\epsilon^0(\omega) \geq 0$ for real frequencies, and the phase ϕ_{ϵ^0} must satisfy $0 \leq \phi_{\epsilon^0} \leq \pi$. The phase of its square root then satisfies $0 \leq \phi_{\sqrt{\epsilon^0}} \leq \pi/2$, and hence

$$0 \leq \phi_k \leq \pi/2. \tag{13}$$

Equation (13) is clearly in contradiction with condition (11), and we are left with the conclusion that spatial nonlocality and dissipation are necessary conditions for the existence of negative refraction. This is the first important result in this Letter. Although a related discussion can be found in Ref. [16], these necessary conditions have not been stated previously to our knowledge. Finally, let us note that the necessity of spatial nonlocality also holds for longitudinal waves. Indeed, in the absence of spatial nonlocality, the longitudinal component of the electric field cannot propagate and, hence, cannot exhibit negative refraction.

We now investigate the existence of a sufficient condition for negative refraction. For nongyrotropic media, due to invariance under point reflection, $\epsilon_{T,L}(\omega,k)$ can be expanded in powers of k^2 in the form (contributions with odd powers of k are forbidden by symmetry)

$$\epsilon_{T,L}(\omega,k) = \epsilon^0(\omega) + \sum_{l=1}^{+\infty} \epsilon_{T,L}^l(\omega)(k\mathcal{L})^{2l}, \tag{14}$$

where $\mathcal{L}$ denotes the largest intrinsic characteristic length of the material (e.g., the lattice constant for crystals, the mean free path of conduction electrons for metals, the Debye screening length for plasmas, etc.) and vanishes in local media. Inserting Eq. (14) into the sum rule (6) and imposing the validity of the resulting equation for any value of k, we obtain

$$\int_0^{+\infty} \mathrm{Im}\epsilon^0(\omega')\omega' d\omega' = \frac{\pi\omega_p^2}{2},$$
$$\int_0^{+\infty} \mathrm{Im}\epsilon_{T,L}^l(\omega')\omega' d\omega' = 0, \quad l > 0. \tag{15}$$

Equations (15) imply that, for $l > 0$, the functions $\mathrm{Im}\epsilon_{T,L}^l(\omega)$ have to change sign in the interval $\omega \in [0,+\infty]$. This apparently surprising result is a direct consequence of causality [20]. This is not in contradiction with Eq. (7), since it only applies separately for each $\epsilon_{T,L}^l(\omega)$ with $l > 0$, and $\mathrm{Im}\epsilon^0(\omega) > 0$, while Eq. (7) implies that the imaginary part of the total sum (14) (for real k) is positive.

For transverse waves, and for isotropic and nongyrotropic media, we shall now prove that the first nonlocal correction to the dielectric function gives a sufficient condition for the existence of negative refraction. Let us consider the dielectric function [31]

$$\epsilon_T(\omega,k) = \epsilon^0(\omega) + \epsilon_T^1(\omega)(k\mathcal{L})^2. \tag{16}$$

Inserting Eq. (16) into (8), we obtain

$$k(\omega) = \frac{\omega}{c}\sqrt{\epsilon^0(\omega)F(\omega)}, \tag{17}$$

where $F(\omega) = [1 - \omega^2\mathcal{L}^2\epsilon_T^1(\omega)/c^2]^{-1}$. From Eq. (15), we have deduced that there exist frequencies ω such that $\mathrm{Im}\epsilon_T^1(\omega) < 0$, leading to $\mathrm{Im}F(\omega) < 0$. Since $\phi_k = \phi_{\sqrt{\epsilon 0}} + \phi_{\sqrt{F}}$, we can state that

$$\exists\,\omega \text{ such that } \pi \geq \phi_k > \phi_{\sqrt{\epsilon^0}} + \pi/2. \tag{18}$$

This condition on the phase of the wavevector amounts to showing that frequencies for which Eq. (11) (defining negative refraction) is satisfied necessarily exist. We conclude that a nonlocal medium, that can be represented as the most general first-order correction (allowed by symmetry) to a local response Eq. (16), necessarily has a range of frequencies in which negative refraction is observed. This is the second important result in this Letter.

Two comments are worth noting. First, applying the same analysis to longitudinal waves does not allow us to conclude on the existence of a sufficient condition for negative refraction in this case. Second, we stress that, while dissipation and spatial nonlocality are necessary conditions, they are not, in general, sufficient conditions for negative refraction. For example, consider the nonlocal transverse dielectric function

$$\epsilon_T(\omega, k) = 1 - \frac{N_0^2}{\omega^2 - D_0^2 + iD_1\omega - D_2k^2 + iD_3\omega k^2}, \quad (19)$$

representing the generic response of a spatially nonlocal (isotropic, homogeneous, and passive) medium near a resonance. N_0 and D_j are the first real positive constant coefficients of the most general expansion in powers of ω and k^2. For particular choices of the system, such coefficients reduce to well-known physical quantities—e.g., the average collision time, viscosity, decay width, plasma frequency squared, square of the frequency gap, etc.—and (19) can describe the response of charged viscous fluids (e.g., electrons in certain metals [13]) or the spatially nonlocal Lorentz model (e.g., exciton-polariton resonance for certain semiconductors [32]). See [20]. Changing the values of the coefficients of the k^2 terms, one can switch from a negative refraction regime to a positive refraction regime at low frequencies, as previously shown for the electron hydrodynamic model [13] (see [20]). The analysis therefore leads to the conclusion that spatial nonlocality supports, but does not necessarily imply, negative refraction [33].

Before concluding, it is worth commenting on the relation between the formalism used in this Letter and the more familiar local formalism. The usual local analysis, in terms of the dielectric function $\epsilon(\omega)$ and a magnetic permeability $\mu(\omega)$, is clearly included in the nonlocal formalism described in this Letter in term of a single spatially nonlocal response function $\epsilon_{ij}(\omega, \boldsymbol{k})$. It indeed emerges as a particular case in the limit $\omega/k \to 0$ and $k \to 0$ (see Refs. [18–20]). In this limit, our result that spatial nonlocality is a necessary condition for negative refraction translates into the well-known fact that, in the local limit, to observe negative refraction it is necessary to have both $\epsilon(\omega) \neq 1$ and $\mu(\omega) \neq 1$. The sufficient condition for negative refraction, usually stated in the local formalism in the form $\mathrm{Re}[\epsilon(\omega)]|\mu(\omega)| + \mathrm{Re}[\mu(\omega)]|\epsilon(\omega)| < 0$ (see, e.g., Refs. [1,5,30,34]), is consistent with (although not implied by) our analysis. Indeed, the sufficient condition of a k^2 nonlocal correction does not require any constraints on the general form of the nonlocal permittivity Eq. (16). However, one should not conclude that the theory described in this Letter is a mere generalization of a well-established approach. The use, in the first place, of a nonlocal description of the electrodynamic response is fundamental at least for three reasons. First, the meaning of $\mu(\omega)$ as a true response function [with $\mathrm{Im}\mu(\omega) \geq 0$ for passive media] is valid only for very small k *and* in a limited range of frequencies in the neighborhood $\omega/k = 0$ [18–20]. Outside this range, $\mu(\omega)$ loses its meaning of a magnetic permeability, and its imaginary part has, in general, an indefinite sign [20]. This is actually the reason behind the sufficient condition for negative refraction proven in this Letter [the function $F(\omega)$ in Eq. (17) can be understood as an effective permeability]. Second, the analysis based on a nonlocal dielectric function reveals that $\mu(\omega)$, even if apparently a local response, encodes (some of) the nonlocal effects to the order of k^2 initially included in the nonlocal dielectric function $\epsilon_{T,L}(\omega, k)$ [20]. Therefore, the existence of a permeability $\mu(\omega) \neq 1$, as a condition for negative refraction in the local approach, can be seen as a disguised residual of spatial nonlocality. Third, the local formalism, although appropriate for a phenomenological analysis of certain materials, cannot be used for a first-principles analysis of negative refraction on the full spectrum of frequencies because of its limited range of validity around $\omega/k = 0$ and $k \to 0$ [17–19]. In the present Letter, we applied constraints coming essentially from causality [except for Eq. (8)]. In the local limit, it is possible to include thermodynamical constraints as well, that are instead, in general, not valid in the nonlocal theory. In Ref. [35], it has been indeed argued that thermodynamics forbids negative refraction in the local limit. Indeed, thermodynamical considerations allow us to conclude that negative refraction is realized exactly outside the regime of applicability of the $\epsilon(\omega)$ and $\mu(\omega)$, and the nonlocality needed in the $\epsilon_{T,L}(\omega, k)$ formalism to include magnetic effects is not enough to support negative refraction, while dependency of the response on the spatial derivatives of the electric field is needed as well. This is true also in the "weak" spatial nonlocal case; see (16).

In conclusion, we have demonstrated that negative refraction of electromagnetic waves is intrinsically related to both dissipation and spatial nonlocality. Their simultaneous presence is a necessary condition for negative refraction. The implications of the theory developed in this Letter are manyfold. At the fundamental level, the analysis can be extended nonisotropic, nonhomogeneous, or non-point-reflection-invariant media, to assess if, and under which conditions, the main results of this Letter remain valid. Very interesting is also to extend the analysis of this Letter to other kinds of waves such as acoustic waves, for which the design of negative refraction materials is an active field as well. At a practical level, the results in this Letter show that nonlocality should not be considered as a refinement of the existing theories, allowing one to improve in an incremental way the precision in the determination of the effective response functions of real materials. Instead, any inverse procedure should include in the first place nonlocality, since its exclusion is fundamentally inconsistent and can lead to uncontrollable systematic errors. Finally, our results open the way to a systematic classification of media with or without negative refraction, providing a clear connection between this phenomenon and the dynamical properties of materials, as already shown in the case of metals [13].

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