Criterion for Negative Refraction with Low Optical Losses from a Fundamental Principle of Causality

Mark I. Stockman*

Department of Physics and Astronomy, Georgia State University, Atlanta, Georgia 30303, USA

(Received 4 November 2006; published 27 April 2007)

From the fundamental requirement of causality, we derive a rigorous criterion of negative refraction (left handedness). This criterion imposes the lower limits on the electric and magnetic losses in the region of the negative refraction. If these losses are eliminated or significantly reduced by any means, including the compensation by active (gain) media, then the negative refraction will disappear. This theory can be particularly useful in designing new left-handed materials: testing the expected polarizabilities of a medium against this criterion would check the compliance with the causality and verify the design feasibility.

DOI: 10.1103/PhysRevLett.98.177404 PACS numbers: 78.20.Ci, 71.45.Gm, 73.20.Mf, 78.67.−n

Recently there has been significant attention devoted to the so-called left-handed materials (LHM), which are also called negative-refraction media [1–13]. In such materials, the directions of energy transfer and wavefront propagation are opposite. This leads to remarkable electromagnetic properties such as refraction at surfaces that is described by a negative-refraction index n. This, in turn, causes a flat slab of a left-handed material with $n = -1$ to act as a ''perfect lens'' creating, without reflections at the surfaces, a nondistorted image. This is a so-called Veselago lens [14]. Moreover, such a lens can also build an image in the near field [15]. Optical losses in LHMs are detrimental to their performance. These losses for LHMs in the near-infrared and visible region are significant [8–11], which drastically limits their usefulness. There have been proposals [16–18] to compensate these losses and also absorption in the plasmonic perfect lens [19] with optical gain. This compensation is similar to the idea of spaser or nanoplasmonic laser [20–22]. This idea appears to be a way to resolve this loss problem.

In this Letter, we show that compensating the optical losses, or by any means (material or structural) significantly reducing the imaginary part of the dielectric permittivity ε and magnetic permeability μ, will necessarily change also the real parts of these quantities in such a way that the negative refraction disappears. This follows from the dispersion relations, i.e., ultimately, from the fundamental principle of causality. This principle is conventionally expressed by the familiar Kramers-Kronig dispersion relations (see, e.g., Ref. [23]). Here, we derive similar dispersion relations for the *squared* refractive index. Using them we show that a significant reduction in the optical losses at and near the observation frequency will necessarily eliminate the negative refraction.

The Kramers-Kronig relations follow from the causality of the dielectric response function in the temporal domain. Then one can prove that in the frequency domain permittivity $\varepsilon(\omega)$ does not have singularities in the upper half-plane of the complex variable ω. From this and the limit $\varepsilon(\omega) \to 1$ for $\omega \to \infty$, one derives the conventional Kramers-Kronig dispersion relation for the dielectric function. For the same causality reason, magnetic permeability $\mu(\omega)$ does not have singularities in the upper half-plane of complex ω. Since also $\mu(\omega) \to 1$ for $\omega \to \infty$, permeability $\mu(\omega)$ satisfies a similar dispersion relation. Note the requirement of the response linearity is essential: nonlinear and saturated polarizabilities generally do not satisfy the Kramers-Kronig relations [24]. We will below consider systems including gain media; in those cases we assume that the optical responses to the *signal* (observed) radiation are linear. This, of course, requires the signal to be weak enough to ensure the linearity of the responses to it and the applicability of the Kramers-Kronig relations.

We consider a material to be an effective medium characterized by macroscopic permittivity $\varepsilon(\omega)$ and permeability $\mu(\omega)$. The squared complex refraction index $n^2(\omega) = \varepsilon(\omega)\mu(\omega)$ has exactly the same analytical properties as $\varepsilon(\omega)$ and $\mu(\omega)$ separately: $n^2(\omega)$ does not have singularities in the upper half-plane of complex ω and $n^2(\omega) \to 1$ for $\omega \to \infty$. Therefore, absolutely similar to the derivation of the Kramers-Kronig relations for the permittivity or permeability (see, e.g., Ref. [23]), we obtain a dispersion relation for $n^2(\omega)$,

$$\mathrm{Re}\, n^2(\omega) = 1 + \frac{2}{\pi}\mathcal{P}\int_0^\infty \frac{\mathrm{Im} n^2(\omega_1)}{\omega_1^2 - \omega^2}\omega_1 d\omega_1, \qquad (1)$$

where $\mathcal{P}$ denotes the principal value of an integral.

Note that in contrast to $n^2(\omega)$, refractive index $n(\omega) = \sqrt{n^2}$ may possess singularities in the upper half-plane and thus is generally not causal; this is true, in particular, when optical gain is present [25]. The refractive index n *per se* does not enter the Maxwell equations; it is not a susceptibility, and it does not have to obey the causality, while n^2 does. This theory is based on n^2, not n; the noncausality of n is irrelevant for its purposes.

Now we assume that at and near the observation frequency ω the material is transparent (e.g., the losses are compensated by gain), which mathematically implies that

 © 2007 The American Physical Society

$\mathrm{Im}n^2(\omega) = 0$ and $\partial[\mathrm{Im}n^2(\omega)]/\partial\omega = 0$ (this vanishing is required only at the observation frequency). Then the principal value in the right-hand side of Eq. (1) can be omitted. Multiplying both sides of this equation by ω^2 and differentiating over ω (one can differentiate under the integral over ω as a parameter, because the point $\omega_1 = \omega$ is not singular anymore), we obtain

$$\frac{\partial\omega^2[\mathrm{Re}n^2(\omega) - 1]}{\partial\omega} = \frac{4\omega}{\pi}\int_0^\infty \frac{\mathrm{Im}n^2(\omega_1)}{(\omega_1^2 - \omega^2)^2}\omega_1^3 d\omega_1. \quad (2)$$

The left-hand side of this equation can be expressed in terms of the phase velocity $\mathbf{v}_p = (\mathbf{k}/k)\omega/k$, where real wave vector is $k = \sqrt{\mathrm{Re}n(\omega)^2}\omega/c$, and c is speed of light, and group velocity $\mathbf{v}_g = (\mathbf{k}/k)\partial\omega/\partial k$. In this way, we obtain

$$\frac{1}{\mathbf{v}_p\mathbf{v}_g} - \frac{1}{c^2} = \frac{2}{\pi c^2} \times \int_0^\infty \frac{\varepsilon''(\omega_1)\mu'(\omega_1) + \mu''(\omega_1)\varepsilon'(\omega_1)}{(\omega_1^2 - \omega^2)^2}\omega_1^3 d\omega_1, \quad (3)$$

where $\varepsilon' = \mathrm{Re}\varepsilon$, $\varepsilon'' = \mathrm{Im}\varepsilon$ and, similarly, $\mu' = \mathrm{Re}\mu$, $\mu'' = \mathrm{Im}\mu$; $\mathrm{Im}n^2(\omega) = \varepsilon''(\omega)\mu'(\omega) + \mu''(\omega)\varepsilon'(\omega)$.

In the case of the negative refraction, the directions of the phase and energy propagation are opposite, therefore $\mathbf{v}_p\mathbf{v}_g < 0$. Consequently, we obtain from Eq. (3) a rigorous criterion of the negative refraction with no (or low) loss at the observation frequency ω as

$$\frac{2}{\pi}\int_0^\infty \frac{\varepsilon''(\omega_1)\mu'(\omega_1) + \mu''(\omega_1)\varepsilon'(\omega_1)}{(\omega_1^2 - \omega^2)^2}\omega_1^3 d\omega_1 \leq -1. \quad (4)$$

This criterion directly imposes the lower bounds on the dielectric losses $[\varepsilon''(\omega_1) > 0]$, overlapping with the magnetic plasmonic behavior $[\mu'(\omega_1) < 0]$ and the magnetic losses $[\mu''(\omega_1) > 0]$ overlapping with the electric plasmonic behavior $[\varepsilon'(\omega_1) < 0]$. The denominator $(\omega_1^2 - \omega^2)^2$ makes the integral to converge for $|\omega_1 - \omega|$ large; it would have diverged at $|\omega_1 - \omega| \to 0$ if the integrand did not vanish at that point. Thus, the major contribution to Eq. (4) comes from the lossy, overlapping electric and magnetic resonances close to observation frequency ω.

The stability of the system requires that no net gains are present at any frequency, i.e., $\varepsilon''(\omega) \geq 0$ and $\mu''(\omega) \geq 0$ everywhere [23]. There is a known condition of negative refraction [26] $\mathrm{Im}n^2(\omega) < 0$, which for exactly compensated losses should be extended as $\mathrm{Im}n^2(\omega) \leq 0$. This condition is always satisfied in the region of left handedness where $\varepsilon'(\omega) < 0$ and $\mu'(\omega) < 0$. Thus, this condition is trivial: in contrast to Eq. (4), it does not impose a lower limit on the losses.

In the absence of magnetic resonances, in the optical region $\mu' = 1$ and $\mu'' = 0$. Then it is obvious than the integral in the left-hand side of Eq. (4) is strictly positive and this criterion is not satisfied; i.e., the negative refraction is absent. In the presence of a magnetic resonance, in a part of its region $\mu' < 0$ and $\mu'' > 0$; thus the criterion (4) can, in principle, be satisfied. However, this requires nonzero losses: $\mu'' > 0$ and/or $\varepsilon'' > 0$.

As an alternative way to satisfy the transparency requirement at the observation frequency, one may attempt to add a gain to exactly cancel out the losses at this frequency [17,18], keeping them elsewhere to satisfy criterion (4) from dispersion relation (3). Is it possible from the positions of causality? Consider a particular example when the left-handed behavior is due to a resonance at some frequency ω_r, and that this resonance dominates the behavior of permittivity $\varepsilon(\omega_1)$. Such a resonant behavior is described by a simple pole of the permittivity [27], $\varepsilon_r(\omega_1) \propto [\omega_1 - \omega_r + i\gamma(\omega_1)]^{-1}$, where $\gamma(\omega_1)$ is a frequency dependent relaxation rate. Because losses are compensated at the observation frequency, $\gamma(\omega) = 0$. Since the losses should nowhere be negative, it is obvious that $\gamma(\omega)$ must have a minimum at frequency ω, which implies that $\partial\gamma(\omega)/\partial\omega = 0$, and $\partial^2\gamma(\omega)/\partial\omega^2 > 0$. Assuming that the resonance and observation frequencies are close enough, one can expand $\gamma(\omega_1)$ about frequency ω and obtain

$$\varepsilon_r(\omega_1) \propto \left[\omega_1 - \omega_r + i\frac{1}{2}(\omega_1 - \omega)^2\frac{\partial^2\gamma(\omega)}{\partial\omega^2}\right]^{-1}. \quad (5)$$

However, this $\varepsilon_r(\omega_1)$ has an extra pole at a complex frequency

$$\omega_1 \approx \omega + 2i(\partial^2\gamma(\omega)/\partial\omega^2)^{-1}. \quad (6)$$

This pole is situated in the *upper* half-plane, while any causal quantity as a function of frequency must be analytical in the upper half-plane. Hence, this behavior *violates causality*. Thus, we conclude that in this manner it is impossible to compensate the losses at a single (observation) frequency.

It is still possible that both the magnetic resonance and electric plasmonic behavior are present, but their losses are compensated by an active-medium gain. However, such compensation must take place not only at the observation frequency ω, but for the *entire* region of such resonances assuming their homogeneous nature. This means that in Eq. (4) whenever $\mu'(\omega_1) < 0$, we have $\mu''(\omega_1) = 0$ and $\varepsilon''(\omega_1) = 0$. However, in this case the contribution of this region to the integral in Eq. (4) vanishes, and the contribution of the region of normal optical magnetic behavior ($\mu = 1$) is always positive. Consequently, the negative-refraction criterion is violated, which implies the absence of the negative refraction.

To obtain the negative refraction, the losses in the magnetic resonance region not only should be present, but they should be significant not only to overcome the positive contribution of the nonresonant region to the integral in Eq. (4), but actually to make it less than -1. Thus, significantly reducing by any means, passive or active (by gain), the losses of the negative-refraction resonances will necessarily eliminate this negative refraction itself. Fundamentally, this stems from the fact that the imaginary part and real part of the squared index of refraction are not independent but must satisfy the requirements imposed by the principle of causality.

One has to explore also a possibility to satisfy the criterion (4) with low losses at the working frequency ω by having a left-handed resonance somewhere else at some resonance frequency ω_r remote from ω to satisfy Eq. (4). The contribution of such a remote resonance to the integral in Eq. (4) can be approximated as

$$\frac{2}{\pi}\frac{\omega_r^3}{(\omega_r^2-\omega^2)^2}\mathrm{Im}\int_{-\infty}^{\infty} n_r^2(\omega_1)d\omega_1. \tag{7}$$

Here $n_r^2(\omega_1)$ is the resonant contribution to the squared index. It is assumed that it decreases rapidly enough when $|\omega_1-\omega_r|\rightarrow\infty$, which is the expression of its resonant behavior. In this case, it is possible to extend the integral in this equation over the entire region, as indicated. As required by the causality, $n_r^2(\omega_1)$ does not have any singularities in the upper half-plane of ω_1. This integral can be closed by an infinite arc in the upper half-plane, which gives the zero result due to this absence of the singularities there. Hence, the distant resonances do not contribute to the negative-refraction criterion (4). This completes the proof that zero (or, very low) losses at and near the observation frequency are incompatible with the negative refraction.

We point out that in reality these losses do not have to be zero to eliminate the negative refraction. If they are merely much smaller than the losses in the adjacent regions that result in the positive contribution to the integral in criterion (4), then the negative refraction will be absent.

Simple, exactly solvable (in the sense that they can be reduced to an explicit transcendental equation), and convincing illustrations of the above theory are provided by the negative refraction of surface plasmon polaritons (SPPs) in films with nanoscale thickness. Note that it is a two-dimensional refraction but our consideration is based on the principle of causality and is general, applicable to refraction in spaces of arbitrary dimensions. We emphasize the examples to follow do not provide a proof but serve merely as illustrations of the above-given proof.

Consider a flat layer with nanoscale thickness d made of a material with dielectric permittivity ε_2 embedded between two half-spaces of materials with permittivities ε_1 and ε_3. The dispersion relation, i.e., wave vector k as a function of ω or vice versa, of the waves (SPPs) bound to the nanolayer can be found from an exact, analytical transcendental equation

$$\tanh(\omega d\varepsilon_2 u_2/c) = -u_2(u_1+u_2)/(u_1u_3+u_2^2), \tag{8}$$

where $u_i=\varepsilon_i^{-1}\sqrt{(kc/\omega)^2-\varepsilon_i}$. In this equation and below, we treat k as a complex wave vector whose imaginary part describes losses.

As the first example, we mention a semi-infinite metal (silver) covered with a nanolayer of dielectric with a half-space of another dielectric covering it [28,29]. This system possesses an extended spectral region of negative refraction [29]; however, in this region the SPP losses are so high that the propagation is actually absent, in accord with the above-presented theory.

Another exactly solvable example of negative refraction also described by Eq. (8) is given by SPPs in a metal film of a nanoscopic thickness embedded in a dielectric [30]. There are two metal-dielectric interfaces and, correspondingly, two modes of SPPs in this system. Because there is symmetry with respect to the reflection in the middle plain, these SPP modes are classified according to their magnetic-field parity: symmetric and antisymmetric. As an example, we consider a silver film with thickness $d=30$ nm in vacuum. The corresponding dispersion relations are shown in Fig. 1. As we see from panel (a), the symmetric SPPs have regions of both the positive refraction ($\mathrm{Re}k<2\times10^5$ cm^{-1}) and negative refraction ($\mathrm{Re}k>2\times10^5$ cm^{-1}), while the antisymmetric SPPs possess only the positive refraction. The optical losses are shown in Fig. 1(b). For most of the positive-refraction region of the symmetric SPPs and in the entire spectral range of the antisymmetric SPPs, these losses are relatively very small: $\mathrm{Im}k\ll\mathrm{Re}k$. However, for the symmetric SPPs (the solid line) close to the negative-refraction region, the losses dramatically increase by orders of magnitude. Inside the negative-refraction region, they are extremely high, $|\mathrm{Im}k|\gtrsim|\mathrm{Re}k|$, so the propagation is overdamped and actually absent, in the full agreement with the conclusions of the present theory [31].

Yet another system that supports the negative-refraction SPPs is a dielectric nanolayer embedded in a metal [30,32]. This system is also symmetric and possesses two metal-dielectric interfaces. Therefore it supports two branches of SPPs that are characterized by parity. The corresponding dispersion relations are displayed in Fig. 2. The real part of the dispersion relation for these two types of modes is displayed in panel (a). From it we see that in the entire spectral region the symmetric SPPs (dashed line) have normal, positive refraction ($v_g>0$), while the antisymmetric SPPs (solid line) are negative refracting ($v_g<0$).

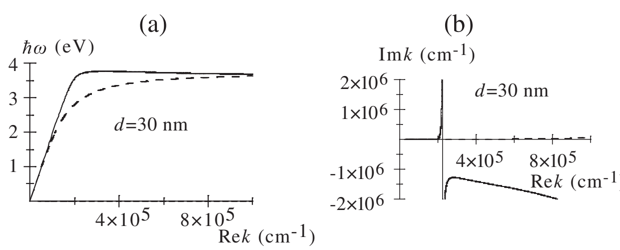

FIG. 1. Dispersion relations for thin silver film in vacuum. The symmetric and antisymmetric modes are displayed with solid and dashed lines, respectively. (a) Real part of dispersion relation: frequency ω as a function of $\mathrm{Re}k$. (b) Imaginary part of the dispersion relation: dependence of $\mathrm{Im}k$ on $\mathrm{Re}k$. Thickness of the silver film is $d=30$ nm.

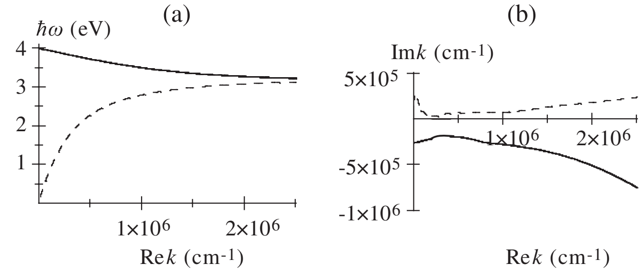

FIG. 2. (a) For a thin ($d = 10$ nm) dielectric layer with $\varepsilon_d = 3$ embedded in silver, dispersion relation of SPPs is displayed of as dependence of frequency $\hbar\omega$ on the real part of wave vector. (b) For the same system, dependence of Imk on Rek. For both panels, the solid lines pertain to the antisymmetric SPP mode, and the dashed lines denote the symmetric SPP mode.

The corresponding losses are displayed in Fig. 2(b). We note that the losses of the positive-refraction, symmetric mode (dashed line) are relatively small in the entire region, Im$k \ll$ Rek. In a sharp contrast, for the antisymmetric, negative-refraction mode (solid line), the losses for small wave vectors are very high, $|\mathrm{Im}k| \gtrsim \mathrm{Re}k$, so the wave propagates through only a few periods before it dissipates.

To conclude, from the fundamental principle of causality, we have derived a dispersion relation (1) for the squared refraction index. From it, assuming a low loss at the observation frequency, we have derived a criterion (4) of the negative refraction. We have shown that the low loss at and near the observation frequency is incompatible with the existence of the negative refraction [33]. While at the THz region the losses may not be significant, they are very large in the optical region. The loss compensation or significant reduction will necessarily lead to the disappearance of the negative refraction itself due to the dispersion relation dictated by the causality.

This work was supported by grants from the Chemical Sciences, Biosciences and Geosciences Division of the Office of Basic Energy Sciences, Office of Science, U.S. Department of Energy, a Grant No. CHE-0507147 from NSF, and a grant from the US-Israel BSF.

Note added in proof.—After this Letter was submitted, a recent experiment [34] found negative two-dimensional refraction for a nanoscopic dielectric layer in metal with significant optical losses, Imk/Re$k \sim 0.25$, in qualitative agreement with this theory.

*Email address: mstockman@gsu.edu
Electronic address: http://www.phy-astr.gsu.edu/stockman

[1] J. B. Pendry, Opt. Express **11**, 639 (2003).
[2] D. R. Smith and N. Kroll, Phys. Rev. Lett. **85**, 2933 (2000).
[3] D. R. Smith, W. J. Padilla, D. C. Vier, S. C. Nemat-Nasser, and S. Schultz, Phys. Rev. Lett. **84**, 4184 (2000).
[4] R. A. Shelby, D. R. Smith, S. C. Nemat-Nasser, and S. Schultz, Appl. Phys. Lett. **78**, 489 (2001).
[5] A. A. Houck, J. B. Brock, and I. L. Chuang, Phys. Rev. Lett. **90**, 137401 (2003).
[6] C. G. Parazzoli, R. B. Greegor, K. Li, B. E. C. Koltenbah, and M. Tanielian, Phys. Rev. Lett. **90**, 107401 (2003).
[7] A. N. Lagarkov and V. N. Kissel, Phys. Rev. Lett. **92**, 077401 (2004).
[8] S. Zhang, W. Fan, N. C. Panoiu, K. J. Malloy, R. M. Osgood, and S. R. J. Brueck, Phys. Rev. Lett. **95**, 137404 (2005).
[9] A. N. Grigorenko, A. K. Geim, H. F. Gleeson, Y. Zhang, A. A. Firsov, I. Y. Khrushchev, and J. Petrovic, Nature (London) **438**, 335 (2005).
[10] V. M. Shalaev, W. S. Cai, U. K. Chettiar, H. K. Yuan, A. K. Sarychev, V. P. Drachev, and A. V. Kildishev, Opt. Lett. **30**, 3356 (2005).
[11] G. Dolling, C. Enkrich, M. Wegener, C. M. Soukoulis, and S. Linden, Science **312**, 892 (2006).
[12] U. Leonhardt, Science **312**, 1777 (2006).
[13] J. B. Pendry, D. Schurig, and D. R. Smith, Science **312**, 1780 (2006).
[14] V. G. Veselago, Sov. Phys. Usp. **10**, 509 (1968).
[15] J. B. Pendry, Phys. Rev. Lett. **85**, 3966 (2000).
[16] M. P. Nezhad, K. Tetz, and Y. Fainman, Opt. Express **12**, 4072 (2004).
[17] A. K. Popov and V. M. Shalaev, Opt. Lett. **31**, 2169 (2006).
[18] A. K. Popov and V. M. Shalaev, Appl. Phys. B **84**, 131 (2006).
[19] S. A. Ramakrishna and J. B. Pendry, Phys. Rev. B **67**, 201101(R) (2003).
[20] D. J. Bergman and M. I. Stockman, Phys. Rev. Lett. **90**, 027402 (2003).
[21] M. I. Stockman and D. J. Bergman, in *Proceedings of SPIE: Complex Mediums IV: Beyond Linear Isotropic Dielectrics*, edited by M. W. McCall and G. Dewar (SPIE, San Diego, California, 2003), Vol. 5221, p. 93.
[22] R. Colombelli, F. Capasso, C. Gmachl, A. L. Hutchinson, D. L. Sivco, A. Tredicucci, M. C. Wanke, A. M. Sergent, and A. Y. Cho, Appl. Phys. Lett. **78**, 2620 (2001).
[23] L. D. Landau and E. M. Lifshitz, *Electrodynamics of Continuous Media* (Pergamon, Oxford and New York, 1984).
[24] R. W. Boyd, *Nonlinear Optics* (Academic, San Diego, London, 2003).
[25] J. Skaar, Phys. Rev. E **73**, 026605 (2006).
[26] R. A. Depine and A. Lakhtakia, Microwave Opt. Tech. Lett. **41**, 315 (2004).
[27] D. J. Bergman and D. Stroud, in *Solid State Physics*, edited by H. Ehrenreich and D. Turnbull (Academic, Boston, 1992), Vol. 46, p. 148.
[28] A. Karalis, E. Lidorikis, M. Ibanescu, J. D. Joannopoulos, and M. Soljacic, Phys. Rev. Lett. **95**, 063901 (2005).
[29] M. I. Stockman, Nano Lett. **6**, 2604 (2006).
[30] A. Alu and N. Engheta, J. Opt. Soc. Am. B **23**, 571 (2006).
[31] In this region Im$k < 0$, describing the propagation of energy opposite to the wave vector.
[32] H. Shin and S. Fan, Phys. Rev. Lett. **96**, 073907 (2006).
[33] The low loss is understood as significantly lower than in the spectral regions adjacent to the magnetic resonance, or the loss that would have be present if the gain medium compensation were not introduced.
[34] H. Lezec, J. A. Dionne, and H. Atwater, Science, **316**, 430 (2007).