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Phys. Rev. Lett. **117**, 107404 — Published 2 September 2016

DOI: [10.1103/PhysRevLett.117.107404](https://doi.org/10.1103/PhysRevLett.117.107404)

Real and Imaginary Properties of Epsilon-near-Zero Materials

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From the fundamental principle of causality we show that epsilon-near-zero (ENZ) materials with very low (asymptotically zero) intrinsic dielectric loss do necessarily possess a very low (asymptotically zero) group velocity of electromagnetic wave propagation. This leads to the loss function being singular and causes high non-radiative damping of optical resonators and emitters (plasmonic nanoparticles, quantum dots, chromophore molecules) embedded into them or placed at their surfaces. Rough ENZ surfaces do not exhibit hot spots of local fields suggesting that surface modes are overdamped. Reflectors and waveguides also show very large losses both for realistic and idealized ENZ.

PACS numbers: 78.67.Pt, 71.45.Gm, 81.05.Zx, 42.70.-a

Recently, materials at frequencies ω close to the bulk plasmon frequency, ω_P , which are characterized by dielectric permittivity ε being small enough, $|\varepsilon| \ll 1$, and are usually referred to as Epsilon Near Zero (ENZ) materials, have attracted a great deal of attention [1–12]. Their optical properties are expected to be quite remarkable: ENZ should totally reflect light at all angles, the phase velocity of light tends to infinity and, correspondingly, the light wave carries almost constant phase, the density of photonic states diverges at $\omega \rightarrow \omega_P$, a waveguide formed inside an ENZ material can confine light at deep sub-wavelength dimensions, there is no reflections even at sharp bands, and the unavoidable roughness of the waveguide walls does not significantly spoil the wave-guiding. As in many other cases in nanooptics [13], dielectric losses present a significant problem deteriorating these unique properties and limiting useful applications of ENZ materials. Several approaches have been proposed to mitigate these unwanted effects of the optical loss: new conducting oxide and nitride materials, in particular, indium tin oxide (ITO), bear promise of significant reduction of the optical losses [14, 15]. Another proposed approach is based on loss compensation by gain [10]. The most radical way to eliminate the loss is using of an all-dielectric metamaterial at optical frequencies whose effective permittivity is near zero and losses are extremely low [16].

The goal of this Letter is to establish fundamental limitations that causality and dielectric losses impose on ENZ materials. We show that a fundamental causality requirement for a perfectly transparent ENZ material (where $\varepsilon = \varepsilon' + i\varepsilon''$ possesses a small real part, $1 \gg |\text{Re}\varepsilon| \rightarrow 0$ and a negligible imaginary part, $\varepsilon'' = \text{Im}\varepsilon = 0$) leads to a vanishing group velocity, $v_g \rightarrow 0$. Thus, such a “perfect” ENZ material cannot transmit energy or information. Also, the establishment time of stationary optical regime diverges $\propto v_g^{-1} \rightarrow \infty$. (Note that it has been understood that in “perfect lens” systems, where $\varepsilon'' \rightarrow 0$ and $\varepsilon' \rightarrow -1$, the asymptotically infinite establishment time has already been found earlier [42, 43].) Therefore

any dielectric metamaterials emulating $\varepsilon' \rightarrow 0$ must be diffractive, i.e., not true metamaterials that transmit and absorb light but not scatter or diffract it. Moreover, we show that the introduction of even very small losses drastically degrades the expected remarkable properties of idealized ENZ materials. Adding gain to reduce ε'' would be no radical solution either because gain affects also ε' leaving the causality limitations in place [18]. Also, the energy loss function, $L = -\text{Im}(\varepsilon^{-1})$, is singular for a low-loss ENZ material, which causes strong damping of embedded nanosystems. In the optical (near-infrared to visible) range, real ENZ materials such as ITO do not actually show remarkable manifestations of the ENZ behavior due to the losses.

Because we are interested in the most fundamental properties of the ENZ materials, we consider such a material as a uniform and isotropic infinite medium (natural or metamaterial [19]). We also assume that the dielectric response of the ENZ material is local (i.e., there is no spatial dispersion), which assures that our results are also applicable to micro- and nano-structures made of it through the use of the Maxwell boundary conditions.

We start first with an idealized case of a material that is lossless at observation frequency ω , i.e., $\varepsilon''(\omega) = 0$. We assume that it is also lossless in the infinitesimal vicinity of ω , which we re-formulate as a condition $d\varepsilon''(\omega)/d\omega = 0$. We will also assume that this material is not magnetic (which is usual at the optical frequencies [20]). Under these conditions, the fundamental causality principle leads to an exact dispersion relation [18],

$$\frac{c^2}{v_g v_p} = 1 + \frac{2}{\pi} \int_0^\infty \frac{\varepsilon''(\omega_1)}{(\omega_1^2 - \omega^2)^2} \omega_1^3 d\omega_1, \quad (1)$$

where c is speed of light, $v_p = c / \sqrt{\varepsilon'(\omega)}$ is phase velocity, and $v_g = c / \frac{d}{d\omega} \omega \sqrt{\varepsilon'(\omega)}$ is group velocity. From this, we can immediately find an exact dispersion relation

for group velocity as

$$v_g = c\sqrt{\varepsilon'(\omega)} \left[1 + \frac{2}{\pi} \int_0^\infty \frac{\varepsilon''(\omega_1)}{(\omega_1^2 - \omega^2)^2} \omega_1^3 d\omega_1 \right]^{-1}. \quad (2)$$

Due to the requirement of stability, $\varepsilon'' \geq 0$. Consequently, $v_g \leq c\sqrt{\varepsilon'(\omega)}$, and $v_g \rightarrow 0$ for $\varepsilon'(\omega) \rightarrow 0$. Thus, a lossless ENZ material in the limit $\varepsilon'(\omega) = 0$ does not transport electromagnetic energy. As a corollary, establishment time τ_s of the stationary regime diverges, $\tau_s \sim a/v_g \rightarrow \infty$, where a is the characteristic size, and the ENZ media may have high (albeit slow) optical nonlinearities (cf. Ref. 21). Note that Eq. (2) is an *exact local* property of the lossless ENZ materials, and it is not affected by their micro- or nano-structuring.

In the experiment of Ref. 16, the medium designed to be a lossless ENZ metamaterial is, in reality, a diffractive medium – a photonic crystal [22, 23] where the linear photon momentum is within the first Brillouin zone, where the diffraction does not show itself. Another class of pertinent systems are parallel-plate waveguides. These are not true continuous media but emulate properties of two-dimensional ENZ media [2–4], which are isotropic in the plane of the waveguide. They may be designed for microwave frequencies where the propagation losses are relatively low but the loss function is large (see the next paragraph), and they cannot be nano-structured.

Now, let us turn to ENZ media with a small but finite loss at the observation frequency, $1 \gg \varepsilon''(\omega) > 0$. More precisely, we will call it an ENZ material if the real part of permittivity is still the smallest part of it and the loss is small enough, which can be stated as $1 \gg \varepsilon'' \gg |\varepsilon'|$. It is well known that energy loss of charged particles in a medium is proportional to the energy-loss function (see, e.g., Ref. 24, Sec. IIIB), $L(\omega) = -\text{Im}[\varepsilon^{-1}(\omega)]$. For an ENZ material with a very low loss, this loss function *diverges*,

$$L(\omega) \approx 1/\varepsilon''(\omega) \rightarrow \infty \quad \text{for} \quad \varepsilon''(\omega) \rightarrow 0. \quad (3)$$

This diverging singularity of the loss function for ENZ materials will lead to anomalously high energy losses of nanophotonic systems (e.g., plasmonic nanoparticles or chromophores) at the surfaces of or embedded into such materials. Such a paradoxical behavior – singularly high loss in the limit of vanishing internal dissipation ($\varepsilon'' \rightarrow 0$) – is due to the singularly low group velocity in this case, cf. Eq. (2), which is also related to the excitation of bulk plasmons.

In a sharp contrast, for all other types of materials, including dielectrics ($\varepsilon' > 0$) and metals ($\varepsilon' < 0$), where $\varepsilon'' \ll |\varepsilon'|$, the loss function *vanishes* for negligible internal dissipation,

$$L(\omega) = \frac{\varepsilon''(\omega)}{\varepsilon'(\omega)^2 + \varepsilon''(\omega)^2} \approx \frac{\varepsilon''}{\varepsilon'(\omega)^2} \rightarrow 0 \quad \text{for} \quad \varepsilon''(\omega) \rightarrow 0. \quad (4)$$

Note that this regime is of especial interest for a case of $\varepsilon'' \ll |\varepsilon'| \ll 1$ which mimics the ENZ behavior and is realistic in the microwave frequency range [25, 26].

The results of Eqs. (3)-(4) are easy to understand. The energy loss density per unit time, \dot{Q} , for a given electric field, E , oscillating inside a medium is given by an universal expression [20] $\dot{Q} = \frac{\omega}{4\pi} \varepsilon'' |E|^2$. (This can also be equivalently written in terms of the real part of conductivity, σ' , as $\dot{Q} = \sigma' |E|^2$.) Obviously, $\dot{Q} \rightarrow 0$ for low internal dissipation, $\varepsilon'' \rightarrow 0$ (or, equivalently, $\sigma' \rightarrow 0$). The energy loss per unit propagation length of a wave inside the medium is determined by the imaginary part of wave vector, $\text{Im}k = \frac{\omega}{c} \text{Im}\sqrt{\varepsilon' + i\varepsilon''} \rightarrow \frac{\omega}{\sqrt{2}c} \text{Im}\sqrt{\varepsilon''} \rightarrow 0$. In contrast, for given charges oscillating inside the medium, $E \propto \varepsilon^{-1}$, and $\dot{Q} \propto 1/\varepsilon'' \rightarrow \infty$ for ENZ media where $\varepsilon'' \gg |\varepsilon'| \rightarrow 0$.

Behavior of these three measures of losses in ENZ with vanishing internal dissipation is fundamentally different: $\propto \varepsilon''^{-1} \rightarrow \infty$, $\propto \varepsilon'' \rightarrow 0$, and $\propto \sqrt{\varepsilon''} \rightarrow 0$. This implies singularity of the ENZ properties, which, as we have already indicated above in conjunction with Eq. (2), is related to $v_g \rightarrow 0$. Physically, this prevents energy removal from an excitation volume and leads to singularly increased fields, which brings about the high loss function. This can be useful to make efficient (“perfect”) thin absorbers [27, 28]. Noteworthy, there is also degeneracy in mathematical sense for $\varepsilon = 0$ because the coefficient at one of the highest derivatives in the wave-propagation equation (obtained from the Maxwell equations) turns to zero.

As an example of the singular behavior of the ENZ materials, the simplest case is a semi-infinite slab where reflection coefficient R is given by the familiar Fresnel formula [we will consider, for certainty, the p- (or, TM-) polarization]. It is expected that a good ENZ material will be highly reflective ($R \approx 1$) for any non-normal incidence. In reality, as the results of Fig. 1(a) show, even for unrealistically small loss, $\varepsilon(\omega) = 0.03 + i10^{-3}$, there is a low reflection for incidence angle $\theta \leq 12^\circ$ with a pronounced Brewster-angle minimum at $\theta \approx 10^\circ$. As a realistic example, in Fig. 1(a), we also plot results for ITO [29] where for the carrier concentration $n = 6.3 \times 10^{20} \text{ cm}^{-3}$ at the telecommunication vacuum wavelength $\lambda = 1.55 \mu\text{m}$, ENZ conditions are attained: $\varepsilon = 0 + i0.57$. Even for a smooth surface, as Fig. 1(a) shows, the reflection is far from perfect: $R \leq 30\%$ for $\theta \leq 60^\circ$. Introduction of a $\sim 50\text{-nm}$ roughness, as the numerical results obtained by FDTD calculations (Lumerical) show, further decreases R by a factor ~ 0.5 . This low reflectivity is very far from what is conventionally expected for an ENZ material. It is physically related to excitation of bulk plasmons: the ENZ materials are those at the bulk plasmon frequency, ω_P , where $\varepsilon'(\omega_P) = 0$. Note that large losses at $\omega \approx \omega_P$ are known and actively exploited in plasma physics for electromagnetic heating of plasmas [30].

To obtain more insight into nano-optical properties of

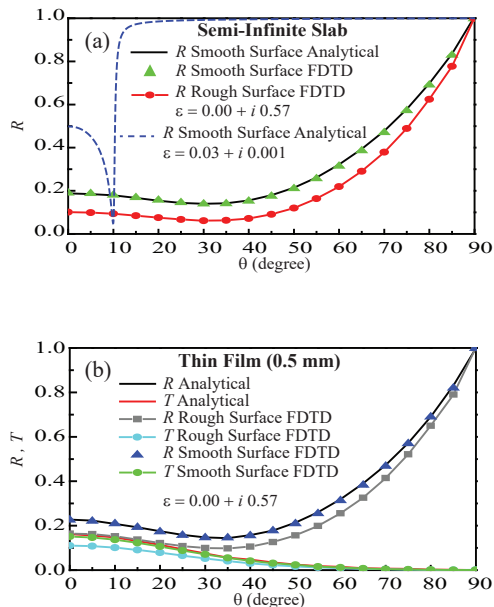


FIG. 1. (Color online) Reflection R and transmission T coefficients of ENZ systems as functions of incidence angle θ . (a) Reflection from a planar surface of semi-infinite ENZ material. Analytical Fresnel reflection coefficient R for smooth surfaces of ITO ($\epsilon = 0. + i0.57$) and idealized low-loss ENZ ($\epsilon = 0.03 + i0.001$). Numerical FDTD reflection coefficient R is displayed for ITO smooth and rough (RMS roughness of 50 nm) surfaces. (b) Analytical and numerical R and T for a 0.5 mm film of ITO ($\epsilon = 0. + i0.57$).

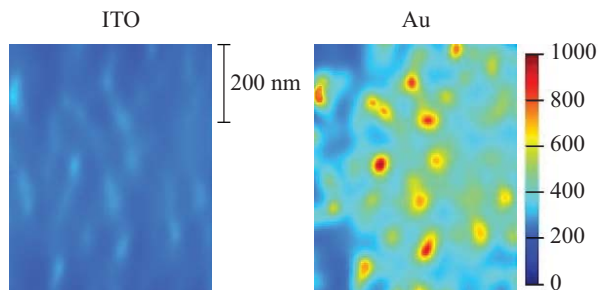


FIG. 2. (Color online) Local field intensity $E^2(\mathbf{r})$ at rough (random Gaussian roughness, MRS 50 nm) surfaces of ITO ($\epsilon = 0. + i0.57$) and gold (dielectric data are adapted from Ref. 31). The excitation radiation is p-polarized incident at 45° . The color scale of intensity (relative units) is indicated to the right of the panels.

ENZ in comparison with conventional plasmonic metals, we display in Fig. 2 local optical field intensity $E^2(\mathbf{r})$ at a rough (random Gaussian roughness, MRS 50 nm) surfaces of ITO ($\epsilon = 0. + i0.57$) and gold [31]. As one can see, the gold surface shows a pronounced picture of nanoscopic hot spots with local fields enhanced by a factor of up to $\sim 10^3$ as expected for a rough plasmonic metal [13, 32, 33]. In a sharp contrast, the ENZ surface of the same geometry does not show such hot spots. This is certainly related to anomalous damping of the plasmonic resonances due to the singularly-enhanced loss function $L(\omega) \rightarrow \infty$ – see Eq. (3) and its discussion above. The hot spots at the rough surface are damped due to the anomalous loss caused by enhanced energy flow into the ENZ.

Now let us consider another important system: a thin film of ENZ where reflection and transmission are also expected to have interesting properties (cf. Ref. 1 and references therein). Such a film with smooth surfaces allows for an exact analytical solution (see, e.g., Ref. 34). The actual analytical results, which are illustrated in Fig. 1(b) for a 0.5 mm ITO film, turn out to be not remarkable: the reflection coefficient is, for most angles, not very high, $R \sim 10\text{--}40\%$, and transmission is rather low due to the losses, typically $T \lesssim 1\text{--}15\%$. We treat a similar thin film with a nanoscopic roughness numerically using the Lumerica package. For the sake of control and testing, we applied this package also to smooth surfaces obtaining an excellent agreement with the analytical formulas – cf. Fig. 1(b). For a nanofilm with rough surfaces [a Gaussian random roughness with root mean-square (RMS) size $\delta = 50$ nm], the reflection is further reduced suggesting the dominating role of loss. In fact, the roughness helps to relax the momentum conservation converting the electromagnetic energy into non-propagating bulk plasmons.

It is widely discussed in the literature that ENZ materials bear high promise for nanoscale waveguiding, which is suggested by the expected strong reflection from ENZ surfaces at all angles [1, 3, 4, 6, 8, 35–40]. Propagation in the plane nanoslit filled by a dielectric material between two semi-infinite slabs made of an ENZ material, which we will call ENZ-I-ENZ waveguide, is amenable to an exact analytical solution. The corresponding dispersion relation is an analytical continuation of the known relation for the metal-insulator-metal (MIM) waveguides [41] and is valid for any values of dielectric functions. For a symmetric waveguide where the ENZ material with dielectric permittivity ϵ surrounds a nanoscopic planar dielectric waveguide with thickness d and permittivity ϵ_d , this dispersion relation for the lower-loss mode of interest, which is a symmetric mode, is

$$\tanh \left[\frac{1}{2} k_0 d \varepsilon_d u(\varepsilon_d) \right] = - \frac{u(\varepsilon)}{u(\varepsilon_d)}, \quad (5)$$

where a function $u(\epsilon)$ is defined as $u(\epsilon) = \frac{1}{\epsilon} \left(\frac{k^2}{k_0^2} - \epsilon \right)^{1/2}$;

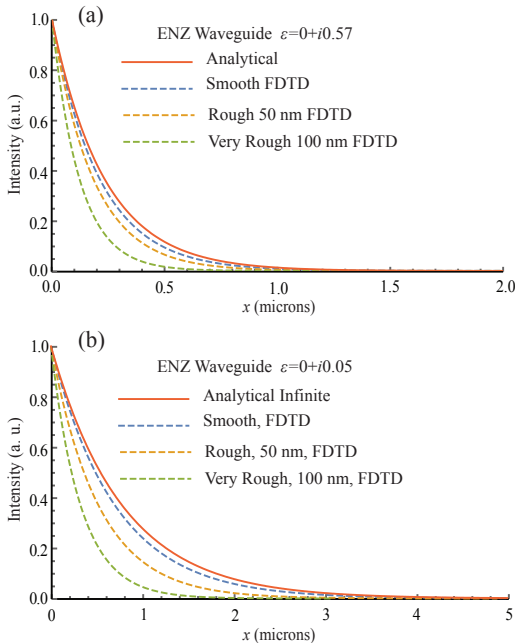


FIG. 3. (Color online) Intensity of wave propagating in ENZ-I-ENZ waveguide with a 500-nm-thick (in the z -direction) SiO_2 dielectric as a waveguide core. The intensity is shown as function of the propagation length (in the x -direction). Analytical results are for a planar waveguide (infinite in the xy -direction). Numerical results are calculated for a $500 \text{ nm} \times 500 \text{ nm}$ rectangular-cross-section SiO_2 waveguide. (a) The ENZ material of the waveguide is ITO at the vacuum wavelength of $1.55 \mu\text{m}$ where $\epsilon = 0. + i0.57$. (b) The ENZ material is idealized with a very low loss, $\epsilon = 0. + i0.05$.

k is the modal wave vector, and $k_0 = \omega/c$ is vacuum wave vector.

For a plasmonic system, where $\epsilon' < -\epsilon_d$, the symmetric mode is highly confined, $k \gg k_0$, and Eq. (5) simplifies,

$$k = \frac{1}{d} \ln \frac{\epsilon - \epsilon_d}{\epsilon + \epsilon_d} \approx -\frac{2}{d} \frac{\epsilon_d}{\epsilon}, \quad (6)$$

where the approximation is valid in a deep plasmonic region, $\epsilon' \ll -\epsilon_d$. Obviously, $k \rightarrow \infty$ for $d \rightarrow 0$, which describes a highly nano-confined guided mode.

In a sharp contrast, for an ENZ material, $k \ll k_0$, and Eq. (5) simplifies to

$$k \approx k_0 \left[\epsilon + \frac{1}{2} (k_0 d \epsilon)^2 \right]^{1/2} \approx k_0 \sqrt{\epsilon}. \quad (7)$$

Thus, the dispersion of the ENZ nano-waveguide is close to that of the embedding ENZ medium, which indicates a very weak mode confinement. In fact, $k \rightarrow 0$ for $|\epsilon| \rightarrow 0$, which is characteristic of the ENZ materials.

We consider numerically a parallel plate ENZ-I-ENZ waveguide whose core is made of a $d = 500 \text{ nm}$ dielectric plate of SiO_2 , surrounded by two semi-infinite ENZ slabs. These are made of ITO at $\lambda = 1.55 \mu\text{m}$, where $\epsilon = 0 + i0.57$. In Fig. 3(a), intensity of the symmetric mode propagating in such an ENZ-I-ENZ waveguide is displayed as a function of the propagation coordinate by a solid red line. As one can see, the real ENZ makes a very poor waveguide: the modal propagation length is only $\approx 0.3 \mu\text{m}$. In comparison, for the same wavelength and geometry, replacing the ENZ by gold (using dielectric data of Ref. [31]), leads to the modal propagation length of $51 \mu\text{m}$. Thus the real ENZ material (ITO) of relatively low loss [14, 15] in the optical range is indeed much inferior to real metals as a waveguiding material. As we have discussed above in conjunction with Eq. 7, this is due to the weak confinement: the energy leaks into the ENZ material where it is absorbed.

For a rectangular SiO_2 waveguide of $500 \times 500 \text{ nm}$ cross section in the ENZ ITO, we used Lumerical package to describe the mode propagation. The results for smooth surfaces, rough surfaces ($\delta = 50 \text{ nm}$) and very rough surfaces ($\delta = 100 \text{ nm}$) are shown in Fig. 3(a) by dashed lines. As one can see, the propagation length in the square waveguide is indeed even shorter than in the parallel-plate one but not by very much. The roughness shortens the propagation length somewhat by increasing coupling to bulk plasmons.

Given the high propagation losses of the waveguides based on ITO as a realistic ENZ material, one may ask how much the loss, i.e., ϵ'' , for a waveguiding ENZ material should be reduced to make it competitive with the real metals in the optical spectral region. To elucidate this question, we plot in Fig. 3(b) results for the same waveguide geometry calculated for an idealized, extremely low-loss ENZ material where dielectric permittivity is set to be $\epsilon = 0 + i0.05$. In this idealized case, the waveguide modal propagation length is, indeed, increased but still it is in a $\lesssim 0.8 \mu\text{m}$ range, i.e., much inferior to gold as a plasmonic waveguiding material.

To conclude, the fundamental principle of causality [as given by Eq. (1)] dictates that any ENZ material with a very low (asymptotically zero) loss at the observation frequency has necessarily asymptotically zero group velocity at that frequency. Physically, this leads to enhanced scattering and dissipative losses as given by the diverging energy-loss function – cf. Eq. (3). Paradoxically, a reduction of the intrinsic loss, $\epsilon'' \rightarrow 0$, leads to an increase of energy-loss function and further deterioration of performance of reflectors and waveguides built from ENZ materials. Both analytically and numerically we have shown that a realistic ENZ material ITO at the bulk plasma frequency causes high reflection and propagation losses. The singular loss function is also responsible for anomalously strong optical damping of resonant systems (plasmonic nanoparticles, dye molecules, quan-

tum dots, etc.) embedded into or positioned at the surfaces of ENZ materials. In contrast to plasmonic metals, there are no pronounced hot spots of local fields at rough ENZ surfaces. Structured dielectric media with practically zero loss in the optical region cannot function as true ENZ materials because of the singular response (3); they necessarily are *diffractive* photonic crystals, and not refractive effective media. Obviously, this anomalous loss of ENZ materials can be gainfully used in energy absorbers, which begets analogy with heating of plasmas at plasma frequency with charged particles or electromagnetic waves. These losses and singularities are fundamental, local properties of the ENZ media, which cannot be eliminated by micro- or nano-structuring.

This work of M.I.S. was supported by Grant DE-SC0007043 from the Physical Behavior of Materials Program, Office of Basic Energy Sciences, the US Department of Energy; M.H.J. was supported by Grant DE-FG02-01ER15213 from the Atomic, Molecular and Optical Sciences Program, Office of Basic Energy Sciences, the US Department of Energy.

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