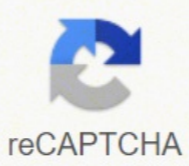




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Enhanced Nonlinear Refractive Index in ϵ -Near-Zero Materials

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Nonlinear propagation of light over long distances is generally low unless there is a resonance or a resonant system. The slow light regime offers a natural solution to this problem. We demonstrate an enhanced nonlinear refractive index in the epsilon-near-zero (ENZ) regime by utilizing the nonlinear refractive index, which enables enhanced light-induced changes in the material properties. Experiments performed in the infrared (IR) show that the ENZ materials exhibit a 10-fold increase in the nonlinear refractive index compared to the ENZ wavelength, located in the ENZ resonance region. This is well beyond the observed light-induced refractive index changes of the order of unity, thus demonstrating a new paradigm for nonlinear optics.

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The nonlinear optical response of matter to light is, by its very nature, a perturbative nonlinear process. Weak effects, applications, e.g., for nonlinear optical switches or optical isolators, are therefore largely underpinned by the continuous enhancement to create stronger and more efficient light-matter interactions. Nonlinear mechanisms are typically classified as resonant or nonresonant, depending on the frequency of light with respect to the characteristic electronic excitation of the material. Resonant mechanisms, like those present in transparent atomic or molecular materials (e.g., doped silica glass), are generally weak and require high light intensities and/or long samples to take advantage of an extended light-matter interaction. Conversely, nonresonant mechanisms can be several orders of magnitude stronger, for this reason in the quest of introducing transparent losses. A typical example is that of metals, which both reflect and absorb light strongly [1–3]. An alternative approach to enhancing the nonlinear response of a material consists of creating artificial electromagnetic resonators, for example, by stacking multiple dielectric layers to form a photonic crystal or composite materials [4–10]. Creating resonant metal-dielectric stacks and composite yields a very strong nonlinear enhancement [11–13], but it inevitably sacrifices the detrimental role of linear and nonlinear losses.

Here, we propose a different approach to enhancing the effective nonlinearity without resorting to optical resonances. The approach relies on enhancing the nonlinear effect, excited in terms of the nonlinear refractive index n_2 , rather than on a direct enhancement of the intrinsic $\chi^{(2)}$ nonlinear susceptibility. In our theory, this enhancement arises due to the fact that the nonlinear refractive index is a function of both the nonlinear susceptibility and the linear refractive index. Recent progress in material design and fabrication has provided access to the full range of linear optical properties bounded by dielectric and metallic regimes [14]. In particular, we focus on the latter set of materials which exhibit a real part of the dielectric permittivity that is close to zero, such as transparent conducting oxides [15] whose linear permeability is typically bounded to the near infrared spectral region.

The linear properties of these "epsilon-near-zero" (ENZ) materials have been investigated [16–17] for applications ranging from controlling the radiation pattern of electron beams [18] to novel transparent irises and optical absorbers. Recently, the nonlinear properties have also been shown to be largely affected by the ENZ condition [19–22], and recently even more theoretically predicted for the coupling between linear and nonlinear properties of ENZ half-metals [23]. However, experimental studies reported so far [24] are limited to glass-coating the condition in two-layer coating [25], enhanced third and second harmonic generation [26–27], and nonlinear optical switching [28].

In order to observe how the nonlinear refractive index can be enhanced to a level of the ENZ linear properties, we employed a 400 nm thick film of oxygen-doped zinc-oxide-doped thin oxide (AZO) [29, 27]. The AZO ENZ thin film was deposited by pulsed laser deposition (PLD) (Pulsar, Inc.) [30, 27] using a 343 nm excimer laser (Lamda Physik GmbH) operating at a wavelength of 248 nm for various excited thicknesses from 200 [27] to 400 nm.

The linear properties, i.e., the real and imaginary parts of ϵ , n , and n_2 , respectively, were measured by a standard



Nonlinear epsilon-near-zero materials explained: opinion

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Abstract: Epsilon-near-zero materials have recently come onto the scene as promising new nonlinear optical materials. However, this field is quite crowded and it is prudent to ask whether they possess any key features which will elevate them above other candidates. It is our opinion that they in fact possess two such features, a simultaneous intrinsic and extrinsic enhancement to light-matter interaction. Here we elucidate these enhancement mechanisms and compare them to other manifestations found in literature, explaining why this combination is unique to epsilon-near-zero materials.

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1. Introduction

The search for exceptional nonlinear materials is as long as the field of nonlinear optics itself. The list of once promising nonlinear media is lengthy and in the end, a majority of them did not live up to their promise for one reason or another as most practical nonlinear devices continue to rely upon the well-established ionic crystals (LiNbO₃, KTP) and on silicon- or gallium-compounds (Si, SiO₂) as in fiber, SiN, GaAs). Typically, the nonlinearity in these materials is weak and for this reason they require either a long length, or the help of photonic structures such as microresonators. To this date, nonlinear optical fiber remains most widely used platform for nonlinear optics for this reason. ENZ materials [1, 2] – a material or spectral region where $Re\{\epsilon\} < 1$ – are the newest entry into the catalogue of nonlinear materials demonstrating exceptional properties from large refractive index tuning [3, 4], enhanced harmonic generation [5], and near unity generation of phase conjugate waves [6]. As a result, they have been projected to become the building blocks for current and future all-optical and electro-optical systems. Yet, it would be prudent to pose a question: is there anything exceptional about ENZ materials that would let them succeed in the brutal nonlinear optics arena where so many others failed ignominiously? It is our opinion that the answer is affirmative, and, moreover, homogeneous ENZ materials such as *transparent oxides possess not one but two salient features* that make them stand out in the crowded field – a high degree of electric field confinement (what we refer below as *extrinsic enhancement*), as well as an ideal balance of strength and speed suited for many applications (to which we refer as *intrinsic enhancement*).

Let us explain this point in a laymen fashion: If one adapts the language of Feynman diagrams [7], optical nonlinearity can be explained as light generating material excitations (typically electron transitions between or within the energy bands) which in turn affect light for as long as they exist. Considering that within the UV to mid-infrared range, the strength of light-matter interaction as represented by an optical dipole is more or less constant according to the oscillator sum rule [8], our goal is to maximize the interaction time – either *intrinsically* by operating close to absorption resonance, or *extrinsically* as in a number of diverse schemes united by a common moniker “slow light”. When it comes to ENZ the enhancement of nonlinearity follows

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