Hyperbolic metamaterial feasible for fabrication with direct laser writing processes

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Stimulated-emission-depletion-microscopy-inspired direct laser writing (STED-DLW) processes can offer diffraction-unlimited fabrication of 3D structures, not possible with traditional electron-beam or optical lithography. We propose a hyperbolic metamaterial for fabrication with STED-DLW. First, we design meandering wire structures with three different magnetic dipoles which can be excited under different incidences of light. Then, based on effective parameters corresponding to normal incidence and lateral incidence, we find that the hyperbolic dispersion relation for a five-layer structure appears between 15 and 20 THz. Finally, we investigate the influence of imaginary parts of the effective parameters on the metamaterial dispersion. The proposed metamaterial structure also has potential for three-dimensionally isotropic permeability despite geometric anisotropy. © 2015 Optical Society of America

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1. INTRODUCTION

The field of electromagnetic metamaterials has provided us with a new look at the materials by mimicking nature through electromagnetic engineering on subwavelength scales. This has led to the possibility of previously unthought-of applications such as flat lens [1], perfect lens [2], hyperlens [3–6], ultimate illusion optics [7–9], perfect absorber [10,11], quantum levitation [12], optical analogue simulators [13–15], compact antennas [16,17], solar photovoltaics [18], metaspaters [19], and many others.

Hyperbolic metamaterials [20–22] have emerged as one of the most interesting and promising subclasses of metamaterials after negative index metamaterials [1,23–25] with practical applications ranging from subwavelength imaging [3–6] to the engineering of spontaneous [26–34] and thermal emission [35]. In 2006, a far-field optical lens with a resolution beyond the diffraction limit was theoretically proposed [3]. This lens was called a “hyperlens,” because the key feature of the lens enabling subwavelength resolution arises from the hyperbolic dispersion of the metamaterials from which the lens is built. The hyperlens allows image magnification and is more robust with respect to losses as opposed to Pendry’s perfect lens [2]. In 2007, the hyperlens was experimentally demonstrated [4]. The fabricated structure showed a subdiffraction resolution of 130 nm under 365 nm ultraviolet (UV) illumination. In 2010, a spherical version of the hyperlens operating at 410 nm visible wavelength and resolving features down to 160 nm was designed and fabricated [6]. This was the first experimental demonstration of a far-field imaging device at a visible wavelength, with resolution beyond the diffraction limit in two lateral dimensions.

Furthermore, it was predicted theoretically and demonstrated experimentally that the spontaneous emission rate of a dipole emitter would be significantly enhanced near or inside the material with hyperbolic dispersion due to the high photonic density of states [26–29,34]. In an experiment, a multilayer metamaterial with hyperbolic dispersion was used for the demonstration of broadband enhanced spontaneous emission from nitrogen-vacancy centers in nanodiamonds [31]. Hyperbolic metamaterials incorporating quantum emitters were proposed for efficient and directional single photon sources for potential applications in quantum information [30]. Roughened surfaces of hyperbolic metamaterials consisting of silver nanowire arrays grown in alumina membranes were demonstrated to scatter light preferentially inside the metamaterial [32]. In a similar experiment, multilayer hyperbolic metamaterial covered with indium-tin-oxide nanoparticles was shown to reduce reflection [33], which may be useful for high-efficiency solar cells and photodetectors. Besides spontaneous emission engineering by controlling the photonic density of states, hyperbolic dispersion was also shown to provide broadband thermal emission beyond the blackbody limit in the near field due to the thermal excitation of unique bulk metamaterial modes [35]. Other found implications of hyperbolic dispersion are extremely high field enhancement [36] and giant optical forces [37] in waveguides.
Among the natural materials, triglycine sulfate and sapphire exhibit hyperbolic dispersion at far-infrared frequencies, bismuth at terahertz frequencies, and graphite at UV frequencies [38, 39]. To date, most optical metamaterial structures have been fabricated by well-established two-dimensional (2D) fabrication technologies, such as e-beam lithography and evaporation of metal films. However, these can only allow stacking of several planar functional layers [40, 41]. Concerning the fabrication of optical hyperbolic metamaterials, layered metal–dielectric structures [4, 6, 28, 29, 31, 33, 42–45] and nanowire arrays [26, 32, 46–50] have appeared as two common approaches. The largest sample size of $1 \, \text{cm} \times 1 \, \text{cm} \times 51 \, \mu \text{m}$ was achieved with nanowire arrays fabricated by electrochemical deposition of a metal on a porous alumina membrane [41]. Multilayer fishnet structures [51] and graphene metamaterials [52, 53] with hyperbolic dispersion have been theoretically proposed. However, to fabricate truly bulk optical metamaterials [54, 55] a three-dimensional (3D) fabrication approach is needed. Particularly, the practical realization of hyperbolic metamaterial devices such as the hyperlens, which is one of the most captivating manifestations of hyperbolic dispersion, demands three-dimensional volume structures.

Direct laser writing (DLW), based on two-photon polymerization, can enable the fabrication of truly bulk and computer-controlled arbitrarily shaped 3D complex structures [56–59] that are not possible with traditional photolithographic processes [60]. DLW has an important potential in fabrication of metamaterials, especially at frequencies ranging from mid-IR to visible, since it offers a viable route as a low-cost and rapid prototyping tool for truly 3D fabrication of nanostructures. Fabrications of large-area, complex metallic nanostructures [61] and metamaterials [57, 58] have been demonstrated with DLW and subsequent metallization. With stimulated-emission-depletion-microscopy-inspired direct laser writing (STED-DLW) [62–65], a feature size reduction by more than a factor of two has been demonstrated [66]. Additionally, with the combination of STED-DLW and the “dip-in” approach [67, 68] metamaterial height can reach the level of 1 mm—where one can think about constructing macroscopic metamaterials [68].

Here, inspired by the rapid progress in DLW technologies, we propose the first blueprint of a hyperbolic metamaterial structure amenable to fabrication with STED-DLW processes followed by electroplating of gold [58]. The structure has operating frequencies at mid-IR frequencies and the features within the resolution of state-of-the-art STED-DLW technologies.

### 2. PHYSICAL GEOMETRY

The unit cell of the hyperbolic metamaterial structure consists of two pairs of meandering wires with inversion symmetry (see Fig. 1). The dimension of the unit cell is $2513 \, \text{nm} \times 2513 \, \text{nm} \times 2290 \, \text{nm}$. The wires are modeled by using experimental Drude model parameters for bulk gold with plasma frequency of $f_p = 2180 \, \text{THz}$ and collision frequency of $f_c = 19.1 \, \text{THz}$ as given in [58]. The simulations are performed by using the finite-integration-method-based CST Microwave Studio software package. Frequency domain solver is used to calculate the $s$-parameters corresponding to the complex reflection and transmission coefficients. Then, these $s$-parameters are used to retrieve the effective medium parameters of the metamaterial [69]. Unit cell boundary conditions are chosen to impose the periodic or quasi-periodic boundary conditions as necessary in the simulations. The tetrahedral meshes with the adaptive meshing method are selected to accurately represent the models to be simulated.

### 3. PHYSICAL MECHANISMS AND EFFECTIVE PARAMETERS

Different magnetic dipoles [70] with close resonance frequencies can be excited inside the structure in Fig. 1 under different incidences of light. The resonances underlying the magnetic dipoles arise from the combined capacitance and inductance of the nanocircuits inside the structure similar to split-ring-resonators. Figure 2 schematically illustrates three different magnetic dipoles that can be excited under different incidences of light. Color arrows represent the current loops for the induced magnetic dipoles. The planes of these current loops are perpendicular to the respective incident magnetic field $H$, since the dipoles are magnetically excited. Below we refer to the current loops in the $y$–$z$ plane [see Fig. 2(a)] and the $x$–$z$ plane [see Fig. 2(b)] as column loops, and the current loops in the $x$–$y$ plane [see Fig. 2(c)] as the joint loops. In particular, Fig. 2(a) illustrates the magnetic dipoles excited by normal incidence. Here, we define the normal incidence such that the incident wave vector $k$ is along the $z$ direction and the structure is periodic in the $x$–$y$ plane. This is the simplest configuration for fabrication with DLW, where the structures are grown on the substrate parallel to the $x$–$y$ plane, and subsequent optical characterization. Optical response under this configuration is polarization-independent in the $x$–$y$ plane. On the other hand, Fig. 2(b) illustrates the magnetic dipoles excited by TE-polarized laterally incident light. We define the lateral incidence such that the incident $k$-vector lies in the $x$–$y$ plane parallel to the substrate and the structure is periodic in the plane perpendicular to the $k$-vector. In this case, the TE-polarized light is described as the electromagnetic field with fixed electric field $E$ along the $z$ direction. In contrast, for the TM-polarized laterally incident light electric field $E$ is replaced with $H$, as shown in Fig. 2(c), where the corresponding magnetic dipoles are also illustrated.

#### A. Effective Parameters and Field Distributions for Different Incidences of Light

In this part, we verify the induced magnetic dipoles illustrated schematically in Fig. 2 based on calculated current density
distributions and show the results for the retrieved effective optical parameters for single-layer metamaterial structures.

First, we consider the configuration in Fig. 2(a) where the structure interacts with normally incident light. Figure 3(a) shows the resultant transmittance (T), reflectance (R), and absorbance (A). In Fig. 3(b), we plot the retrieved effective refractive index, $n = n' + i n''$. Retrieved effective permittivity, $\varepsilon = \varepsilon' + i \varepsilon''$, and permeability, $\mu = \mu' + i \mu''$, are shown in Figs. 3(c) and 3(d), respectively. Notice that a magnetic resonance with a Lorentzian-like lineshape [71,72] appears around 26 THz and $\mu''$ is negative between 26 and 30 THz. The ratio of the vacuum wavelength to unit cell size in the propagation direction (i.e., $\lambda/a$ ratio) is about 5, which is reasonably large for homogenous effective medium approximation, near the magnetic resonance. Figures 3(e) and 3(f) show the current density distribution at $f = 27$ THz (i.e., near the magnetic resonance frequency). This verifies the column loops illustrated in Fig. 2(a).

When the structure interacts with laterally incident light such as in Figs. 2(b) and 2(c), then two other magnetic resonances originate depending on the polarization of incident light. Figure 4 shows the case for the TE-polarized light. There exists magnetic resonance around 22 THz. In this case, the $\lambda/a$ ratio is also about 5. The resultant current density in the $x$ direction (i.e., $J_x$) is excited by $H$ perpendicular to the plane of the loops, and the current density in the $y$ direction (i.e., $J_y$) is excited by $H$ parallel to the plane of the loops.
distribution, which verifies the column loops illustrated in Fig. 2(b), is shown in Figs. 4(e) and 4(f). Finally, Fig. 5 shows the results for the TM-polarized light. Particularly, Figs. 5(e) and 5(f) show the current density distribution at 40 THz near the magnetic resonance, which verifies the joist loops illustrated in Fig. 2(c). For this case, the \( \lambda/a \) ratio is about 3. Although the structure might not seem to be sufficiently subwavelength under this configuration, we should note that the results are
still reliable, because (i) no discontinuities are observed in the retrieved results and (ii) the retrieved refractive index is below the first Brillouin zone edge.

B. Hyperbolic Dispersion

The magnetic dipoles discussed above can be used to obtain hyperbolic dispersion. As an example we choose the magnetic dipoles in Fig. 3. We start with considering the TE-polarized electromagnetic waves propagating in the $x$–$z$ plane. The electric field is fixed along the $y$ direction. When the incident $k$-vector changes its direction from the $z$ direction to the $x$ direction, the corresponding incident field configuration changes from Figs. 3–5. Therefore, one might expect hyperbolic dispersion around the region where $\mu'_z < 0$ [see Fig. 3(d)] since $\mu'_z > 0$ in the same region. However, we show below that imaginary parts also have important contribution to the dispersion of the metamaterial.

In order to demonstrate the hyperbolic dispersion relation, we calculate the tangential and normal components of the effective wave vector inside the metamaterial which are defined as $k_x$ and $k_z$, respectively. The inverted Fresnel formula \[ k_{z,d} = \pm \cos^{-1}\left(1 - r^2 + t^2\right) + 2\pi m, \] (1)

where $d$ is the unit cell thickness along the propagation direction. $r$ and $t$ are the reflection and transmission coefficients, respectively. $m$ is the branch number. The sign is chosen to guarantee a positive imaginary part of $k_z$. $r$ is selected to promise a continuous real part of $k_z$. On the other hand, at the vacuum–metamaterial interface the tangential components of the wave vectors are continuous. Thus, $k_x$ can be expressed as $k_x = k_0 \sin \theta$, where $k_0$ is the wave number in free space and $\theta$ is the angle of incidence with respect to the surface normal. Different angles of incidence are set up in the CST simulations and corresponding reflection and transmission coefficients along with $k_x$ values are obtained. $k_z$ values are then calculated from Eq. (1). Finally, based on different pairs of $k_z$ and $k_x$, we obtain the equifrequency contours describing the dispersion for the metamaterial.

The equifrequency contours for a one-layer structure (i.e., single unit cell along the $z$ direction and infinite in the $x$–$y$ plane) gives elliptical dispersion contrary to anticipated hyperbolic dispersion due to the contribution of relatively large imaginary parts of effective optical parameters. However, increasing the number of layers of the proposed metamaterial structure leads to a transition from elliptical dispersion to hyperbolic dispersion. For example, the equifrequency contours in Fig. 6 correspond to the five-layer structure (i.e., five unit cells along the $z$ direction and infinite in the $x$–$y$ plane) which manifests hyperbolic dispersion. The blue, green, red, and cyan lines represent the frequencies of 15, 16, 18, and 20 THz, respectively.

In Figs. 7 and 8, we show the effective parameters for two five-layer structures with different configurations. Figure 7 shows effective permittivity and permeability under normal incidence with the same geometric configuration as in Fig. 6. Comparing Fig. 7 with Fig. 3, we notice that the magnetic resonance becomes weaker and redshifts from about 26 to 20 THz with an increasing number of layers. The structure homogenizes rather slowly as can be seen from relatively different effective parameters compared to a single layer.

Figure 8 shows effective permittivity and permeability for the five-layer structure under $y$-polarized laterally incident light with the same geometric configuration as in Fig. 5, except that there exists five layers along the $x$ direction. Around 20 THz, we observe an electric resonance, which does not appear in the one-layer structure. This suggests that unlike the magnetic resonances, the electric resonance arises from the interaction between neighboring unit cells rather than an intra-unit cell effect.
In the following we show that the retrieved effective parameters in Figs. 7 and 8 are consistent with the hyperbolic dispersion displayed in Fig. 6. For the TE-polarization considered in Fig. 6, because $E$ is along the $y$ direction and the vectors $k$ and $H$ are in the $x\,–\,z$ plane, only $\varepsilon_y$, $\mu_x$, and $\mu_z$ contribute to hyperbolic dispersion, which can be expressed as

$$\frac{k_x^2}{\varepsilon_y \mu_z} + \frac{k_z^2}{\varepsilon_y \mu_x} = \frac{\omega^2}{c^2}. \quad (2)$$

We consider, for example, $f = 20$ THz in Fig. 6. From the retrieved results in Figs. 7 and 8, $\varepsilon_y = 0.6869 - i0.7442$, $\mu_x = 1.006 + i1.345$, and $\mu_z = 1.016 - i0.2175$. The negative imaginary parts in $\varepsilon_y$ and $\mu_x$ are due to inherent periodicity effects commonly seen in metamaterials [74]. First, considering $\theta = 0^\circ$ (i.e., normal incidence), in Eq. (2) we substitute $k_x = 0$, $k_z$ calculated from Eq. (1), and the retrieved parameters, $\varepsilon_y$ and $\mu_x$ (see Fig. 7), we find the corresponding complex frequency as $19.912 + i1.3258$ THz, which is very close to 20 THz.

When $\theta \neq 0^\circ$, $\mu_z$ is also needed. However, the retrieved $\mu_z$ in Fig. 8 does not accurately describe the equiphase contours in Fig. 6, because the structure corresponding to Fig. 6 has infinite unit cells in the $x\,–\,y$ plane and only 5 unit cells along the $z$ axis. In contrast, the structure corresponding to Fig. 8 has 5 unit cells along the $x$ axis and infinite unit cells in the $y\,–\,z$ plane. Although the numbers of unit cells in different directions are different, the optical properties of these two structures should be qualitatively similar since they are two different pieces of the identical bulk metamaterial. Therefore, starting with the above $\mu_z$ as a guess value and feeding this into Eq. (2) we find through an iterative process that the best fitting value is $\mu_z = 1.75 - i0.6175$. Although the real part is relatively large compared to $\mu_z$ in Fig. 8, the negative imaginary part implies that electric resonance seems to persist.

4. DISCUSSION

Based on the above analysis, the imaginary part of the effective parameters plays an important role on the type of metamaterial dispersion. In the previous reports (see, for example, [51]), since the imaginary parts are usually small compared to the real parts, considering only the real parts of the effective parameters alone are usually sufficient to predict the type of metamaterial dispersion. While, here, the imaginary parts of the effective parameters are of the same order as the real parts or even larger. Therefore, contrary to the metamaterials with low imaginary parts in the effective parameters, the one-layer structure, here, results in elliptical dispersion with $\varepsilon_y > 0$, $\mu_x > 0$, and $\mu_z > 0$ and the five-layer structure results in hyperbolic dispersion with $\varepsilon_y > 0$, $\mu_x > 0$, and $\mu_z > 0$. We should note that, in the latter, especially the negative imaginary part of $\mu_z$ around electric resonance as a result of periodicity effects has an important contribution to the transition from elliptical to hyperbolic dispersion. Despite this sharp transition in optical properties as the monolayer structure is transformed into a multiple-layer stack (i.e., this can be regarded analogous to graphene versus graphite), the structure slowly homogenizes with an increasing number of layers and approaches a bulk hyperbolic metamaterial.

The metamaterial structures, here, were designed specifically for fabrication with DLW processes and subsequent metallization. The functional optical metamaterials resulting from this fabrication approach are usually free-standing structures in air [57, 58, 75]. Therefore, vacuum was selected as a background material in our simulations. Direct metallization in the dielectric host media using DLW is also possible [76, 77]. However, no functional optical metamaterial with this approach has been shown.

If the meandering wires are embedded in a dielectric media with a larger refractive index than vacuum, we find that the resonances and effective material properties redshift. Therefore, it is expected that the hyperbolic dispersion should also redshift with a larger refractive index. Considering the underlying resonant magnetic dipole modes (see Fig. 2) the redshift in magnetic resonances in Figs. 3–5, for example, can be easily explained by a simple LC circuit model [18, 71, 78]. Effectively, the meandering wires in Figs. 3 and 4 behave similar to a two-gap split-ring resonator (SRR) and the meandering wires in Fig. 5 behave similar to a four-gap SRR. Embedding these SRR-like structures inside host media with a larger refractive index than vacuum results in an increase in equivalent circuit capacitance, hence a redshift in resonance frequency.

On the other hand, if we decrease the length or the diameter of the wires, blueshift occurs in optical magnetic response, because decreasing the length of the wires reduces the equivalent circuit impedance and decreasing the diameter of the wires by keeping the wire positions fixed reduces the equivalent circuit capacitance due to larger gaps. Thus, in both cases magnetic resonance frequency blueshifts consistent with the LC circuit model.

We should note that the retrieved effective parameters above are obtained by inverting transmission and reflection coefficients in accordance with the homogeneous effective medium (HEM) approximation discussed in [69, 73, 74]. This retrieval procedure uniquely returns the impedance ($z = z' + iz''$) and $n''$ by making use of the physical requirements that $z' > 0$ and $n'' > 0$ for passive material. However, there exists an ambiguity in determining $n'$ due to multiple solutions. Once this ambiguity is resolved, the effective permittivity ($\varepsilon$) and permeability ($\mu$) are determined from $\varepsilon = n/z$ and $\mu = nz$, respectively, without any constraint on imaginary parts of $\varepsilon$ and $\mu$. In our case, $n'$ was obtained from the continuous 0th order branch under the first Brillouin zone edge, which in turn was verified by (1) the resultant resonances in effective constitutive parameters that are consistent with the field distributions and (2) multiple-layer simulations.

The origin of resultant commonly observed negative imaginary parts in the retrieved constitutive parameters under the HEM approximation has been extensively investigated [74, 79–84]. It was shown that these negative imaginary parts near the resonances arise from the inherent periodicity of the metamaterial if the actual periodic metamaterial structure is approximated by a HEM with the same scattering parameters as the periodic structure [74, 85]. However, every inhomogeneous medium exhibits spatial dispersion (i.e., polarization and magnetization at a given location depends on the spatial distribution of the fields) [82], which is not considered in
we plot in the same graph the effective DLW processes. For discussions on fabrication of the structures with STED-cussions on meandering wire structures and Martin Wegener. We would like to thank Costas M. Soukoulis for fruitful dis-

Finally, in Fig. 2 we plot in the same graph the effective permeability values corresponding to three different magnetic dipoles illustrated in Fig. 2 and studied in Figs. 3–5. It is worth mentioning that three effective permeability values intersect nontrivially around 40 THz (i.e., convergence below 20 THz is uninteresting due to asymptotic nonmagnetic response at low frequencies). This shows that the structure has potential for three-dimensionally isotropic permeability despite geometric anisotropy. Despite operating at a single frequency, considering the difficulty of making isotropic metamaterials at optical frequencies, this result is still interesting. Moreover, the structure may be optimized to operate over a wider bandgap. Tunability with the incorporation of, for example, liquid crystals can also be utilized to mitigate the bandwidth issue.

5. CONCLUSION
In summary, we propose a hyperbolic metamaterial structure operating at mid-infrared frequencies. The structure is feasible to fabricate using combined state-of-the-art STED-DLW technolo-

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