ACS Photonics

pubs.acs.org/journal/apchd5 Letter

Photon Acceleration Using a Time-Varying Epsilon-near-Zero Metasurface

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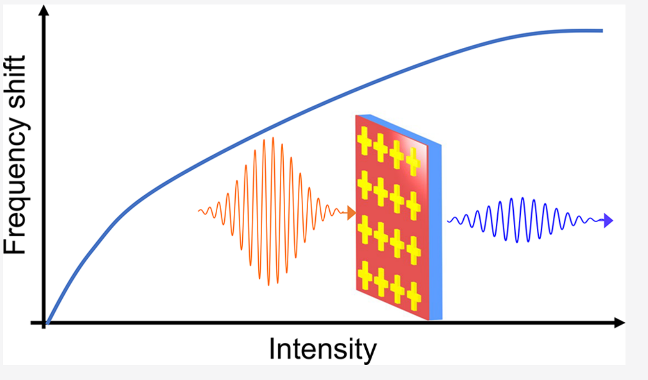
Cite This: https://dx.doi.org/10.1021/acsphotonics.0c01929 Read Online

ABSTRACT: A light beam's frequency can blueshift when the beam travels through a medium that exhibits a time-dependent decrease in the refractive index. Here we show that a metasurface made of a plasmonic antenna array on a thin indium tin oxide (ITO), which exhibits epsilon-near-zero (ENZ) response, can behave as a time-varying medium and change the frequency of a sufficiently strong light beam through self-action effect. Specifically, we observe that a near-resonant optical excitation of the 92 nm thick metasurface leads to an intensity-dependent blueshift of the excitation pulse. We measured a maximum blueshift of ~1.6 THz with ~4 GW/cm^2 incident intensity. The observed effect using an ITO-based ENZ metasurface has an energy requirement that is up to 200× lower than implementations using ITO alone.



KEYWORDS: *time-varying medium, epsilon-near-zero, metasurface, frequency conversion, nonlinear optics, plasmonics*

An ENZ material exhibits a near-zero real permittivity, and consequently, a small linear refractive index, over a certain spectral range. They exhibit intriguing linear effects such as energy squeezing through narrow channels and arbitrary bends,[1−3] geometry-invariant resonance,[4−6] quantum optical properties,[7] and unprecedentedly large nonlinear optical responses.[8,9] It has recently been demonstrated that large spectral translations of a probe beam can be induced by pumping an ITO-based thin ENZ film through an adiabatic frequency conversion (AFC) process.[10−12] Prior work by Zhou et al. explains the AFC process by time-refraction, that is, a pump-induced, time-dependent change in the refractive index (Δn) of an ITO film leads to a frequency blueshift or a redshift, depending on the sign of Δn. In this work we investigate a complementary pathway for frequency translation: a frequency blueshift of a beam through a self-action effect where a carrier frequency of the pump beam changes due to the time-varying effect induced by the same beam. This effect was first experimentally observed in the optical domain using gaseous plasma and is known as photon acceleration (PA).[13−15] PA is a specific case of the time-refraction process that results in only frequency blueshift. We note that our experimental observation can also be explained as a delayed-time self-phase modulation effect.[16]

A high-intensity laser pulse propagating through an ionization front experiences a rapidly decreasing refractive index, leading to a spectral blueshift of the light beam. While propagating through such a medium, the leading edge of the pulse experiences a higher index than its trailing part. As a result, the local phase velocity increases over the duration of the pulse. For a sufficiently long propagation distance, the trailing edge of the pulse accelerates to catch up to the leading edge, resulting in temporal compression, as well as spectral broadening and blueshifting of the pulse. If the change in the refractive index occurs at a time scale that is slower than the characteristic oscillation time of the carrier frequency, this process leads to AFC through a self-action effect (Figure 1).

To demonstrate the self-action-induced PA effect, we consider a plasmonic ENZ-based metasurface. Such an engineered metasurface exhibits a nonlinear response that is orders of magnitude larger than the nonlinear response of the

Received: December 20, 2020

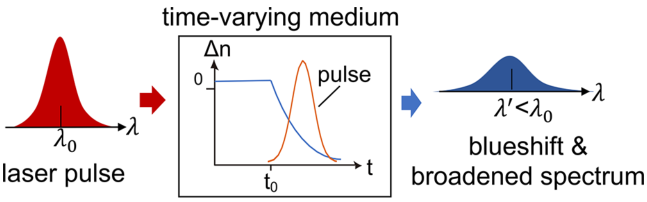


Figure 1. Conceptual illustration of photon acceleration. When a laser pulse propagates in a medium exhibiting a time-dependent refractive index (n), a decrease in n results in a blueshift of the pulse and a broadening of the spectrum.

ENZ medium itself.[17−22] The operation power requirement in such a metasurface is reduced compared to that of a bare ENZ film due to (a) the plasmonic-antenna-enhanced near-field interactions with the ENZ layer, (b) increased coupling efficiency of the electromagnetic energy into the ENZ layer, and (c) dynamic tuning of the antenna resonance due to the change in the refractive index of the ENZ layer (Figure 2a−c).

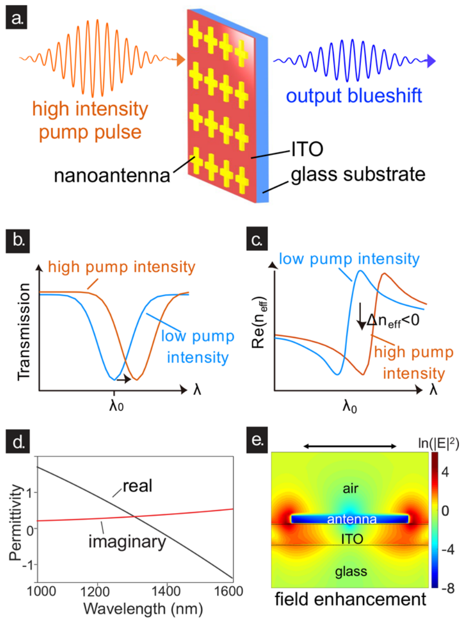


Figure 2. (a) Schematic of the metasurface structure: An ENZ-based metasurface can be optically induced as a time-varying medium, where (b) a dynamic redshift of the resonance wavelength (λ_0) leads to (c) a decrease in the effective refractive index of the metasurface at λ_0. This results in a frequency blueshift of a sufficiently intense pump pulse. (d) The measured linear permittivity of the ITO layer using ellipsometry. (e) The numerically obtained field enhancement in the vicinity of an antenna at the λ_0 of the metasurface when pumped with an electric field polarized along the horizontal axis, as indicated by the horizontal arrow in the figure. The vertical view of the field enhancement in $\ln(|E|^2)$ is plotted from the center of one antenna unit.

Consequently, the metasurface also exhibits an effective change in the refractive index Δn that is as large as $|2.5|$.[17] We note that there have been several other demonstrations of AFC using solid-state systems, such as a high-Q optical cavity and photonic crystal waveguides.[23−25] In all of those cases, a beam needs to propagate through many wavelength-long materials for a frequency shift of less than 200 GHz.

Here, we report that nonlinear interactions of a near-IR pump beam with a 92 nm thick ENZ metasurface leads to a frequency blueshift of ∼1.6 THz with an incident intensity of ∼4 GW/cm^2 due to the PA effect. That incident intensity is up to 200× lower than the previous demonstrations using pure ENZ thin films.[10,11] We define the performance of such an interaction as the achievable frequency shift per unit required intensity per unit nonlinear propagation distance, which is 4.35 × 10^{-3} GHz·(GW/cm^2)$^{-1}$·μm^{-1} of this ENZ metasurface. We note that in our metasurface the thick glass substrate is for structural integrity and does not induce any nonlinear response. Therefore, the nonlinear propagation distance for our metasurface is the total thickness of the ITO film and the antenna. Furthermore, we only use this performance metric for the ease of comparison with the reported demonstrations of the effect using different nonlinear media. This performance metric for the metasurface reported in this work is up to 7 orders of magnitude larger than previous demonstrations of PA in gaseous plasmas.[14,15] We also demonstrate that the observed blueshift is accompanied by a spectral compression of the pump beam when operated near the metasurface resonance, which is due to the anomalous dispersion.

The metasurface investigated in this work comprises an array of a 27 nm thick gold plasmonic antenna on top of a 65 nm thick ITO layer on a 1 mm thick float glass substrate. The ITO film exhibits a zero real permittivity at ∼1390 nm wavelength, as measured using spectroscopic ellipsometry (Figure 2d). A thin ITO−ENZ film exhibits a number of leaky and surface modes.[26] Due to the near-field interactions between the ITO and the antenna array, the metasurface exhibits a large field enhancement inside the ITO layer (Figure 2e). We present numerically calculated dispersion curves in the wavelength−wavevector space in Figure 3. Close to the zero-permittivity

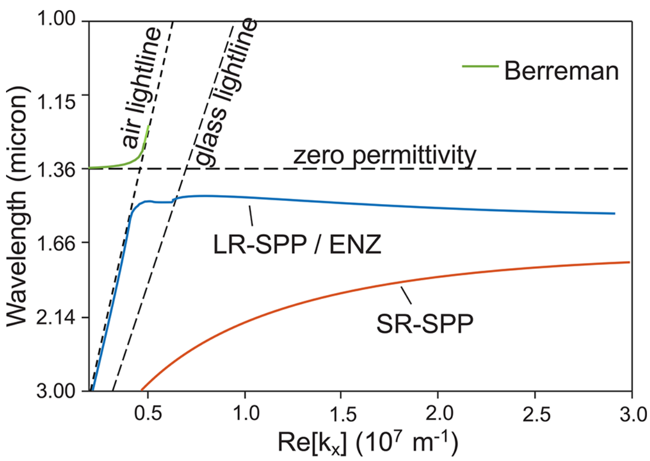


Figure 3. Calculated dispersion relations of the ITO layer in the metasurface using the FDTD method. LR-SPP: Long-range surface plasmon polariton, which is also referred to as the ENZ mode; SR-SPP: Short-range surface plasmon polariton.

frequency, the thin ITO film supports an ENZ mode that resides outside the light line cone. The polarization-insensitive plasmonic antenna array is fabricated using e-beam lithography on top of the ITO film. We choose the dimensions of the plasmonic antenna array (inset of Figure 4) such that the resonance wavelength, in the absence of the ITO layer, is ∼1300 nm. Figure 4 shows the simulated (using FDTD) and the experimentally obtained linear responses of the metasurface measured using a thermal light source. The antenna array geometry is chosen to be cross-shaped such that there is no

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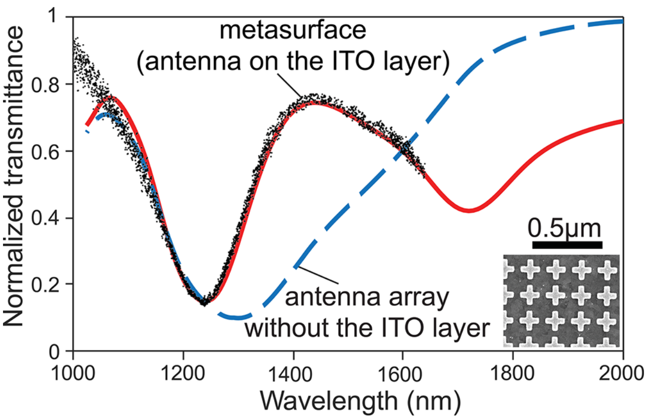

Figure 4. Normalized linear transmittance of the ENZ metasurface (black dots: experimental measurement; red line: FDTD simulation). The blue dashed line shows the simulated transmission resonance of the antenna array in the absence of the ITO layer. The inset shows a SEM image of the fabricated metasurface. Each antenna unit has a dimension of 125 × 450 nm, with a periodicity of 600 nm.

polarization dependence in the linear or nonlinear response. We note that the measurements shown in Figure 4 are insensitive to the input light linear polarization. In the presence of the ITO layer, the localized plasmonic mode of the antenna array interacts with the ENZ mode of the ITO layer. This near-field mediated interaction leads to a strong coupling-induced resonance splitting of ∼500 nm. This splitting is larger than the line widths of both the plasmonic resonance and the ENZ mode. The strong coupling-induced resonance splitting results in two distinct dips in the linear transmittance response of the metasurface.[17,18,20,27] We observe a dominant resonance at ∼1240 nm and a weaker resonance at ∼1720 nm. As presented in Figure 4, the two resonances are clearly visible in the numerical simulation (red line). The main dip is observed in the measured linear transmission (black line) of the metasurface. We note that (i) the simulation and experimental results of the linear transmission are close, and we attribute the minor disagreements to various fabrication imperfections; (ii) the weaker resonance of the metasurface at ∼1720 nm is out of the measurement range of our optical spectrum analyzer (OSA), and thus, we could not measure it in the laboratory.

We measure the nonlinear response using a single pump configuration. The pump pulse, obtained from the output of a Ti:Sapph laser-pumped optical parametric amplifier, has a pulse duration of ∼60 fs and a repetition rate of 1 kHz. We set the central wavelength of the pump pulse to be at ∼1240 nm, which is nearly resonant with the main dip of the metasurface. An adjustable free-space attenuator is used to vary the intensity of the pump light. The average power is measured using a calibrated photodiode. We use a 200 mm long focal length lens to focus the beam onto the metasurface with a measured focal spot size of 250 μm. The transmitted light from the metasurface is collected by a multimode fiber that has a core diameter of 50 μm with the help of another 100 mm focal length lens. The collected light is then sent to an OSA to record the spectral information as a function of incident intensity. Four representative normalized spectra of the measurements are presented in Figure 5a. In Figure 5b we show the central frequency of the pump light, as collected after the metasurface, versus the incident intensity. We observe that the central frequency of the pump blueshifts with an increase in pump intensity and that the maximum achievable blueshift of ∼1.6 THz occurs with a pump intensity of ∼4 GW/cm^2. For

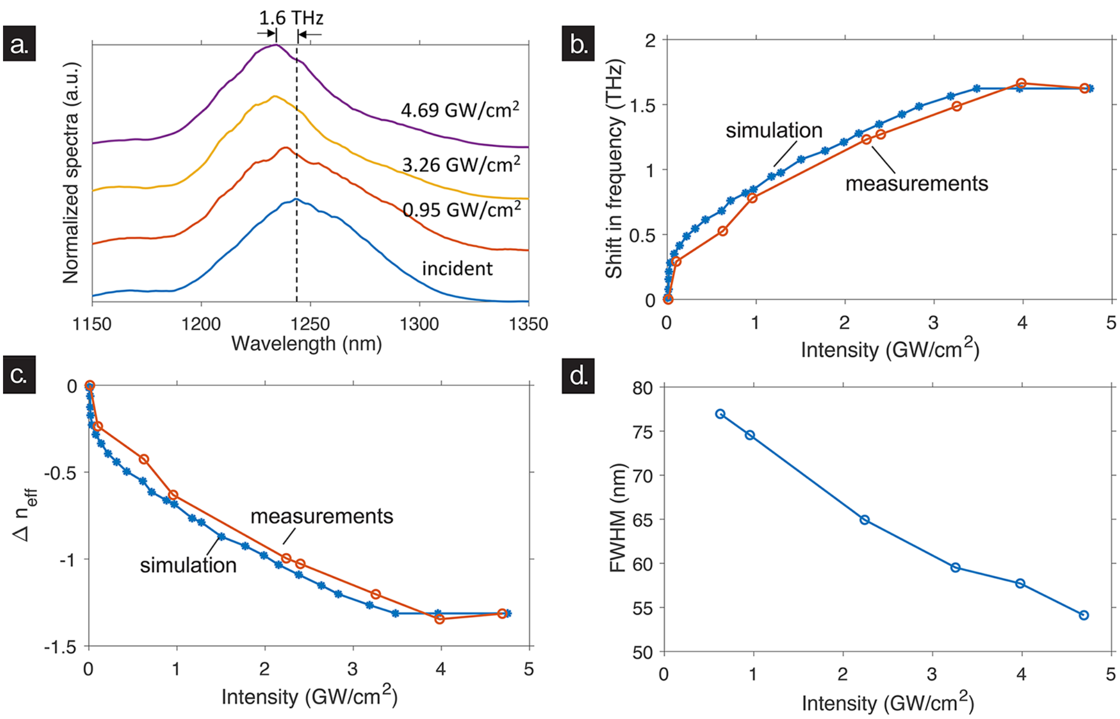

Figure 5. (a) Four representative intensity-dependent measured output spectra of the metasurface. The spectral amplitudes are normalized to their respective peak values recorded by the spectrum analyzer. (b) The pump frequency shifts as functions of the pump intensity. (c) The intensity-dependent Δn_{eff} induced by the pump pulse. The blue curves are simulation results, while the orange curves are experimental results in (b) and (c). (d) The measured full width at half-maximum (FWHM) of the output pulses as a function of incident intensity.

https://dx.doi.org/10.1021/acsphotonics.0c01929
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an incident pump intensity larger than ~4 GW/cm^2, the frequency shift saturates.

To verify the model of AFC, we perform a set of FDTD simulations to numerically obtain the nonlinear frequency shift. We model the ITO layer with a delayed, normalized $\chi^{(3)}$ response while setting the intrinsic nonlinearity of the gold layer to zero. We note that, in our simulation, $\chi^{(3)} \cdot |E|^2$ leads to the effective nonlinear change in the refractive index (Δn_{eff}), as observed in our experiment. The results of the numerical simulation are shown in Figure 5b. The simulation results are in excellent agreement with the experiment, confirming our hypothesis that the large nonlinearity of the ITO layer is primarily responsible for the overall nonlinear response of the system.

Next, we estimate the total intensity-dependent change in the Δn_{eff} of the material as experienced by the pump from the experimental and the simulation data. To estimate the Δn_{eff}, we can express the change in the frequency as $\Delta f = -(kL/2\pi)(\Delta n_{\text{eff}}(t)/\Delta t)$.[8] Here, k is the free space wavevector and L is the thickness of the metasurface. Since the time scale of the nonlinear dynamics is set by the pump pulse duration, we can set Δt to be equivalent to the pump pulse duration. We present these results in Figure 5c. We find that, under pulsed excitation, the metasurface exhibits an absolute Δn_{eff} that is as large as ~1.45, which is equivalent to the linear refractive index of most common optical materials (e.g., glass). We note that, although the Δn of the ITO layer is positive due to the intraband dynamics,[8] the Δn_{eff} of the coupled structure is negative.[17] This is because a positive Δn of the ITO layer leads to a beam that is resonant with the structure to become nonresonant due to the time-varying change of the resonant condition.

We observe that the frequency blueshift in the experiment is accompanied by a spectral compression (Figure 5d). This observation is in contrast to previous reports of adiabatic frequency blueshift[10] and theoretical predictions[28] showing that, in a normally dispersive medium, the frequency blueshift is accompanied by spectral broadening. We attribute our observation due to near-resonant excitation of the coupled system; we inherently operate in the anomalous dispersion regime. Due to the large Δn, the pulse also experiences a large time-varying change in the dispersion condition. We posit that a time-dependent change of anomalous dispersion compresses the spectra in a dual fashion to a temporal compression obtained using a time-invariant anomalously dispersive medium. Our findings indicate the possibility of spectral and, by extension, temporal manipulation of an ultrafast pulse using dispersion engineering in the time domain.

In conclusion, we have demonstrated that a time-varying ENZ-based metasurface can be used for frequency blueshifting. We emphasize that the nonlinear effect we observed in an ENZ metasurface has a significantly reduced propagation distance and an energy requirement that is many orders of magnitude smaller compared with the implementation using gaseous plasma. Furthermore, due to the extraordinarily large and ultrafast effective decrease in the refractive index, a time-varying change in the dispersion condition can be exploited for the spectral manipulation of an ultrafast pulse in the time domain. The findings may find practical applications in designing on-chip or fiber-based frequency translators and in investigating light−matter interactions at the nanoscale using a time-varying medium.

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https://dx.doi.org/10.1021/acsphotonics.0c01929
ACS Photonics XXXX, XXX, XXX−XXX

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Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

This research was developed with funding from the Defense Advanced Research Projects Agency (DARPA) under the NLM Nascent Light−Matter Interactions Program (Grant No. W911NF-18-0369).

■ REFERENCES

(1) Silveirinha, M.; Engheta, N. Tunneling of electromagnetic energy through subwavelength channels and bends using ε-near-zero materials. *Phys. Rev. Lett.* **2006**, *97*, 157403.
(2) Liberal, I.; Engheta, N. Near-zero refractive index photonics. *Nat. Photonics* **2017**, *11*, 149−158.
(3) Engheta, N. Pursuing near-zero response. *Science* **2013**, *340*, 286−287.
(4) Liberal, I.; Mahmoud, A. M.; Engheta, N. Geometry-invariant resonant cavities. *Nat. Commun.* **2016**, *7*, 10989.
(5) DeVault, C. T.; Zenin, V. A.; Pors, A.; Chaudhuri, K.; Kim, J.; Boltasseva, A.; Shalaev, V. M.; Bozhevolnyi, S. I. Suppression of near-field coupling in plasmonic antennas on epsilon-near-zero substrates. *Optica* **2018**, *5*, 1557−1563.
(6) Schulz, S. A.; Tahir, A. A.; Alam, M. Z.; Upham, J.; De Leon, I.; Boyd, R. W. Optical response of dipole antennas on an epsilon-near-zero substrate. *Phys. Rev. A: At., Mol., Opt. Phys.* **2016**, *93*, 063846.
(7) Lobet, M.; Liberal, I.; Knall, E. N.; Alam, M. Z.; Reshef, O.; Boyd, R. W.; Engheta, N.; Mazur, E. Fundamental radiative processes in near-zero-index media of various dimensionalities. *ACS Photonics* **2020**, *7*, 1965−1970.
(8) Alam, M. Z.; De Leon, I.; Boyd, R. W. Large optical nonlinearity of indium tin oxide in its epsilon-near-zero region. *Science* **2016**, *352*, 795−797.
(9) Caspani, L.; Kaipurath, R. P. M.; Clerici, M.; Ferrera, M.; Roger, T.; Kim, J.; Kinsey, N.; Pietrzyk, M.; Di Falco, A.; Shalaev, V. M.; Boltasseva, A.; Faccio, D. Enhanced nonlinear refractive index in ε-near-zero materials. *Phys. Rev. Lett.* **2016**, *116*, 233901.
(10) Zhou, Y.; Alam, M. Z.; Karimi, M.; Upham, J.; Reshef, O.; Liu, C.; Willner, A. E.; Boyd, R. W. Broadband frequency translation through time refraction in an epsilon-near-zero material. *Nat. Commun.* **2020**, *11*, 2180.
(11) Khurgin, J. B.; Clerici, M.; Bruno, V.; Caspani, L.; DeVault, C.; Kim, J.; Shaltout, A.; Boltasseva, A.; Shalaev, V. M.; Ferrera, M.; Faccio, D.; Kinsey, N. Adiabatic frequency shifting in epsilon-near-zero materials: the role of group velocity. *Optica* **2020**, *7*, 226−231.
(12) Bruno, V.; Vezzoli, S.; DeVault, C.; Carnemolla, E.; Ferrera, M.; Boltasseva, A.; Shalaev, V. M.; Faccio, D.; Clerici, M. Broad frequency shift of parametric processes in epsilon-near-zero time-varying media. *Appl. Sci.* **2020**, *10*, 1318.
(13) Wilks, S.; Dawson, J.; Mori, W.; Katsouleas, T.; Jones, M. Photon accelerator. *Phys. Rev. Lett.* **1989**, *62*, 2600.
(14) Lopes, N.; Figueira, G.; Dias, J.; Silva, L.; Mendonça, J.; Balcou, P.; Rey, G.; Stenz, C. Laser pulse frequency up-shifts by relativistic ionization fronts. *EPL (Europhysics Letters)* **2004**, *66*, 371.
(15) Wood, W. M.; Siders, C.; Downer, M. Measurement of femtosecond ionization dynamics of atmospheric density gases by spectral blueshifting. *Phys. Rev. Lett.* **1991**, *67*, 3523.
(16) Boyd, R. W. *Nonlinear Optics*; Academic Press, 2020.
(17) Alam, M. Z.; Schulz, S. A.; Upham, J.; De Leon, I.; Boyd, R. W. Large optical nonlinearity of nanoantennas coupled to an epsilon-near-zero material. *Nat. Photonics* **2018**, *12*, 79−83.
(18) Yu, N.; Capasso, F. Flat optics with designer metasurfaces. *Nat. Mater.* **2014**, *13*, 139−150.
(19) Boyd, R. W.; Gehr, R. J.; Fischer, G. L.; Sipe, J. Nonlinear optical properties of nanocomposite materials. *Pure Appl. Opt.* **1996**, *5*, 505.
(20) Reshef, O.; De Leon, I.; Alam, M. Z.; Boyd, R. W. Nonlinear optical effects in epsilon-near-zero media. *Nature Reviews Materials* **2019**, *4*, 535−551.
(21) Jun, Y. C.; Reno, J.; Ribaudo, T.; Shaner, E.; Greffet, J.-J.; Vassant, S.; Marquier, F.; Sinclair, M.; Brener, I. Epsilon-near-zero strong coupling in metamaterial-semiconductor hybrid structures. *Nano Lett.* **2013**, *13*, 5391−5396.
(22) Bruno, V.; DeVault, C.; Vezzoli, S.; Kudyshev, Z.; Huq, T.; Mignuzzi, S.; Jacassi, A.; Saha, S.; Shah, Y. D.; Maier, S. A.; Cumming, D. R. S.; Boltasseva, A.; Ferrera, M.; Clerici, M.; Faccio, D.; Sapienza, R.; Shalaev, V. M. Negative refraction in time-varying strongly coupled plasmonic-antenna−epsilon-near-zero systems. *Phys. Rev. Lett.* **2020**, *124*, 043902.
(23) Preble, S. F.; Xu, Q.; Lipson, M. Changing the colour of light in a silicon resonator. *Nat. Photonics* **2007**, *1*, 293−296.
(24) Upham, J.; Tanaka, Y.; Asano, T.; Noda, S. On-the-fly wavelength conversion of photons by dynamic control of photonic waveguides. *Appl. Phys. Express* **2010**, *3*, 062001.
(25) Kondo, K.; Baba, T. Dynamic wavelength conversion in copropagating slow-light pulses. *Phys. Rev. Lett.* **2014**, *112*, 223904.
(26) Campione, S.; Brener, I.; Marquier, F. Theory of epsilon-near-zero modes in ultrathin films. *Phys. Rev. B: Condens. Matter Mater. Phys.* **2015**, *91*, 121408.
(27) Campione, S.; Wendt, J. R.; Keeler, G. A.; Luk, T. S. Near-infrared strong coupling between metamaterials and epsilon-near-zero modes in degenerately doped semiconductor nanolayers. *ACS Photonics* **2016**, *3*, 293−297.
(28) Xiao, Y.; Agrawal, G. P.; Maywar, D. N. Spectral and temporal changes of optical pulses propagating through time-varying linear media. *Opt. Lett.* **2011**, *36*, 505−507.