

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 a 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 nonradiative damping of the 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 materials.

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Recently, materials at frequencies ω close to the bulk plasmon frequency ω_p , which are characterized by the dielectric permittivity ϵ being small enough, $|\epsilon| \ll 1$, 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 materials should totally reflect light at all angles, the phase velocity of light in these materials tends to infinity and, correspondingly, the light wave carries almost constant phase, the density of photonic states in ENZ materials diverges at $\omega \rightarrow \omega_p$, a waveguide formed inside an ENZ material can confine light at deep subwavelength dimensions, there are no reflections even at sharp bands in these materials, and the unavoidable roughness of the waveguide walls does not significantly spoil the waveguiding by ENZ materials. As in many other cases in nano optics [13], dielectric losses present a significant problem, deteriorating these unique properties and limiting the useful applications of ENZ materials. Several approaches have been proposed to mitigate these unwanted effects of optical loss: new conducting oxide and nitride materials, in particular, indium tin oxide (ITO), bear the promise of a significant reduction of 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 whose losses are extremely low [16].

The goal of this Letter is to establish the 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 $\epsilon = \epsilon' + i\epsilon''$ possesses a small real part, $1 \gg |\text{Re}\epsilon| \rightarrow 0$ and a negligible imaginary part $\epsilon'' = \text{Im}\epsilon = 0$) 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 the stationary optical regime diverges $\propto v_g^{-1} \rightarrow \infty$. (Note

that it has been understood that in “perfect lens” systems, where $\epsilon'' \rightarrow 0$ and $\epsilon' \rightarrow -1$, the asymptotically infinite establishment time has already been found earlier [17,18].) Therefore, any dielectric metamaterials emulating $\epsilon' \rightarrow 0$ must be diffractive, i.e., not true metamaterials that transmit and absorb light but that do 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 [19]. Also, the energy loss function $L = -\text{Im}(\epsilon^{-1})$ 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 the remarkable manifestations of ENZ behavior due to the losses.

Because we are interested in the most fundamental properties of ENZ materials, we consider such a material as a uniform and isotropic infinite medium (natural or metamaterial [20]). We also assume that the dielectric response of the ENZ material is local (i.e., there is no spatial dispersion), which ensures that the results are also applicable to micro- and nanostructures 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 the observation frequency ω , i.e., $\epsilon''(\omega) = 0$. We assume that it is also lossless in the infinitesimal vicinity of ω , which we reformulate as the condition $d\epsilon''(\omega)/d\omega = 0$. We will also assume that this material is not magnetic (which is usual at optical frequencies [21]). Under these conditions, the fundamental causality principle leads to an exact dispersion relation [19]

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

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

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

Because of the requirement of stability, $\epsilon'' \geq 0$. Consequently, $v_g \leq c\sqrt{\epsilon'(\omega)}$, and $v_g \rightarrow 0$ for $\epsilon'(\omega) \rightarrow 0$. Thus, a lossless ENZ material in the limit $\epsilon'(\omega) = 0$ does not transport electromagnetic energy. As a corollary, the 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. [22]). Note that Eq. (2) is an exact local property of lossless ENZ materials, and it is not affected by their micro- or nanostructuring.

In the experiment of Ref. [16], the medium designed to be a lossless ENZ metamaterial is, in reality, a diffractive medium—a photonic crystal [23,24] 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 the 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 nanostructured.

Now, let us turn to ENZ media with a small but finite loss at the observation frequency, $1 \gg \epsilon''(\omega) > 0$. More precisely, we will call it an ENZ material if the real part of the permittivity is still the smallest part of it and the loss is small enough, which can be stated as $1 \gg \epsilon'' \gg |\epsilon'|$. It is well known that the energy loss of charged particles in a medium is proportional to the energy-loss function (see, e.g., Ref. [25], Sec. III B) $L(\omega) = -\text{Im}[\epsilon^{-1}(\omega)]$. For an ENZ material with a very low loss, this loss function diverges:

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

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 ($\epsilon'' \rightarrow 0$)—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 sharp contrast, for all other types of materials, including dielectrics ($\epsilon' > 0$) and metals ($\epsilon' < 0$), where $\epsilon'' \ll |\epsilon'|$, the loss function vanishes for negligible internal dissipation:

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

Note that this regime is of especial interest for a case of $\epsilon'' \ll |\epsilon'| \ll 1$, which mimics the ENZ behavior and is realistic in the microwave frequency range [26,27].

The results of Eqs. (3) and (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 a universal expression [21]: $\dot{Q} = (\omega/4\pi)\epsilon''|E|^2$. (This can also be equivalently written in terms of the real part of the conductivity σ' as $\dot{Q} = \sigma'|E|^2$.) Obviously, $\dot{Q} \rightarrow 0$ for low internal dissipation, $\epsilon'' \rightarrow 0$ (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 the wave vector $\text{Im}k = (\omega/c)\text{Im}\sqrt{\epsilon' + i\epsilon''} \rightarrow (\omega/\sqrt{2}c)\sqrt{\epsilon''} \rightarrow 0$. In contrast, for given charges oscillating inside the medium, $E \propto \epsilon^{-1}$, and $\dot{Q} \propto 1/\epsilon'' \rightarrow \infty$ for ENZ media where $\epsilon'' \gg |\epsilon'| \rightarrow 0$.

The behavior of these three measures of losses in ENZ media with vanishing internal dissipation is fundamentally different: $\propto \epsilon''^{-1} \rightarrow \infty$, $\propto \epsilon'' \rightarrow 0$, and $\propto \sqrt{\epsilon''} \rightarrow 0$. This implies a 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 [28,29]. Noteworthy, there is also a degeneracy in the mathematical sense for $\epsilon = 0$ 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 ENZ materials, the simplest case is a semi-infinite slab where the reflection coefficient R is given by the familiar Fresnel formula [we will consider, for certainty, 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 an unrealistically small loss, $\epsilon(\omega) = 0.03 + i10^{-3}$, there is a low reflection for an 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 the results for ITO [30], where for a carrier concentration $n = 6.3 \times 10^{20} \text{ cm}^{-3}$ at the telecommunication vacuum wavelength $\lambda = 1.55 \mu\text{m}$, the ENZ conditions are attained: $\epsilon = 0. + i0.57$. 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 finite difference time domain (FDTD) calculations (Lumerical) show, further decreases R by a factor of ~ 0.5 . This low reflectivity is very far from what is conventionally expected for an ENZ material. It is

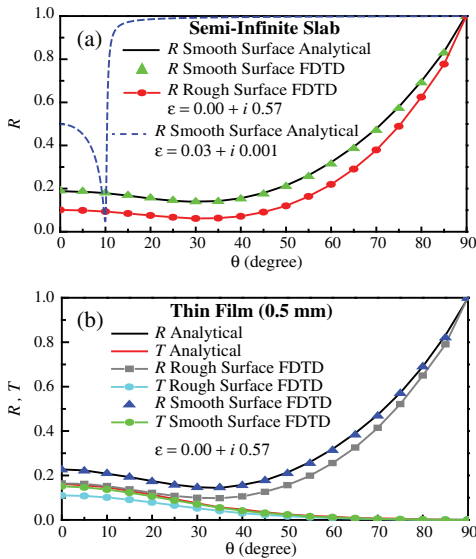


FIG. 1. Reflection R and transmission T coefficients of ENZ systems as functions of the incidence angle θ . (a) Reflection from a planar surface of a semi-infinite ENZ material. Analytical Fresnel reflection coefficient R for smooth surfaces of ITO ($\epsilon = 0. + i0.57$) and an idealized low-loss ENZ material ($\epsilon = 0.03 + i0.001$). The 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 ($\epsilon = 0. + i0.57$).

physically related to the excitation of bulk plasmons: the ENZ materials are those at the bulk plasmon frequency ω_p , where $\epsilon'(\omega_p) = 0$. Note that large losses at $\omega \approx \omega_p$ are known and actively exploited in plasma physics for the electromagnetic heating of plasmas [31].

To obtain more insight into the nano-optical properties of ENZ media in comparison with conventional plasmonic metals, we display in Fig. 2 the local optical field intensity $\mathbf{E}^2(\mathbf{r})$ at rough (random Gaussian roughness, root-mean square (RMS) 50 nm) surfaces of ITO ($\epsilon = 0. + i0.57$) and gold [32]. As one can see, the gold surface shows a pronounced picture of nanoscopic hot spots with the local fields enhanced by a factor of up to $\sim 10^3$ as expected for a rough plasmonic metal [13,33,34]. In a sharp contrast, the ENZ surface of the same geometry does not show such hot spots. This is certainly related to the 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 the enhanced energy flow into the ENZ.

Now, let us consider another important system: a thin film of ENZ material where the 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. [35]). The actual analytical results, which are illustrated in Fig. 1(b) for a 0.5 mm ITO film, turn out to be not

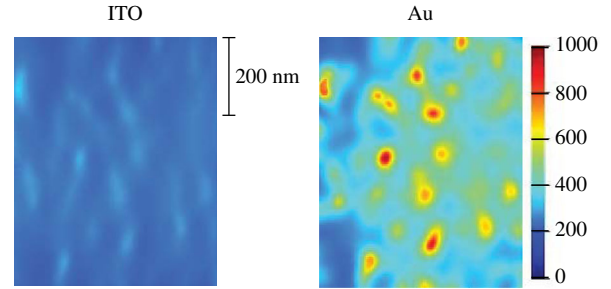


FIG. 2. Local field intensity $\mathbf{E}^2(\mathbf{r})$ at rough (random Gaussian roughness, MRS 50 nm) surfaces of ITO ($\epsilon = 0. + i0.57$) and gold (dielectric data are adapted from Ref. [32]). The excitation radiation is p polarized, incident at 45° . The color scale of the intensity (relative units) is indicated to the right of the panels.

remarkable: the reflection coefficient is, for most angles, not very high, $R \sim 10\%–40\%$, and the transmission is rather low due to the losses, typically $T \lesssim 1\%–15\%$. We treat a similar thin film with a nanoscopic roughness numerically using the Lumerical 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 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 nonpropagating 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 [1,3,4,6,8,36–41]. Propagation in the plane nanogap filled by a dielectric material between two semi-infinite slabs made of an ENZ material, which we will call an ENZ-I-ENZ waveguide, is amenable to an exact analytical solution. The corresponding dispersion relation is an analytical continuation of the known relation for metal-insulator-metal waveguides [42] and is valid for any values of the 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

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

where the function $u(\epsilon)$ is defined as $u(\epsilon) = (1/\epsilon) [(k^2/k_0^2) - \epsilon]^{1/2}$; k is the modal wave vector and $k_0 = \omega/c$ is the 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 nanoconfined guided mode.

In 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 nanowaveguide 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 a characteristic of ENZ materials.

We consider numerically a parallel plate ENZ-I-ENZ waveguide whose core is made of a $d = 500$ nm dielectric plate of SiO₂, 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), the 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 material 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 material by gold (using the

dielectric data of Ref. [32]) leads to a 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 SiO₂ waveguide of 500×500 nm cross section in the ENZ ITO, we used the Lumerical package to describe the mode propagation. The results for smooth surfaces, rough surfaces ($\delta = 50$ nm), and very rough surfaces ($\delta = 100$ 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 the 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 answer this question, we plot in Fig. 3(b) the results for the same waveguide geometry calculated for an idealized, extremely low-loss ENZ material where the 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 the $\lesssim 0.8 \mu\text{m}$ 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 an 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 the energy-loss function and a further deterioration of the 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 losses and propagation losses. The singular loss function is also responsible for the anomalously strong optical damping of resonant systems (plasmonic nanoparticles, dye molecules, quantum 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 an analogy with the heating of plasmas at the plasma

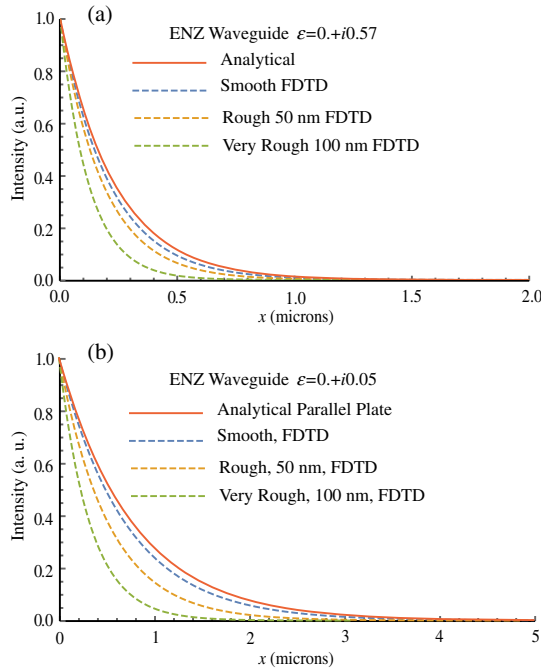


FIG. 3. Intensity of a wave propagating in an ENZ-I-ENZ waveguide with a 500-nm-thick (in the z direction) SiO₂ dielectric as a waveguide core. The intensity is shown as function of the propagation length (in the x direction). The analytical results are for a planar waveguide (infinite in the xy direction). The numerical results are calculated for a 500×500 nm rectangular-cross-section SiO₂ waveguide. (a) The ENZ material of the waveguide is ITO at a vacuum wavelength of $1.55 \mu\text{m}$ where $\epsilon = 0. + i0.57$. (b) The ENZ material is idealized with a very low loss, $\epsilon = 0. + i0.05$.

frequency with charged particles or electromagnetic waves. These losses and singularities are fundamental, local properties of ENZ media, which cannot be eliminated by micro- or nanostructuring.

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