Experimental Verification of \( n = 0 \) Structures for Visible Light

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We fabricate and characterize a metal–dielectric nanostructure with an effective refractive index \( n = 0 \) in the visible spectral range. Light is excited in the material at deep subwavelength resolution by a 30-keV electron beam. From the measured spatially and angle-resolved emission patterns, a vanishing phase advance, corresponding to an effective \( \varepsilon = 0 \) and \( n = 0 \), is directly observed at the cutoff frequency. The wavelength at which this condition is observed can be tuned over the entire visible or near-infrared spectral range by varying the waveguide width. This \( n = 0 \) plasmonic nanostructure may serve as a new building block in nanoscale optical integrated circuits and to control spontaneous emission as experimentally demonstrated by the strongly enhanced radiative optical density of states over the entire \( n = 0 \) structure.

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In the past several years, developments and advances in the field of electromagnetic metamaterials and plasmonic optics have enabled control over the flow of light in engineered nanomaterials, providing the possibility to realize materials with effective refractive indices that range all the way from positive to negative values [1–5]. These developments have led to exciting features and applications, such as subwavelength imaging and superlensing [6,7], hyperlensing [8–11], cloaking and transformation optics [12–16], and subwavelength scale cavities and waveguides [17,18]. Indeed, the ability to tailor the indices of refraction in engineered nanomaterials has opened many new vistas in the design of optical structures with desired characteristics.

While metamaterials with a negative index have been explored recently, metamaterials and structures with a refractive index near zero have received relatively modest attention (e.g., Refs. [19–23]). Metamaterials with low effective permittivity, i.e., epsilon-near-zero (ENZ) metamaterials, have been studied theoretically [24–34], and some of their characteristics have been investigated experimentally in the microwave frequency [27,28] and mid-IR regimes [35]. Since the index of refraction is directly related to the permittivity via \( n = \sqrt{\varepsilon/\mu} \), these structures exhibit near-zero refractive index. These ENZ metamaterials offer unique and exciting wave characteristics and potential applications such as supercoupling [24–29], displacement-current wires and nanocircuit boards [30,31], antenna-to-waveguide coupling [34], and emission enhancement in plasmonic structures, where the Purcell enhancement may occur almost independently of the location of the emitter [32,33]. There are several approaches to achieve metamaterials and structures with ENZ properties and near-zero refractive index. One way is to design metamaterials by stacking subwavelength structures such as thin layers [22,23] or embedding complementary metallic split-ring resonators in host media [36]. Another method is by exploiting the dispersion characteristics of composite metal–dielectric waveguides [27,28]—a technique based on structural dispersion instead of material dispersion that has been utilized in the past to construct structures with tailored dispersion characteristics [37]. In particular, it has been shown that a metallic waveguide operating around its cutoff frequency behaves, under proper conditions, as a structure with an effective refractive index and effective permittivity near zero. This concept has been exploited to experimentally verify some of the ENZ properties, such as supercoupling, at microwave frequencies [27,28].

However, to the best of our knowledge, there has been no experimental realization of ENZ metastructures in the visible spectral range nor a demonstration of the exciting characteristics such as enhanced spontaneous emission almost independent of the location of the emitter [33], because fabrication of these structures at optical length scales presents a challenging set of experimental tasks. It is particularly challenging to place an emitter inside an ENZ structure to probe the optical density of states.

In the present work, we show how these challenges can be overcome by bringing together a combination of experimental, theoretical, and numerical efforts. Our ENZ metastructure design is based on a nanoscale dielectric waveguide core surrounded by a metallic cladding (metal-insulator-metal, MIM waveguides) that is designed to operate at the frequency where the waveguide’s modal dispersion exhibits cutoff, i.e., \( k = 0 \). At this frequency, effectively \( n = 0 \). It is interesting to note that in conventional dielectric waveguides (e.g., waveguides with the core and cladding made of dielectric materials), at the cutoff frequency, the wave number of the mode equals
the propagation constant in the outside medium, and therefore for any frequency below this cutoff, the mode would leak out. Since in such dielectric waveguides, at the cutoff frequency, the wave number is not zero, the phase advance of such a mode is not zero, and thus such a dielectric waveguide at cutoff would not behave as an ENZ or $n = 0$ structure. Here in our geometry we have a MIM rectangular waveguide and therefore near the cutoff frequency the wave number of the mode is small, thus acting similarly to the ENZ and $n = 0$ structures.

We probe the enhanced radiative optical density of states in these ENZ waveguides using a special technique in which we use a 30 keV electron beam as a point dipole source that serves as a spatially resolved probe of the optical density of states.

Figure 1(a) shows a schematic of the waveguide engineered to exhibit $n = 0$ behavior in the visible. The waveguide consists of an 85-nm-thick SiO$_2$ core that is fully surrounded by an Ag cladding. The structure is fabricated using a sequence of evaporation, sputtering, and focused-ion-beam milling of an SiO$_2$/Ag layer structure on a Si substrate. Scanning electron micrographs of the fabricated structures are shown in Figs. 1(b) and 1(c).

To probe the dispersion of the waveguide with deep subwavelength accuracy, we utilize a tightly focused ($\sim 10$ nm) electron beam penetrating the waveguide [38,39] from the top as a broadband point dipole emitter [40]. Analogous to a vertical point dipole, coupling of the radiation field of the electron beam to optical modes of the waveguide [41] is governed by the local density of optical states [40,42]. The far-field radiation emitted by the waveguide, known as cathodoluminescence (CL), is collected in the scanning electron microscopes using a parabolic mirror [43] and analyzed using a spectrometer.

Figures 2(a) and 2(b) show a two-dimensional scan of the CL emission over a 400-nm-wide and 2-$\mu$m-long waveguide. The recorded intensity is plotted for two wavelengths. The scan clearly shows the outline of the waveguide; it is brighter than the surroundings. The CL emission that is excited along the waveguide shows a strong spatial modulation: at 910 nm emission wavelength we observe 6 maxima along the waveguide axis while at 624 nm 11 maxima are found. We attribute these maxima in CL emission to the antinodes of a standing wave [41,44–46] of the surface plasmon polariton mode that is confined in the 2-$\mu$m-long plasmonic Fabry-Perot cavity: the larger number of maxima corresponds to the shorter plasmon wavelength.

The standing wave nodal pattern is plotted for a broad spectral range in Fig. 2(c), which shows a line scan of the CL emission along the waveguide axis; the full emission spectrum is plotted for 5-nm steps along a waveguide with 400-nm width. A large number of resonances is clearly resolved, each corresponding to an integer number of half-wavelengths fitting in the waveguide length. Towards shorter wavelengths, up to 15 antinodes within the waveguide are seen ($\lambda_0 = 500$ nm). The signal vanishes for shorter wavelengths due to increasing losses of the MIM plasmon mode. Figure 2(d) shows a line scan of a 190-nm-wide waveguide. The CL emission for this waveguide shows a completely different behavior. In the 600–700-nm spectral band, this waveguide supports resonances, similar to the wider waveguide, although at a larger period between the maxima. However, at longer wavelengths, the resonances become more closely spaced in the wavelength domain, in contrast to the data for the wider waveguide, where the spacing between the maxima (free spectral range) increases as the wavelength is increased. Correspondingly, for the narrower waveguide, the spacing between maxima along the waveguide rapidly increases with increasing wavelength and at $\sim 800$ nm the standing wave antinodes merge together in a single band. The constant CL intensity along the MIM waveguide at 800 nm and the fact that no antinodes are observed for longer wavelength clearly show that at this wavelength cutoff occurs. At the cutoff wavelength, the electromagnetic field in the waveguide oscillates in phase along the entire waveguide, corresponding to an effective $n = 0$. Since the mode of this waveguide has a quasi-TE character, the behavior near its cutoff resembles the wave in a structure with an effective zero permittivity or $n = 0$ [32,33]. Therefore, this set of experimental results clearly verifies an ENZ structure at visible wavelength.

For wavelengths longer than the cutoff wavelength, no propagating modes exist and the field of a radiating dipole in the waveguide will decay exponentially. Indeed,
Fig. 2(d) shows decaying intensity tails near the ends of the waveguide above 800 nm.

Full-wave numerical simulations utilizing the finite-difference time domain method using the software package LUMERICAL FDTD SOLUTIONS™ [47] were performed using a dipole source placed in the core of the waveguide. Figures 2(e)–2(h) show the simulated total power emitted by the structure, normalized by the power of the dipole emitted in free space. Excellent agreement with the experimental data is obtained, reproducing the standing waves below the cutoff wavelength, a bright band at cutoff, the decaying tails at the waveguide ends, and very low emission above the cutoff wavelength.

To further investigate the relation between cutoff wavelength and waveguide width, Fig. 2(i) shows integrated spectra from a range of waveguides with different widths. Data are taken by integrating spectra over a 500-nm-long line scan along the center of the waveguide. The extent of this region is indicated by the dashed lines in Fig. 2(d). The spectrum for the 190-nm-wide waveguide shows cutoff around 800 nm as evidenced by the sharp decrease in intensity beyond this wavelength, and a number of resonant peaks at shorter wavelengths, as also observed in Fig. 2(d). For narrower waveguides the cutoff wavelength monotonically shifts towards shorter wavelength: for a 120-nm-wide waveguide cutoff occurs at 570 nm. These data show that cutoff, at which the effective permittivity and effective refractive index is near zero, can be controlled over the full visible or near-infrared spectral range by varying the waveguide width from 100 to 200 nm. In all cases, the emission is clearly enhanced at cutoff [33], an effect that is most pronounced at 150 nm width. These experimental results clearly verify the theoretical predictions for the enhancement of emitter radiation placed inside the plasmonic waveguide at cutoff particularly with the unique property of the uniform phase distribution along the waveguide as described in Ref. [33]. As a consequence, in this structure the Purcell enhancement is independent of the location in the waveguide, in strong contrast with other work in which Purcell enhancement is only observed at specific locations, such as, e.g., enhanced spontaneous emission near an interface [48], modified spontaneous
emission near antennas [49], or in metal nanoparticle arrays [50]. Figure 2(j) presents the simulated analog of Fig. 2(i), showing identical behavior of the enhancement as a function of wavelength and waveguide width. Differences in the peak wavelengths between theory and experiment are attributed to small differences in the geometry and material constants.

From the experimental data, the dispersion of the MIM plasmon mode in this waveguide can be derived (see the Supplemental Material for details [51]). Excellent agreement is obtained with the calculated dispersion over the entire experimental spectral range, including the value of the cutoff wavelength. The dispersion has a small slope towards low frequency corresponding to a strong reduction in the group velocity \( v_g = d\omega/dk \), and correspondingly a large increase in density of states. The strongly enhanced CL intensity observed at cutoff is a direct measure of the enhanced density of states. The results are in good agreement with the calculated dispersion in this waveguide, following the theory discussed in Ref. [32].

At cutoff, \( k = 0 \) and there is no phase advance of the light inside the waveguide; all free electrons of the metallic waveguide oscillate in phase, effectively generating a line dipole. To characterize this special \( n = 0 \) condition in detail, we measure the angle-resolved CL emission pattern. The radiation, collected by the parabolic mirror, is directed onto a CCD camera; from the image the azimuthal and polar angular distribution of the emission is derived. Figure 3 shows the emission at 700 nm from two waveguides with widths of 210 and 170 nm that are excited by an electron beam at the center of the waveguide. The waveguides are oriented vertically in the figure. At 700 nm, the 210-nm-wide waveguide is far from cutoff and its dispersion lies below the light line. Hence, when this waveguide is excited, the emission at 700 nm mostly emerges from the waveguide ends. Figure 3(a) shows a clear fringe pattern which is attributed to interference of the emission from the two waveguide ends. The narrower 170-nm-wide waveguide is at cutoff at 700 nm. The oscillation in the waveguide is therefore in phase and above the light line, generating a line dipole and thus allowing direct coupling of the plasmon resonance to the far field to the side of the guide (i.e., normal to the line dipole). The measured angular emission pattern at 700 nm in Fig. 3(b) clearly shows two lobes corresponding to the angular emission pattern of a line dipole antenna. This confirms that the waveguide oscillates in phase \( (k = 0) \). Figures 3(c) and 3(d) show the simulation results using the software package CST MICROWAVE STUDIOTM [52] for the emission patterns, which reproduces both the interference fringes above cutoff and the line dipole emission pattern at cutoff where \( n = 0 \). The simulations are carried out at 800 nm wavelength, which is the simulated cutoff wavelength for the 170-nm-wide waveguide. Because of slight differences in the cutoff wavelength between the experiment and the simulation as well as slight differences in the end facets, Fig. 3(a) shows the emission of an odd mode whereas Fig. 3(b) shows an even mode [53].

In conclusion, we have fabricated and characterized a nanostructure that guides light with vanishing phase advance \( (n = 0) \), using a metal-insulator-metal waveguide at cutoff, verifying several important features of the ENZ metasurfaces including enhanced emission at cutoff, and uniform phase distribution along the waveguide. Correspondingly, the Purcell enhancement is essentially independent of the location of the emitter placed inside the plasmonic waveguide. Resonant guided plasmon modes buried in the metal-insulator-metal stack were excited using a 30 keV electron beam as a point source exciting the waveguide. Strong modal dispersion is observed, in particular, near the cutoff frequency. The observation of an effective \( n = 0 \) is corroborated by angle-resolved measurements of the emission that show that the waveguide radiates in a line dipole antenna emission pattern. The metal-insulator-metal plasmon waveguides operating at \( n = 0 \) provide a key new tool to control dispersion in nanoscale integrated optical circuits and nanolasers and enable enhanced radiative decay rates and superradiance in active metamaterials. By realizing three-dimensional architectures [54] of coupled plasmonic
$n = 0$ waveguides, bulk metamaterials with such properties may be envisioned.

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**Viewpoint**

**Metal-Coated Waveguide Stretches Wavelengths to Infinity**

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*A zero refractive index at optical frequencies, as demonstrated in a new nanoscale waveguide, enables opportunities for better control and enhancement of light propagation in waveguides, as well as development of photonic nanocircuits.*

Subject Areas: **Optics, Metamaterials, Plasmons**

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**A Viewpoint on:**

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The index of refraction is arguably the most important parameter for an optical medium and is critically pertinent to most, if not all, of the phenomena of light in optical materials and devices. Recent advances in metamaterials, which are artificially structured media with engineered electromagnetic properties, have provided us with an unprecedented ability to realize unconventional values of the refractive index that were not considered possible in the past. Now, writing in *Physical Review Letters* [1], Ernst Vesseur of the FOM Institute for Atomic and Molecular Physics (AMOLF), Netherlands, and his colleagues have demonstrated a metallic waveguide that exhibits a zero refractive index at visible frequencies. Light in such a material has a spatially uniform phase, characterized by a quasi-infinite wavelength.

What we know about the index of refraction of various materials has progressed over many centuries. Early investigations into geometrical optics introduced this dimensionless quantity as a way to characterize the ability of a material to bend light rays, as described by Snell’s law. Later it was discovered that the refraction of light in an anisotropic material is direction sensitive, and in this situation the refractive index possesses the form of a tensor rather than a scalar value. With the development of wave optics, the refractive index is now recognized as a measure describing the reduction of the speed and wavelength of light compared to their free-space counterparts, following the simple relations $n = c/n$ and $\lambda = \lambda_0/n$. (The frequency $\omega = 2\pi c/\lambda_0 = 2\pi \nu / \lambda$ doesn’t vary in “linear” optical materials.) In a general case where the wave has a propagation factor $e^{-i(\omega - k z)}$, the refractive index (and consequently the wave vector $k = \omega n / c$) assumes a complex value $n = n' + i n''$, where $\exp(i2\pi n' z /\lambda_0)$ describes the phase advance the wave acquires during its propagation, and $\exp(-2\pi n'' z /\lambda_0)$ gives the exponential decay of the field magnitude. The past decade has witnessed an explosive development of metamaterials, as researchers have striven to develop and utilize man-made materials with anomalous refractive indices, ranging from the negative to the infinite (Fig. 1, top), with exotic applications such as super-resolution lenses and invisibility cloaks [2].

Recently, growing attention has been devoted to materials with near-zero index of refraction, which are sometimes referred to as epsilon-near-zero (ENZ) materials due to the connection between the refractive index and the dielectric constant $n = \sqrt{\epsilon}$. Within an ENZ material, the wave exhibits little or no spatial variation, as the wavelength $\lambda = \lambda_0/n$ approaches infinity (see Fig. 1, top middle). In addition, the speed of light in the material, as indicated by $\nu = c/n$, becomes superluminal, posing an apparent contradiction to relativity. However, this velocity is the “phase velocity,” which describes the rate at which the phase of the wave propagates. In contrast, the optical power or information travels at the “group velocity,” which is limited by relativity, so causality still holds. To give a specific example, consider noble metals like silver or gold, whose conducting electrons can be approximated as an electron gas. In this case, the index of refraction is $n = \sqrt{1 - (\omega_p/\omega)^2}$, where $\omega_p$ is the so-called plasma frequency. Based on the definitions of the phase velocity $\nu_p = \omega / k$ and the group velocity $\nu_g = d\omega / dk$, we obtain $\nu_p = c/\sqrt{1 - (\omega_p/\omega)^2}$ and $\nu_g = c\sqrt{1 - (\omega_p/\omega)^2}$. Consequently, a zero index occurs at the plasma frequency, where $\nu_p \to \infty$ and $\nu_g \to 0$. In general, any material with $|n| < 1$ will be dispersive (with the index of refraction depending on wavelength), and any modulated signal in the material will travel below the speed of light in vacuum, albeit with a superluminal phase velocity for certain frequencies.

Besides noble metals, other homogeneous materials may exhibit a near-zero index in a specific wavelength...
range, such as polar dielectrics, heavily doped semiconductors, and some metal oxides. Artificially structured materials also have a long history as a viable route to near-zero refractive index. Examples include a metallic wire mesh that mimics an electric plasma with tailored plasma frequency [3], stratified structures with alternating metallic and dielectric layers that provide an anisotropic ENZ medium, and metamaterials with resonant inclusions that allow impedance matching [4]. Moreover, the recent developments in negative-index metamaterials offer zero index as a by-product. The reasoning is as follows: all negative-index media are dispersive, and the refractive index is a continuous function of wavelength, so there must exist a frequency between the negative-index band and the (positive-index) non-resonant spectral range at which the effective index is exactly zero.

In the present work, Vesseur et al. demonstrate a near-zero index utilizing the cutoff frequency in a metallic (plasmonic) waveguide built at the nanoscale. The basic idea dates back to earlier microwave studies from half a century ago, but it has been revived very recently at optical frequencies [5]. An ENZ material in a waveguide configuration opens up many applications, such as subwavelength channel propagation [6]. The strategy takes advantage of the particular dispersion relation between frequency and wave vector in a metallic waveguide. A normal dielectric waveguide has a dispersion relation that cuts off at the point where it crosses the light line (\(\omega/k = c/n\)) of the low-index cladding medium (which is air in the example given in Fig. 1, bottom). Below this cutoff frequency, light leaks out of the waveguide. In contrast, a metallic hollow waveguide possesses a dispersion curve that starts near the light line of the embedded dielectric and ends at the vertical axis where \(k = 0\) (Fig. 1, bottom). A vanishingly small wave vector at a nonzero frequency indicates a waveguide mode with near-zero index \(n_{eff} = k_0/k\), as well as a phase velocity \(\nu_p = \omega/k\) approaching infinity.

The waveguide used by Vesseur et al. has a rectangular glass core (85 by 190 nanometers), surrounded by a silver cladding on all sides. Embedding an emitting source inside an ENZ material can be challenging, but the researchers overcame this by using an electron beam to excite the plasmonic waveguide and thus probe the optical density of states. They observed the near-zero-index mode occurring near the far-red end of the visible spectrum, but they can potentially tune this wavelength by altering the dimensions of the dielectric slot. This tunability in the waveguides is a significant advantage over other ENZ materials, like noble metals and polar dielectrics, where the wavelength of zero index is fixed. There also exists the possibility of packing many of these waveguides together into an array to form a bulk metamaterial with a zero effective index [7].

A vanishing refractive index is certainly more than a mathematical trick. It enables far-reaching possibilities for both fundamental physics and exciting novel applications. The spatially uniform phase distribution (or vanishing phase advance) for the zero-index mode implies directed and enhanced radiation from emitters either embedded inside the medium [3], or externally excited as shown by Vesseur and coauthors. Potential applications of zero-index materials include arbitrary wave front design, supercoupling with subwavelength channels, air-core waveguides, innovative lenses, insulating media for optical lumped nanocircuits, and many more [9, 10].

By stretching the wavelength of light at optical frequencies, zero-index materials provide an interesting contrast to surface plasmons, which have a flat dispersion relation (see Fig. 1, bottom) that shrinks light at optical frequencies down to x-ray wavelengths. Once again, the ever-evolving field of nanophotonics challenges our tradi-
tional understanding of optics, and offers unconventional means to manipulate light waves.

References


About the Author

Wenshan Cai

Wenshan Cai joined the faculty of Georgia Institute of Technology as an associate professor in January 2012. Prior to this, he worked as a postdoctoral fellow at Stanford University for three years after receiving his Ph.D. from Purdue University in 2008. His impact on the evolving field of plasmonics and metamaterials is exemplified by over 3,000 citations of his recent papers. He authored the book Optical Metamaterials: Fundamentals and Applications (Springer, 2010), which is used as a textbook or reference at many universities around the world. He leads the Laboratory for Advanced Photonics and Optoelectronics (LAPO) at Georgia Tech, which is focused on the design and implementation of nanophotonic materials and devices.