Single-material semiconductor hyperbolic metamaterials

D. Wei,1 C. Harris,2 C. C. Bomberger,1 J. Zhang,1 J. Zide,1 and S. Law1,*

1Department of Materials Science and Engineering, University of Delaware, Newark, Delaware 19716, USA
2Department of Chemistry and Physics, Lincoln University, Oxford, Pennsylvania 19352, USA

*slaw@udel.edu

Abstract: Layered semiconductor hyperbolic metamaterials for the mid-infrared are grown by molecular beam epitaxy using a single material system, doped and undoped InAs. The onset wavelength for metamaterial behavior can be tuned from 5.8 μm to beyond 10 μm, while the fill factor ranges from 0.25 to 0.75, resulting in designer optical behavior. The reflection and transmission behavior were studied by Fourier transform spectroscopy and modeled using effective medium theory. We also conducted a geometric optics experiment to demonstrate negative refraction of our materials.

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References and links

1. Introduction

Hyperbolic metamaterials (HMMs) have been of great interest recently, due to their unique optical properties. Normal materials exhibit a closed optical dispersion surface which restricts the wavevectors of propagating light. In HMMs, the optical dispersion surface is an open hyperboloid, which allows light with large wavevectors to propagate within the material, rather than decay exponentially close to the surface of the sample [1,2]. The propagation of high-k light can be harnessed for diverse applications including subwavelength imaging [3,4], emission engineering [5–8], focusing and waveguiding [9–11], and more [12].

There are a variety of ways to create HMMs; two common ways are embedding a regular array of subwavelength metallic rods within a dielectric matrix or building a superlattice of subwavelength layers of metal and dielectric [5,11,13,14]. This paper will focus on the latter type. These composite materials act as effective media due to the subwavelength nature of their components [15–17]. In general, HMM properties are determined both by the optical properties of the constituent metals and dielectrics as well as the details of the subwavelength geometry. Most research using these types of HMMs has focused on visible wavelength devices using traditional metals (gold, silver) and traditional dielectrics (alumina, silicon) with good success. However, there are many applications at longer wavelengths that could benefit from metamaterials, including infrared sensing, emission, waveguiding, and imaging. Moving HMMs to the infrared is not merely a matter of changing the subwavelength geometry. In order for an HMM to exhibit hyperbolic dispersion, the magnitude of the permittivity of the metal components (|ε_m|) must be similar to the permittivity of the dielectric (|ε_r|).

Traditional metals have large negative permittivities in the infrared: |ε_m|~1000. This large permittivity makes it impossible to find a matching dielectric. In addition, many traditional dielectric materials like alumina have strong absorption resonances in the infrared, making...
device design and data interpretation complicated. In order to take advantage of HMMs in the infrared, we must therefore look for new materials.

It has been demonstrated by a number of authors that doped semiconductors, including InAs, InSb, and Si, act as low-loss infrared optical metals for wavelengths from ~3-20μm [18–22]. The optical properties of these materials can be tuned by changing the doping density, enabling wavelength-specific design. Semiconductors can be grown using molecular beam epitaxy (MBE), resulting in crystalline films with precise control over layer thicknesses. MBE is a natural technique for growing superlattice-type HMMs. Indeed there have been two reports of HMMs created using silicon-doped InGaAs as the metal layer and undoped InAlAs as the dielectric layer [23,24]. Negative refraction was demonstrated in these materials, and the HMM onset wavelength was shown to be tunable by changing the silicon doping density.

In this paper, we report on the creation of tunable infrared HMMs created from single-material superlattices of metallic doped InAs and dielectric undoped InAs. Due to its small electron effective mass and high maximum doping density, InAs exhibits the shortest reported plasma wavelength (5.5μm) of any III/V material as demonstrated in [20]. This makes InAs particularly exciting for use in HMMs, as it will enable a broader working wavelength range than other III/V HMM material systems. Since the HMMs are created by layering doped/undoped InAs, growth is straightforward and much simpler than alternating materials, with no lattice-matching required beyond an initial buffer layer. In this paper, we will describe growth and measurement of InAs HMMs with a variety of doping densities and metal:dielectric thickness ratios, resulting in materials with designer optical properties across the infrared.

2. Experiment

2.1 Experiment

Samples were grown by molecular beam epitaxy (MBE) on semi-insulating GaAs substrates. The substrate temperature during growth was 450°C as determined by band edge thermometry, and the film is grown at 1.25~1.85 μm/hour. A fast growth rate is necessary to efficiently incorporate silicon donor atoms at the high doping levels used in these materials [21]. A 500nm undoped InAs buffer layer is initially grown on the substrate to account for the lattice mismatch between InAs and GaAs, thereby minimizing defect densities in the HMMs. This buffer is followed by the HMM superlattice comprising metal/dielectric Si:InAs/InAs layers. The thickness of the doped Si:InAs metallic layer, \( t_m \), and the thickness of the undoped InAs dielectric layer, \( t_d \), were varied, but the number of periods of Si:InAs/InAs is held constant at 10. The overall sample thicknesses varied with the thickness of the superlattice components and was confirmed post-growth by profilometry. A series of Si:InAs calibration films were grown and room-temperature Hall measurements conducted to determine the doping density of the metal layers in the HMM.

To obtain the optical properties of the samples, we collected polarized reflection and transmission data using an external port of a Bruker Vertex 70V Fourier transform infrared (FTIR) spectrometer, with a spectral resolution of 4cm\(^{-1}\) and a scan velocity of 40 kHz. A liquid-nitrogen-cooled HgCdTe detector, ZnSe lenses, a KRS-5 holographic wire grid polarizer, and a gold mirror are also used to create an optical setup allowing transmission and specular reflection measurements as a function of angle. Transmission data were taken from 0° to 50° in 5° increments, while reflection data were collected from 25° to 70° in 5° increments. The range of incident angles for light focused on the sample is ~2.1°. The reflection data were normalized to polarized reflection from the gold mirror set at 25° angle and the transmission data normalized to polarized transmission through the undoped GaAs substrate. A schematic of our optical setup is shown in Fig. 1.
In addition to standard optical measurements, we also performed experiments to demonstrate the negative refraction properties of our samples. A schematic is shown in Fig. 1. The setup was placed in transmission mode with the sample at 30 degrees. We first performed standard transmission measurements using the full beam for both polarizations. Then half of the transmitted beam was blocked using a blade placed about 1cm behind the sample. We conducted experiments with the blade blocking half the light in four different positions: left, right, top, and bottom (see inset of Fig. 1). As discussed in detail below, when the sample exhibits negative refraction, the beam is bent away (toward) from the blade for the blade placed in the left (right) position using TM-polarized light. This results in an increase (reduction) in signal reaching the detector. Data is normalized to transmission with no blade. When the blade is placed on the top or bottom, or for TE-polarized light, no change in signal is observed, indicating that our samples are indeed acting as HMMs.

2.2 Modeling

The angle- and polarization-dependent reflection and transmission properties were modeled using an anisotropic transfer matrix method [25] combined with effective medium theory (EMT) [15–17]. According to EMT, our multi-layered HMM sample can be modeled as one effective medium with differing permittivities in the parallel and perpendicular directions, given by

\[
\varepsilon_\parallel = \frac{\varepsilon_m + \eta \varepsilon_d}{1 + \eta} \quad \varepsilon_\perp = \frac{1 + \eta}{1/\varepsilon_m + \eta/\varepsilon_d}
\]  

(1)

where \(\varepsilon_m\) and \(\varepsilon_d\) are the permittivities of Si:InAs metal layer and InAs dielectric layer, respectively, and \(\eta\) is the ratio of their thicknesses (\(\eta = t_d / t_m\)). The permittivity of the Si:InAs layer is given by the Drude model [26,27]:

\[
\varepsilon_m = \varepsilon_0 \left(1 - \frac{\omega_p^2}{\omega^2 + i\omega\Gamma}\right)
\]

(2)
where \( \varepsilon_r \) is the high-frequency permittivity of the undoped InAs, \( \omega_p \) is the plasma frequency of the bulk Si:InAs, and \( \Gamma \) is the scattering rate. The plasma wavelength depends on the doping density (\( n \)) and effective mass (\( m^* \)) as

\[
\lambda_p = \frac{ne^2}{\varepsilon_r \varepsilon_m (n)}
\]  

(3)

By controlling the doping density, we can control the plasma frequency and, consequently, the permittivity of the metal layers in our HMM. The model used for these samples comprised four layers: air, HMM, undoped InAs buffer layer, and the GaAs substrate. The variable parameters were the HMM thickness, plasma frequency, and scattering rate. All samples were modeled using the reflection data collected at 40 degrees. Although calibration films of Si:InAs were grown to guide the HMM growths, the plasma frequency and scattering rate were allowed to vary slightly in the model. This is partially due to the sensitivity of the growth. As discussed above, a fast growth rate is required to incorporate silicon at these high doping densities. A slight deviation in the silicon or indium cell temperatures during growth will lead to slightly different doping densities than were obtained in calibration films. Additionally, the actual thickness of the metal and dielectric layers was allowed to vary slightly from the intended thicknesses. This may be due to carrier diffusion away from the intended regions of high doping into the undoped regions, which could also account for slight shifts in plasma frequency. Since we used a single material to create these HMMs, there is no conduction band offset to confine carriers. Instead, the heavy doping in the material leads to band bending which then confines the carriers. The amount of band bending was modeled using a self-consistent Schrödinger and Poisson solver. For heavy doping densities or relatively thick metal layers, the carriers were well-confined and the carrier density as a function of depth was essentially a square-wave function. However, for samples with lighter doping or with thin layers, the carriers were less well-confined. Indeed, in experiments with very thin layers (50nm metal and 50nm dielectric, not shown in this paper), a very poor match between experiment and theory was obtained. The Poisson solution for this sample showed a sinusoidal variation in the carrier concentration with depth, rather than a square wave. Although these samples still showed HMM-like behavior, we were unable to model them with the EMT shown above. This deviation from a perfect square-wave electron profile is most severe for samples 3-5 below, which had the lightest doping. Despite these difficulties, we continued to use the EMT model, as it is simple to implement and satisfactorily reproduces the relevant experimental features as discussed in detail below.

3. Results and discussion

A variety of samples were grown for this study. Two series of samples will be shown: one in which the metal:dielectric thickness ratio remained the same but the doping density in the metal layer was changed, and one in with the doping density remained the same, but the metal:dielectric ratio varied. Data for all samples is given in Table 1. The HMM thickness, metal thickness, dielectric thickness, fill factor, plasma wavelength, and scattering rate are all determined by fitting the angle-dependent transmission and reflection measurements as described above. The bandwidth of HMM behavior is determined from the sample permittivity and described in more detail below.
We will first consider samples in which the fill factor $\rho = t_w / (t_w + t_d)$ is held constant near 0.5 (Samples 1, 2, and 3), but the doping density and therefore the plasma wavelength is changed. TM-polarized reflection as a function of wavelength for incident angles from 25 to 70 degrees in five degree increments for three samples are shown in Fig. 2: Sample 1 with $\lambda_p = 5.8\,\mu m$, Sample 2 with $\lambda_p = 6.7\,\mu m$, and Sample 3 with $\lambda_p = 9.5\,\mu m$. All samples have similar scattering rates, as shown in Table 1. Modeled data using EMT is shown in Figs. 2(b), 2(d), and 2(e) for Samples 1, 2, and 3 for comparison.

It should be noted that, although we generally have good agreement between our experimental and modeled data, there are a few discrepancies. To begin, we sometimes see a double feature near the plasma wavelength in the experimental data which is not replicated in the model. This feature becomes stronger as the angle is increased. For an incident angle of 25 degrees, this “extra” feature is not visible, and the experimental results match the simulation quite well. As the angle increases, this extra feature becomes more prominent. In addition, the match between experiment and theory becomes worse overall at higher angles. Finally, we occasionally see long-wavelength behavior that is non-monotonic. For example, in Fig. 2(c), the reflectivity appears to “cross” itself around 11\,\mu m. We do not believe there is any physical significance to this feature.

There are multiple factors to consider when discussing the fit between experiment and theory. To begin, it is difficult to precisely position the sample and detector both when measuring our samples as well as when taking background data from our gold mirror. If the mirror or the sample is not perfectly aligned with the detector, the data may be spuriously high or low. In addition, small misalignments will be magnified as the angle is increased.
the simulation side, we are using effective medium theory, which is known to break down at higher angles, as well as when approaching the Brewster angle [28–31]. EMT will therefore be a worse approximation to our data as the angle increases, which could explain worse match between experimental and modeled data as the angle increases. Despite these difficulties, our simulated data reproduces the salient experimental features quite well.

At short wavelengths, oscillations in reflection are observed for all samples; this is due to Fabry-Perot interference across the HMM and buffer layer optical cavities. Likewise, reflection is ~30% for all samples at short wavelengths, as is expected for a semiconductor material. Finally, for all three samples, a feature in the reflection is observed near the plasma wavelength, indicated by arrows in Fig. 2. This occurs when \( \varepsilon_{\perp} = 0 \) and indicates the onset of HMM behavior. For sample 1, with the shortest plasma wavelength, the long wavelength reflection approaches 1 around 10\( \mu \)m, while samples with longer plasma wavelength have correspondingly shifted optical properties. As the angle increases, we can also observe a discontinuity in the Brewster angle, a result of \( \varepsilon_{\perp} \) approaching zero. For wavelengths longer than \( \lambda_p \), the Brewster angle is around 60 degrees (the exact angle depends on wavelength), while for shorter wavelengths, the Brewster angle is around 70 degrees. The Brewster angle can be somewhat difficult to determine due to the strong Fabry-Perot oscillations. This discontinuity in Brewster angle across the plasma wavelength is perhaps most obvious for Sample 2, but the same features occur in all samples.

Fig. 3. Data for Sample 1. Experimental (a) and simulated (b) TE reflection, experimental (c) and simulated (d) TM transmission, and experimental (e) and simulated (f) TE transmission. Other samples look similar. Both experimental and simulated TM and TE transmission data has been normalized to 1 for easier comparison of the relevant features.

For comparison, Fig. 3 shows TE-polarized reflection data and both TE- and TM-polarized transmission data for Sample 1; other samples are similar. In these plots, the simulated transmission and the experimental transmission are both normalized to 1, for ease of comparison. In the experimental data, transmission was somewhat lower than predicted,
due to absorption from defects at the InAs/GaAs interface. Figures 3(a) and 3(b) plot the TE-polarized reflection for Sample 1. Note the lack of feature near the plasma wavelength of 5.8 μm and the similarity in data as the angle is changed, in contrast to the data obtained for TM-polarized light in Figs. 1(a) and 1(b). Figures 3(c) and 3(d) plot the TM-polarized transmission. The strong absorption dip near the plasma wavelength is due to increased losses as the material approaches the wavelength at which $\varepsilon_\perp = 0$. At long wavelengths for both TE- and TM-polarized light, the transmission goes to zero as the reflection approaches 1.

![Fig. 4. Parallel (red) and perpendicular (gray) permittivity for samples with plasma wavelengths 5.8 μm (a), 6.7 μm (b), and 9.5 μm (c). Gray shaded region indicates HMM behavior.](image)

We can understand this optical response by investigating the wavelength-dependent parallel and perpendicular components of the permittivity. These are determined from Eq. (1), using the plasma wavelength and scattering rate from the reflection fitting and plotted in Fig. 4. The shaded gray region indicates type I HMM behavior, defined as $\varepsilon_\perp < 0$ and $\varepsilon_\parallel > 0$. The bandwidth of this region is given in Table 1 for all samples. The bandwidth in microns is determined by measuring the width of this region, while the bandwidth in percentage normalizes the width of the HMM behavior to the central wavelength. By analyzing the parallel and perpendicular components of the permittivity, as shown in Fig. 4, we can see that shifting the plasma wavelength merely shifts the material response to longer wavelengths while keeping the magnitude of the response quite similar; all plots have the same y-axes. For Sample 3, the bandwidth is 25%, while for Samples 1 and 2, it is around 30%. We can therefore tune the HMM response across the mid-infrared using a single-material system which can be grown by MBE.

We now turn to a series of samples in which the plasma wavelength is held constant near 9.5 μm, but the filling fraction is changed. Three samples will be discussed: Sample 3 with fill factor 0.44, Sample 4 with fill factor 0.24, and Sample 5 with fill factor 0.74. The TM reflection experimental and modeled data for these samples is shown in Fig. 5. It should be noted that, in some samples, the model predicts a resonance at longer wavelengths that we do not observe experimentally, most notably near 15 μm in Fig. 5(d). We have found that the strength of this resonance depends on the scattering rate: a smaller scattering rate leads to a strong resonance. Using larger scattering rates, however, led to a much worse fit overall. We therefore chose to keep the smaller scattering rates. The data was modeled using a single scattering rate, but it is also possible that a frequency-dependent scattering would be more accurate at long wavelengths.

Although the optical properties of the metal and dielectric layers are quite similar in all three samples, the observed reflection properties differ dramatically. We again see a feature in the reflection at the plasma wavelength in all samples as well as a discontinuity in the Brewster angle. However, at long wavelengths, we observed a marked difference in reflection. Sample 4 ($\rho = 0.24$) continues to show ~30% reflectivity out to 18 μm. Sample 3 ($\rho = 0.44$) exhibits reflection gradually approaching 1, while sample 5 ($\rho = 0.74$) exhibits a sharp increase in reflectivity after the plasma wavelength.
Fig. 5. Experimental TM reflection for Sample 4 (a), Sample 3 (c), and Sample 5 (e) for various angles, as indicated in the legend. For all samples, plasma wavelength is \(\sim 9.5\mu m\). Simulated TM reflection using EMT for Sample 4 (b), Sample 3 (d), and Sample 5 (f). Dotted lines are simulated reflection data that are not shown on the experimental plots due to the inaccessibly high angle.

Fig. 6. Parallel (red) and perpendicular (gray) permittivity for Sample 4 (a), Sample 3 (b), and Sample 5 (c). Note the different y-axis scales. Gray region indicates HMM behavior.

This behavior can be understood by considering the parallel and perpendicular components of the permittivity, plotted in Fig. 6. As shown in Fig. 6(a), Sample 4 has a narrow HMM region with a bandwidth of only 10\%, after which both components of the permittivity become positive, indicating that the material will act like a dielectric. This is mirrored by the reflection data, in which the sample exhibits a small amount of reflection even at wavelengths much longer than the plasma wavelength. Sample 3 exhibits the largest bandwidth in this set of samples at 25\%. At longer wavelengths, reflectivity increases slowly to 1 as the parallel component of the permittivity becomes negative while the perpendicular component becomes positive. Finally, Sample 5 again has a narrow HMM bandwidth at 14\%.
However, HMM behavior in this sample is not ended when $\varepsilon_\perp$ becomes positive as in Samples 1 and 2, but when $\varepsilon_\parallel$ becomes negative. This results in the material behaving strongly metallic, which a large and negative permittivity (note the difference in y-axis scales in Fig. 5) and a correspondingly sharp increase in reflectivity. To the best of our knowledge, this is the first experimental report detailing how layered hyperbolic metamaterial properties depend on filling fraction. These data show that in a single material HMM, the optical response can be tailored by fine adjustments in the HMM fill factor, which is easily accomplished using a technique like MBE.

Finally, to demonstrate the HMM behavior of our samples, we performed a standard beam optics experiment. When a material is acting like an HMM, it will exhibit negative refraction behavior. TM-polarized light will be bent in the opposite direction when crossing the HMM interface as would be expected in a material with a positive index of refraction. The difference between normal and negative refraction is shown schematically in Fig. 7(a). One commonly-used way to demonstrate negative refraction behavior is to block half the light transmitted through the sample using a razor blade [23,32–34]. The blocked transmission is then normalized to the total light transmitted through the unblocked sample. Depending on the type of refraction (positive or negative) and the blade position (left, right, top, bottom), different behavior is expected. A schematic of this setup including a drawing of the various blade positions is shown in Fig. 1. In our experiment, the incident angle of the light on the sample was held at 30 degrees. For a blade blocking the left half of the light transmitted through the sample, we expect to observe a peak in the transmission signal when negative refraction occurs. For a blade blocking the right half of the sample, we expect to see a dip. For TE-polarized light, we expect to see monotonic behavior.

Fig. 7. (a) Schematic of negative refraction measurement setup. When negative refraction occurs, the beam shifts to the right with respect to normal refraction. (b) Negative refraction data for Sample 1 shown for a blade blocking the left (black) or right (red) side of the transmitted beam for both TM-polarized light (solid) and TE-polarized light (dotted). (c) Negative refraction data for TM-polarized light with the blade in the left position for Sample 1 (black), Sample 2 (blue), and Sample 3 (green). Data for Samples 1 and 2 is cut off when reflection goes to 1, as it becomes very noisy. Arrows indicate plasma wavelength. In figure (c), Sample 1 data has been shifted vertically by 0.05, while Sample 3 data has been shifted vertically by $-0.05$ for visual clarity.

Negative refraction data for Sample 1 is shown in Fig. 7(b). For TM-polarized light with the blade on the left (black line), we see a peak in transmission at the plasma wavelength as we would expect for negative refraction. For TM-polarized light with the blade on the right, we see a dip at the corresponding wavelengths, again as expected for negative refraction. For TE-polarized light, we see no peak or dip, just a monotonically varying signal. We attribute the upward (downward) trend for the right (left) blade position to the overall change in refractive index as the wavelength changes. In Fig. 7(c), transmission data is shown for Samples 1, 2, and 3 for the blade on the left (curves have been shifted vertically for clarity). The peak in transmission shifts matches the position of the plasma wavelength, denoted by
colored arrows. These data strongly support the idea that our materials are acting like tunable infrared hyperbolic metamaterials.

4. Conclusion

In conclusion, we have demonstrated hyperbolic metamaterials fabricated from a single material system, using doped InAs for the metal layers and undoped InAs for the dielectric layers. This choice of material offers two advantages: it is relatively easy to grow by MBE and offers the shortest plasma wavelength of any III/V material. We have demonstrated the ability to control the onset of HMM behavior simply by adjusting the doping level in the metal layers, tuning across the mid-infrared. In addition, by changing the ratio of the metal:dielectric layer thicknesses, we can obtain designer wavelength-specific optical properties, including dielectric, metallic, and HMM behavior. Finally, we performed a geometric optics experiment to show that our materials exhibit negative refraction, a hallmark of the hyperbolic metamaterial. The fabrication of a layered HMM using a single-material semiconductor system has great potential for applications in hyperlensing and emission.

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