Light propagation in multilayer metamaterials

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Optical materials with a dielectric constant near zero have the unique property that light advances with almost no phase advance. Although such materials have been made artificially in the microwave and far-infrared spectral range, bulk threedimensional epsilon-near-zero (ENZ) engineered materials in the visible spectral range have been elusive. Here, we present an optical metamaterial composed of a carefully sculpted parallel array of subwavelength silver and silicon nitride nanolamellae that shows a vanishing effective permittivity, as demonstrated by interferometry. Good impedance matching and high optical transmission are demonstrated. The ENZ condition can be tuned over the entire visible spectral range by varying the geometry, and may enable novel micro/nano optical components, for example, transmission enhancement, wavefront shaping, controlled spontaneous emission and superradiance.

2.1 Introduction

Optical metamaterials have been the topic of intense study in recent years because they enable the realization of optical properties that do not occur in natural
Experimental realization of an ENZ metamaterial at visible wavelengths

Materials. Metamaterials are composed of elements with subwavelength scales in one or more dimensions. For this reason, metamaterials have been studied in the microwave spectral range, where index near-zero [31] and epsilon-near-zero (ENZ) behaviours have been predicted theoretically [32–34] and demonstrated experimentally [35–40]. With the appearance of advanced ion- and electron-beam nanolithography methods, the realization of ENZ metamaterials in the visible spectral range has now come within reach. Recently, a one-dimensional waveguide nanochannel with ENZ behaviour was demonstrated in the visible [41]. If three-dimensional ENZ metamaterials could be made in the visible, entirely new forms of waveform shaping [33, 42], light tunnelling [34] and spontaneous emission enhancement [43] would become possible over this technologically important spectral range.

Here, we present a metamaterial design composed of alternating layers of Ag and SiN with subwavelength layer thicknesses (Fig. 2.1, inset). This geometry has been studied previously to construct hyperbolic wavevector diagrams and increase the photonic density of states [23, 25, 44–46]. In the limit of deep subwavelength layer thicknesses, and for transverse electric (TE) polarized light (that is, the electric field parallel to the waveguides), the effective permittivity of this metamaterial equals the geometrically averaged permittivity [23],

\[ \varepsilon_{\text{av}} = \rho \varepsilon_m + (1 - \rho) \varepsilon_d, \]

where \( \rho \) is the metal filling fraction and \( \varepsilon_m \) and \( \varepsilon_d \) are the permittivity of metal and dielectric, respectively. Figure 2.1a shows experimentally determined values of the real (\( \varepsilon' \), solid lines) and imaginary (\( \varepsilon'' \), dashed lines) values of the permittivity for a thin Ag layer. The plot for \( \varepsilon' \) demonstrates the well-known decreasing trend towards strongly negative values due to the Drude-type response of free electrons in metal at long wavelengths. The effective permittivity of SiN is also shown in the figure and is positive and only slightly dispersive. The imaginary part of the permittivity of both Ag and SiN is measured to be very small over the entire visible range (Supplementary section "Ellipsometry").

Figure 2.1b shows the calculated effective permittivity for TE polarized light for a metamaterial composed of deep subwavelength Ag and SiN layers with \( \rho = 20, 30 \) and 50 \( \% \), based on the optical constants in Fig. 2.1a. With these metal filling fractions, the ENZ condition occurs at \( \lambda = 662, 545 \) and 428 nm, respectively, demonstrating the ENZ condition can be controlled over a broad spectral range by varying the composition. At the same time, the imaginary part of the effective permittivity \( \varepsilon'' \) is close to zero (\(< 0.2\)) over the entire visible range, indicating low loss.

The permeability of the metamaterial is assumed to be unity throughout the entire visible spectrum, as there is no magnetic resonance in the system. This means that when \( \varepsilon' = 1 \) the metamaterial is completely matched to free space, and almost no light would be reflected. Figure 2.1b shows that the wavelength at which this condition is met can be tuned throughout the visible range by varying \( \rho \). As an example, for \( \rho = 30\% \) we find \( \varepsilon' = 1 \) at \( \lambda = 491 \) nm. Interestingly, the losses are relatively low in this case (\( \varepsilon'' <0.1\)), even though a significant portion of the metamaterial consists of metal, and the optical frequency is quite close to the bulk
2.1 Introduction

![Figure 2.1: Metamaterial optical constants.](image)

- **Figure 2.1**: Metamaterial optical constants. 
  
  **a**, Measured real ($\epsilon'$, solid line) and imaginary ($\epsilon''$, dashed line) permittivity of Ag (grey) and SiN (blue). Inset: schematic of the multilayered metamaterial structure composed of Ag and SiN layers ($a$, $d$, and $m$ are unit cell dimension and dielectric and metal layer thicknesses, respectively). Light is incident along the $x$-direction and polarized along the $y$-direction. The dashed lines for Ag and SiN coincide. 
  
  **b**, Effective permittivity of an Ag/SiN multilayered metamaterial with metal filling fraction $\rho = 20\%$ (red), $30\%$ (green) and $50\%$ (blue), calculated using an effective medium approximation. The real ($\epsilon'$, solid lines) and imaginary ($\epsilon''$, dashed lines) parts of the permittivity are shown. The dashed lines for the three filling fractions coincide.

Probing the ENZ condition on this layered metamaterial requires samples that are optically accessible along the planar layers ($x$-direction), which cannot be achieved using geometries made by thin-film evaporation. Here, we demonstrate a layered metamaterial architecture composed of vertically oriented parallel lamellae of Ag and SiN, fabricated from a SiN membrane using a combination of focused ion beam (FIB) milling, reactive ion etching and thermal evaporation of Ag. This configuration allows us to isolate the ENZ condition using TE polarized light. Note that TE and transverse magnetic (TM) polarized light are degenerate when incident normal to the layers. Moreover, in the latter geometry, higher order Bloch harmonics can complicate the analysis. Figure 2.2a shows a top-view scanning electron micrograph (SEM) of a completed metamaterial sample (see Supplementary section "Fabrication", for more details). The $8 \mu m \times 8 \mu m$
waveguide arrays are highly regular, and the Ag and SiN layers are clearly visible as bright and dark bands, with layer thicknesses of 110 nm and 130 nm, respectively. A cross-section of the metamaterial structure is shown in Fig. 2.2b. The metamaterial layer can be clearly seen, on top of a remaining underlayer of SiN.

![Figure 2.2: Metamaterial sample.](image)

**Figure 2.2: Metamaterial sample.** a, SEM top view of the completed metamaterial structure. The Ag and SiN layers are visible as bright and dark bands, with layer thicknesses of 110 nm and 130 nm, respectively. b, SEM view of a cross-section showing the metamaterial (MM) layer, with alternating layers of Ag (bright) and SiN (dark), as well as a continuous SiN layer under the metamaterial. Pt layers are deposited to emphasize the top and bottom interface.

A total of 16 different multilayered metamaterial samples were fabricated. The exact waveguide dimensions, metamaterial thickness and underlayer thickness of
2.2 Interferometric phase measurements

Each sample was determined from cross-sections made using FIB. The metal layer thickness was varied from 40 to 150 nm, and the dielectric layer thickness between 36 and 135 nm. In this way, the metal filling fraction $\rho$ was systematically varied between 30 and 80% to study its effect on metamaterial dispersion. The unit cell size was varied between 85 and 280 nm to investigate the transition from an effective medium material to that of a waveguide-based metamaterial.

2.2 Interferometric phase measurements

To determine the effective optical properties of each multilayered metamaterial, we used a specially designed Mach-Zehnder interferometer to measure the optical path length of the metamaterial structures \( [9] \) (Supplementary section "Interferometry"). The optical path length was determined from the difference between the phase of light focused through the metamaterial and light focused through an air reference hole made in the same sample.

Figure 2.3a shows the measured phase shifts for metamaterials composed of a unit cell of a thin metal layer (40, 58 nm) in combination with a thin (45 nm, blue) and thick (135 nm, red) dielectric layer, respectively. Figure 2.3b shows similar data for a thick metal layer (155 nm, 150 nm) in combination with thin (36 nm, blue) and thick (130 nm, red) dielectric layers. The experimental data, taken at seven different wavelengths, show a gradual transition from a large phase shift at short wavelengths to a small phase shift at longer wavelengths.

Figure 2.3 (solid lines) also shows finite-difference time-domain simulations (FDTD Solutions 8.0, Lumerical Solutions) of the phase shift as a function of wavelength. In these simulations, the metamaterial waveguide dimensions determined from cross sections are used to define a periodic unit cell, and the experimentally determined optical constants of Ag and SiN (Fig. 2.1a) are used, so no fitting parameters are applied. Figure 2.3 shows very good agreement between experiment and simulations for all four samples. The gradual decrease in phase shift for longer wavelengths is clearly reproduced by the simulations. Also, the distinct feature in the data around $\lambda = 480$ nm for the largest unit cell in Fig. 2.3b is well reproduced. Its origin will be discussed in the following.

To determine the effective permittivity of the metamaterial from the measured phase shift, a transfer matrix method was applied to a double-layer structure composed of the metamaterial sample and the SiN underlayer, surrounded by air. This model expresses the effective complex transmission coefficient $t_{eff}$ of the combined metamaterial and underlayer structure in terms of their respective thickness and (effective) permittivity. The phase shift of light transmitted through the metamaterial relative to that through the reference hole $\Delta \phi = \phi_s - \phi_r$ directly follows using $\phi_s = \arg(t_{eff})$ and $\phi_r = k_0(d_s + d_{SiN})$, where $k_0$, $d_s$, and $d_{SiN}$ are the free-space wavevector, the thickness of the metamaterial sample and the thickness of the SiN underlayer, respectively. From the measured phase shift, the known thickness of metamaterial and SiN underlayer and the known permittivity of SiN, the effective
permittivity of the metamaterial can be derived. This procedure is illustrated by the inset in Fig. 2.4a, which shows the phase shift calculated as described above for a metamaterial sample \((d_s = 45 \text{ nm}; d_{SiN} = 93 \text{ nm}, \text{ in blue})\) at \(\lambda = 633 \text{ nm}\) as a function of metamaterial permittivity. The imaginary part of the effective permittivity is assumed to be \(\varepsilon'' = 0\). The validity of this assumption is discussed in the Supplementary section "Effective permittivity". Also shown in the inset is the measured phase shift for this geometry (horizontal red line). The effective permittivity then directly follows from the intersection between the two lines. The phase shift was measured 20 times for each metamaterial sample at each wavelength, and every measured phase shift was separately converted to an effective permittivity. The mean of this distribution was taken to be the effective permittivity of the sample, with the standard deviation of the distribution an indicator for the error. For metamaterial thicknesses greater than 15 nm, using simulations, we verified that the effective permittivity is independent of thickness (Supplementary section "Thickness dependence").
2.3 Changing effective parameters

Figure 2.4a shows the effective permittivity derived in this way for three different metamaterials, with each structure having the same unit cell size of 200 nm, and the metal filling fraction varied ($\rho = 30, 60$ and $80\%$). For all filling fractions a clear trend of decreasing permittivity with wavelength is observed. The measured permittivity ranges from $\epsilon' = 6.0 \pm 1.2$ at $\lambda = 364$ nm for the lowest filling fraction to $\epsilon' = -13 \pm 3.4$ at $\lambda = 633$ nm for the highest filling fraction. Figure 2.4a also shows analytical calculations of the metamaterial effective permittivity (solid lines). This effective permittivity is determined by calculating the supported eigenmodes of a planar waveguide array using a transfer matrix formalism for a periodic unit cell \cite{21, 29}. The eigenmodes are calculated assuming light propagation parallel to the waveguides, with the electric field parallel to the metal-dielectric interfaces (TE polarization). Only the fundamental mode (characterized by the lowest propagation losses) is assumed to contribute to wave propagation through the metamaterial. The calculations are in good agreement with the experimental trends. The strong difference in dispersion between the three geometries is also well represented by the calculations, confirming that the wavelength at which the effective permittivity of the metamaterial becomes zero can be controlled by the metal filling fraction. This cutoff condition occurs at $\lambda = 807, 567$ and $394$ nm for the three geometries, respectively.

Figure 2.4b shows the measured and calculated effective permittivity of three structures with an approximately constant metal filling fraction $\rho = 50\%$. Here, the unit cell size is varied ($a = 85, 170$ and $280$ nm). The experimental trends are well reproduced by the calculations for the $85$ and $170$ nm layer thicknesses. For the $280$ nm layer thickness, differences are observed between calculations and experiment that are attributed to the excitation of higher-order waveguide modes that are not included in the calculation. This aspect will be discussed shortly. The black dashed curve in Fig. 2.4b shows the geometrically averaged permittivity $\epsilon_{av}$, calculated based on the effective medium theory assuming $\rho = 50\%$. Clearly, the experimental data and the results of the analytical calculation are strongly redshifted compared to this simple model. Indeed, in our metamaterial geometry, with metal thicknesses larger than the skin depth, the electric field is partially excluded from the metal region, causing the effective medium approximation to be invalid. Because of this exclusion, the metal filling fraction is effectively lowered, resulting in a redshift. The data in Fig. 2.4b show how the ENZ condition can be controlled through the 400-800 nm spectral range for any given unit cell size by an appropriately chosen metal filling fraction.

To further study the metamaterial dispersion, we measured the transmission spectra for each metamaterial structure. Data were taken using a white-light source and a spectrograph equipped with a charge-coupled device (CCD) detector, and the spectra were normalized to the transmission through a reference hole. Figure 2.5a-d shows the transmission spectra for the four structures represented in Fig. 2.3. In all cases, the transmission is highly wavelength dependent; the transmission peaks
Figure 2.4: Metamaterial effective permittivity. **a**, Measured (data points) and calculated (solid lines) effective permittivity for 200 nm unit cell structures with metal filling fraction $\rho = 30\%$ (red), 60\% (green) and 80\% (blue). Inset: calculated phase shift (solid lines) as a function of effective metamaterial permittivity for a metamaterial sample ($d_s = 45 \text{ nm}; d_{\text{SiN}} = 93 \text{ nm}, \text{in blue}$) at $\lambda = 633 \text{ nm}$. The measured phase shift is indicated by the horizontal red line. **b**, Measured and calculated effective permittivity for structures with a metal filling fraction of $\rho = 50\%$ and different unit cell size $a = 280 \text{ nm}$ (red), 170 nm (green) and 85 nm (blue). The effective permittivity based on the geometric average is also shown as the black dashed line. Error bars correspond to the standard deviation of the measured phase shifts converted to effective permittivity.
2.3 Changing effective parameters

at a value as high as 90% for the 58 nm Ag/135 nm SiN metamaterial. Figure 2.5 also shows the simulated transmission (solid green lines) for the measured waveguide dimensions, taking into account the SiN underlayer. Overall, the experimental peak wavelengths are well reproduced by the simulation. The lower transmission for the 155 nm Ag/36 nm SiN metamaterial is also reproduced in the calculations. We attribute the difference in spectral shape between measurement and simulation to variations in waveguide dimensions across the sample. The reduced signal-to-noise ratio observed for wavelengths below 400 nm is caused by the low sensitivity of the CCD detector in this regime.

![Transmission spectra and matching condition](image)

**Figure 2.5: Transmission spectra and matching condition.** Measured (black) and simulated (solid green) transmission spectra for four different metamaterial structures. The simulated transmission without the SiN underlayer (dashed green line) peaks at the same wavelength where the effective permittivity of the structure is around 1 (vertical grey dashed line), corresponding to air. The wavelength at which the effective permittivity is near zero is indicated by the vertical black dashed line.

To explain the origin of the transmission maximum, we also simulated the transmission of the same metamaterial geometries, but now without the SiN underlayer (dashed green lines). Also indicated in Fig. 2.5 are the wavelengths
2 Experimental realization of an ENZ metamaterial at visible wavelengths

at which the calculated effective permittivity of the metamaterial equals 1 (vertical grey dashed lines). In all cases, the wavelength at which $\epsilon' = 1$ agrees well with the wavelength of the peak in transmission (when there is no SiN underlayer), indicating that the transmission maximum results from a matched effective permittivity of the metamaterial to the surrounding air. The measured transmission peaks occur at slightly blueshifted wavelengths because, with the SiN underlayer present, the metamaterial sample effectively acts as an antireflection coating for the SiN underlayer. Indeed, such optimum impedance matching occurs for a metamaterial permittivity between that of air and SiN, which is in the blueshifted spectral range.

The vertical black dashed lines in Fig. 2.5 indicate the wavelengths at which the metamaterials reach the ENZ condition. The calculated transmission for the case without the SiN underlayer is still very high (for example, $T = 87\%$ in Fig. 2.5b), although somewhat reduced compared to the transmission at the impedance-matched $\epsilon' = 1$ condition wavelength. This is caused by an increased impedance mismatch and a reduced propagation length (Supplementary section "Propagation length"), leading to both increased reflection and increased absorption.

2.4 Size and angular dependence

An important criterion for metamaterials is that the constituent elements are subwavelength. Therefore, it is interesting to study how the effective optical properties change with unit cell size. As Fig. 2.4b demonstrates, a distinct redshift of the dispersion is observed for larger unit cell sizes with the same filling fraction. To further quantify this effect we calculated the supported waveguide eigenmode for different unit cell sizes. Figure 2.6a shows the calculated effective mode index using the same transfer matrix method described above, for a periodic unit cell with a constant metal filling fraction of 30%, and a unit cell size ranging from $a = 1$ nm to 600 nm at a free-space wavelength of $\lambda = 515$ nm. As can be seen, the real part of the mode index of the fundamental mode (solid black line) clearly converges to the expected index based on the effective-medium theory (dashed green line) for small unit cell dimensions ($n'_{\text{eff}} = 0.75$ for this geometry). For large unit cell dimensions, the mode index approaches the SiN index (dashed blue line), as most of the light is excluded from the thick metal layers. The same behaviour is observed for the imaginary part of the index, as can be seen in Fig. 2.6b.

Around a unit cell size of a 250 nm, the first higher-order waveguide mode is no longer cut off at $\lambda = 515$ nm. The effective index for this mode is also shown in Fig. 2.6a as the solid grey line. Figure 2.6b shows that, for a unit cell larger than 300 nm, the imaginary part of this mode becomes comparable to that of the fundamental waveguide mode. The presence of higher-order modes limits the maximum unit cell dimensions allowed in the design of this metamaterial. Interestingly, the first higher-order mode was in fact observed in the measured and simulated phase shift for the largest fabricated unit cell ($a = 280$ nm), as can be seen in Fig. 2.3b. The
Figure 2.6: Isotropic index. a,b, Real (a) and imaginary (b) part of the mode index of the fundamental TE waveguide mode (black) and the first higher-order mode (grey) calculated versus unit cell size. For small unit cell sizes the effective index approaches the geometrically averaged index (green dashed lines), and for large unit cell sizes the mode index approaches the refractive index of SiN (blue dashed lines). c, Wavevector diagram for $\lambda = 460$ nm ($a = 85$ nm, $\rho = 47\%$), demonstrating an effective index that is independent of angle. The green dashed curve corresponds to the expected isotropic index, and the black dashed curve is the wavevector in free space. d, Wavevector diagram calculated for $\lambda = 475$ nm. The mode index is reduced and still angle independent.
distinct oscillatory feature observed around 480 nm is due to a transition between the fundamental and the first higher-order mode.

So far, we have considered normally incident light. Next, we address the response of our three-dimensional ENZ metamaterial in the angular range between parallel incidence (along $x$) and perpendicular incidence (along $z$). Figure 2.6c shows the calculated wavevector diagram for light in the $x-z$ plane (free-space wavelength $\lambda = 460$ nm), for $a = 85$ nm and $\rho = 47\%$. To determine the response of the metamaterial for all incident angles, we performed two different calculations [21]. The first calculation determines the fundamental waveguide mode for a given small ($k_z < k_0$), real-valued $k_z$ component (shown in blue). This calculation determines the wave propagation for light mainly along $x$. The second calculation determines the fundamental Bloch wavevector, assuming a small real-valued wavevector component along $x$ ($k_x < k_0$) (shown in red). This situation represents wave propagation mainly along $z$. Also shown is the wavevector diagram of light propagation in air (black dashed line), and light propagation with an isotropic index equal to the waveguide mode index when $k_z = 0$ (green dashed line). The fact that the calculated wavevector diagram follows the isotropic index in their respective regions of interest indicates that for a unit cell of $a = 85$ nm the effective index of the structure is nearly angle independent. A small unit cell is required to obtain an angle independent refractive index, as light propagation normal to the waveguide interfaces (along $z$) is described by a Bloch wave composed of a series of harmonics, separated by the reciprocal lattice constant $\Lambda = 2\pi/a$ (ref. [29]): $\Lambda$ must then remain much larger than wavevector $k_z$.

Figure 2.6d shows a wavevector diagram at a free-space wavelength of $\lambda = 475$ nm. Here, the mode index is strongly reduced because the metamaterial approaches cutoff, yet the response is still angle independent. When the mode index further approaches zero, the wavevector diagram will no longer be spherical, and the calculated value of $k_x$ ($k_z$) will become independent of the $k_z$ ($k_x$) component.

## 2.5 Conclusions

In conclusion, we have designed, fabricated and characterized an optical multilayered metamaterial consisting of thin Ag and SiN layers with an effective permittivity in the visible spectrum ranging from $\epsilon' = 6.0 \pm 1.2$ to $\epsilon' = -13 \pm 3.4$. The metamaterial is highly dispersive through the $\lambda = 400 - 800$ nm spectral range, and the dispersion is determined by the layer thicknesses and relative filling fraction. The epsilon-near-zero condition is experimentally demonstrated at wavelengths in the range $\lambda = 351 - 633$ nm. The optical transmission of the metamaterial is as high as 90% percent, and impedance matching to air is observed when $\epsilon' = 1$. The experimental data correspond well with analytical calculations and simulations. The transition from an effective medium material for small unit cell size to that of a coupled waveguide material for larger dimensions is studied. For the unit
cell size \( a = 85 \text{ nm} \), calculations show that the effective index is independent of angle. This highly dispersive metamaterial may find applications in transmission enhancement, wavefront shaping, control of spontaneous emission and superradiance.

2.6 Methods

Metamaterial samples were fabricated from a SiN membrane. The waveguide pattern was written in a Cr masking layer (20 nm) using FIB milling at normal incidence. The pattern was transferred to the SiN using an anisotropic CHF\(_3\)/O\(_2\) reactive ion etch recipe. The etched channels were in-filled with Ag using physical vapour deposition. The surface of the metamaterial was then polished using FIB milling at oblique incidence.

The optical path length of the metamaterial sample was measured in a Mach-Zehnder interferometer. Light was focused through the metamaterial, and the length of the reference arm was changed with a piezo-electrically driven mirror, causing a sinusoidally oscillating intensity at the output. Light was then focused through a reference hole in the membrane, and the same reference mirror movement was performed. The sinusoidal signal was now displaced by a phase shift, caused by the optical path length of the metamaterial sample.

The simulated phase shift was obtained from FDTD simulations (Lumerical). We recorded the phase and amplitude of the transmitted plane wave after propagation through the structure. First the structure consisted only of a single SiN layer. The thickness of this SiN layer was increased from zero to the realized thickness, in steps of 1 nm. The thickness of the metamaterial on top of the SiN layer was then increased to the fabricated thickness.

2.7 Supplementary information

2.7.1 Phase advance

To illustrate how light propagates through an ideal ENZ material we have calculated the normalized field profile of a plane wave incident on an ENZ slab 400 nm thick with a permittivity \( \varepsilon = 10^{-4} \) and \( \mu = 1 \) for a free-space wavelength \( \lambda_0 = 500 \text{ nm} \). Figure 2.7(a) shows the real part of the \( H_z \) and \( E_y \) field profile. The magnetic field is constant inside the slab, and the electric field has a linear dependence on the position in the slab, as expected from Ref. [34] of the main text. Figure 2.7(b) shows the extracted phase shift and absolute amplitude of light transmitted through the slab as a function of slab thickness \( L \). The phase converges to 90 degrees while the amplitude vanishes. This initial change in phase is caused by a changing field at the first interface of the slab due to an increased reflected wave. Eventually this phase change becomes thickness independent.
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Figure 2.7: Phase advance. a, Calculated field profile of a plane wave propagating through an ENZ slab. The slab has a thickness of 400 nm, and assumed permittivity and permeability $\epsilon = 10^{-4}$ and $\mu = 1$, the free-space wavelength for the calculation is taken to be 500 nm. b, Calculated phase shift and absolute amplitude of the plane wave transmitted through the slab as a function of slab thickness $L$.

2.7.2 Ellipsometry

The optical constants of thin Ag and SiN layers given in Fig. 2.1(a) were determined using variable-angle spectroscopic ellipsometry (VASE, J.A. Woollam Co., Inc.). For these measurements Ag layers of different thicknesses (30 - 60 nm) were deposited on a silicon wafer using thermal evaporation. A point by point fit was used to obtain the optical constants of the evaporated metal layers. The optical constants of the SiN layer were determined using VASE by measuring on a part of the silicon window frame, which was covered by the same SiN membrane as was freestanding in the middle of the window. A Cauchy model was used to fit the refractive index of the SiN dielectric layer to the measured data, and good agreement was observed.

2.7.3 Fabrication

We fabricated the multilayered metamaterial starting with a commercially available 200-nm-thick low-stress SiN membrane grown with low-pressure chemical vapor deposition (Norcada Inc.). The membrane is covered by a 20 nm Cr masking layer using thermal evaporation. An array of parallel linear slots is written in the masking layer using a 1.5 pA, 30 keV Ga$^+$ ion beam focused to a diameter of 10 nm, as sketched in Fig. 2.8(a). Subsequently the pattern is transferred into the SiN layer by anisotropic reactive ion etching using a CHF$_3$/O$_2$ mixture. As shown in Fig. 2.8(b-c), after the etching process there still is a thin layer of SiN left underneath the etched waveguide stripes. This underlayer provides mechanical stability for the otherwise freestanding SiN stripes.
Figure 2.8: Metamaterial fabrication. a, Patterning of the Cr masking layer using FIB. b, Anisotropic etching of the SiN membrane with an anisotropic reactive ion etching recipe. c, Thermal deposition of Ag over the entire sample surface. d, FIB polishing of the metamaterial surface to remove excess metal.

The channels etched in the SiN layer are then infilled with Ag using thermal evaporation at a pressure of $10^{-6}$ mbar. By carefully aligning the sample in a direct line of sight to the source the channels were completely filled with metal. As a result of the deposition process, the entire membrane surface is covered with an optically thick metal layer. To allow access to the metamaterial, the metal layer on top of the metamaterial is polished with a focused ion beam at an oblique angle of incidence ($\theta \approx 3^\circ$), see Fig 2.8(d). This polishing is continued until the multilayered metamaterial structure is revealed. Square reference holes are placed at a distance of 5 $\mu$m away from the exposed metamaterial structures by FIB milling through the entire Ag/SiN layer stack at normal incidence. After the optical measurements SEM cross sections of the metamaterial samples were made using focused ion beam milling. To enhance contrast, the metamaterial was first covered with Pt using electron-beam induced deposition at the top and bottom side of the membrane.
2.7.4 Interferometry

Figure 2.9(a) shows a sketch of the interferometer setup. Light from either an Ar+ or HeNe laser is split into a reference and a sample path using a 50/50 beam splitter. Microscopes objectives (numerical aperture NA = 0.45) are used to focus and collect light from the metamaterial. The spot size was around 5 \( \mu \)m, illuminating 20 to 60 unit cells, depending on the exact geometry of the metamaterial. The transmitted beam is interfered with the reference beam using a second 50/50 beam splitter. The intensity of the interfering beams is measured on a photodiode placed behind this beam splitter. A piezo-electrically driven mirror in the reference arm is continuously displaced using a saw tooth driving function, leading to a constant advance of the phase of the reference beam with time. Typical interference oscillations versus time are shown in Fig. 2.9(b) when light is focused through the metamaterial sample (red) and for light transmitted through a reference hole through the membrane (blue). A second piezo-electric stage is used to switch between measurements through the metamaterial area and through the reference hole. A sine wave is fitted to both interference traces, and the relative phase shift \( \Delta \phi = \phi_s - \phi_r \), where \( \phi_s \) and \( \phi_r \) are the phases for sample and reference trace respectively. This phase shift is a direct measure for the optical path length of the metamaterial. The entire measurement cycle alternating between the metamaterial area and the reference hole is synchronized and computer controlled, and is repeated multiple times to allow for an accurate determination of the phase shift.

Figure 2.9(c) shows a histogram of phase shifts determined from 400 measurements on a metamaterial structure at a free-space wavelength \( \lambda = 633 \) nm (in red). The average phase shift of these measurements is \( 37\pm10^\circ \). The error is caused by the reproducibility of the piezo mirror position, which varies by approximately 25 nm. Figure 2.9(c) also shows a histogram of phase shifts measured on the same metamaterial structure at \( \lambda = 364 \) nm (purple). To determine the full dispersion of the metamaterial structure the phase shift is measured using six different lines of an Ar+ laser (in the range \( \lambda = 351-515 \) nm) and a HeNe laser (\( \lambda = 633 \) nm). The coherence length of these light sources is sufficiently long to allow us to observe interference in the setup where the sample and reference beam paths of the interferometer are of different length. All phase shifts reported in the main text are measured relative to an air reference hole.

2.7.5 Phase shifts and effective permittivity

The inset of Fig. 2.4(a) shows the dependence between the effective permittivity of a metamaterial sample and the measured phase shift. In this model, it is assumed that the imaginary part of the permittivity is zero, such that only the real part of the permittivity contributes to the measured phase shift. We justify this assumption by the observation that the imaginary part of the geometrically averaged permittivity \( \varepsilon'' < 0.2 \) in the \( \lambda = 400 - 650 \) nm spectral range (Fig. 2.1(b)). To study the effect of a non-negligible imaginary part, we plot the same graph as the inset of Fig. 2.4(a)
Figure 2.9: Interferometry. a, Sketch of the Mach-Zehnder interferometer setup used to measure the optical path length of the metamaterial samples. b, Interference signal on photodiode as the mirror position is varied over time. The fitted sinusoidal function through the metamaterial data (red) has a different phase than the fitted sinusoidal function of the reference measurement (blue). c, Histogram of 400 measurements demonstrating a distinct difference in average phase shift between measurements at $\lambda = 633$ nm (red) and $\lambda = 364$ nm (purple).
for four different $\varepsilon''$: 0, 0.1, 0.5 and 1. The effect of adding this imaginary part can be seen in Fig. 2.10(a). The relation between the calculated phase shift and the effective metamaterial permittivity is only slightly shifted by adding an imaginary part to the permittivity. This shift is most pronounced in the regime where $\varepsilon' \ll 0$. However, this paper mainly focuses on the regime where $\varepsilon' \approx 0$. To demonstrate the effect of a nonzero imaginary part in this ENZ regime, we have calculated the expected phase shift in this regime, with an imaginary part $0 < \varepsilon'' < 1$. Figure 2.10(b) shows the result of this calculation. The calculated phase shift only changes by a few degrees in this range, well within the experimental error of our setup.

**Figure 2.10: Phase shift and permittivity.** a, Effect of a nonzero imaginary part of the permittivity. The relation between the calculated phase shift and effective metamaterial permittivity is slightly shifted for higher imaginary parts. The effect is most pronounced in the regime where the real part is negative. b, The calculated effect on phase shift at the ENZ condition ($\varepsilon' \approx 0$), which shows only a change of a few degrees.

### 2.7.6 Thickness dependence

The effective optical properties of a metamaterial should be independent of thickness. To determine the effective permittivity as a function of thickness, we use a parameter retrieval method, where we utilize the simulated complex reflection and transmission coefficients in order to extract the material parameters [47]. The extracted permittivity and permeability are calculated as a function of metamaterial thickness and free-space wavelength, to ensure the correct branches of the solution are used.

Figure 2.11(a) shows the extracted real part of the permittivity as a function of metamaterial thickness. This permittivity is calculated for a unit cell with dimensions $m=40$ nm and $d=45$ nm, at three different wavelengths. As can be seen for $\lambda = \ldots$
475 nm and $\lambda = 650$ nm, the permittivity becomes almost thickness-independent for metamaterials thicker than 15 nm. Note, the experiments were preformed for thicknesses well beyond this thickness. This indicates that the effective measured permittivity is indeed determined by the waveguide dispersion. The oscillation in the calculation for $\lambda = 350$ nm is attributed to interference inside the layer that results from the relatively high effective permittivity at this wavelength ($\epsilon' = 3.0$).

![Graph showing the dependence of effective permittivity and propagation length on metamaterial thickness and wavelength.](image)

**Figure 2.11: Thickness dependence and propagation length.** a, Calculated real part of the effective permittivity as a function of thickness. b, Propagation length as a function of wavelength, calculated for a unit cell with $m = 58$ nm and $d = 135$ nm. The vertical gray line indicates the wavelength at which the free-standing metamaterial is impedance matched to free-space, and the vertical black line indicates the ENZ condition.

### 2.7.7 Propagation length

In the main text we demonstrated high transmission through the fabricated metamaterial samples. The observed maximum in transmission was attributed to an optimized impedance leading to low reflection. There is however also absorption in the metamaterial, which attenuates the electromagnetic wave as it propagates through the sample. To determine this attenuation, we calculate the propagation length, defined as $L_p = 1/(2 * \text{Im}(k_{wg}))$, where $k_{wg}$ is the wavevector of the waveguide mode in a periodic system. Figure 2.11(b) shows the calculated propagation length as a function of wavelength. The vertical gray line indicates the wavelength at which this metamaterial is impedance matched to air, and the vertical black line indicates the wavelength at which the effective permittivity becomes zero.

As is evident from this figure, the propagation length is strongly reduced as the ENZ condition is approached. As the ENZ point is crossed, the material becomes increasingly metallic, explaining the reduced propagation length. However, very close to this ENZ condition, the propagation length is approximately 500 nm, well beyond the thickness of the film, in agreement with the relatively high transmission observed from the metamaterial.