Supporting Online Material for

Negative Refraction at Visible Frequencies

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1. Negative index and negative refraction

Consider an electromagnetic mode propagating with wavevector $k$ in an isotropic medium. The phase and group velocity of this mode are collinear and given by, respectively, $v_p = (\omega / k) \hat{k}$ and $v_g = (d\omega / dk) \hat{k}$, where $k = |k|$, and $\hat{k} = |k| / k$. The phase velocity characterizes the speed and direction of propagation of the phase fronts, whereas the group velocity determines the direction of power flow $S = E \times H = W v_g$, where $S$ is the Poynting vector and $W$ is the energy density of the electromagnetic field.

When $d\omega / dk > 0$, $v_p$ and $v_g$ are parallel and the standard forward-wave condition $v_p \cdot v_g > 0$ is obtained. $k$ and $S$ are parallel under such a condition and power propagates in the same direction as the phase-fronts. When $d\omega / dk < 0$, $v_p$ and $v_g$ are anti-parallel and the anomalous backward-wave condition $v_p \cdot v_g < 0$ is obtained. $k$ and $S$ are anti-parallel and power propagates in a direction opposite to that of the phase-fronts.

When a plane wave traveling in medium 1, characterized by phase and group velocities $v_p^1$ and $v_g^1$, is incident at an angle $\phi_1$ upon a boundary with medium 2, characterized by phase and group velocities $v_p^2$ and $v_g^2$, the resulting refraction angle of the transmitted beam, $\phi_2$, is dictated by conservation of the parallel component of $k$ across the boundary as well as conservation of energy (see ref. S1):

$$\frac{\sin(\phi_2)}{\sin(\phi_1)} = \frac{\text{sign}(v_p^1 \cdot v_g^1) c / |v_p^1|}{\text{sign}(v_p^2 \cdot v_g^2) c / |v_p^2|} \equiv n_1 / n_2. \quad (S1)$$

By analogy with the classical form of the Snell-Descartes law of refraction $\sin(\phi_2) / \sin(\phi_1) = n_1 / n_2$, an effective refractive index for each medium can be identified based on Eq. (S1):

$$n_{1,2} = \text{sign}(v_p^{1,2} \cdot v_g^{1,2}) c / |v_p^{1,2}|. \quad (S2)$$

If $v_p^1 \cdot v_g^1 < 0$, medium 1 acts as if it had a negative index of refraction. When light exits such a medium into a second medium characterized by a positive index of refraction, it will be refracted, according to Eq. (S1), to the same side of the normal, i.e. to a negative angle.
2. Derivation of mode index of thick Ag/Si$_3$N$_4$/Ag waveguide

To derive the actual value of the mode index of the thin Ag/Si$_3$N$_4$/Au waveguide, $n_1$, the mode index in the surrounding Ag/Si$_3$N$_4$/Ag waveguide, $n_2$, must be known. To circumvent the difficulty of estimating $n_2$ theoretically, given the predicted multimodal dispersion properties (Fig. 1A), we measure $n_2$ directly using an interferometric technique. To this end, Ag/Si$_3$N$_4$/Ag waveguides with 500-nm-thick dielectric cores were fabricated with semi-transparent Ag cladding layers of 150nm on each side; no additional Al cladding layer was added. The separation between input and output slit was varied from $d = 1$ µm to 3 µm in 25-nm increments. Each device was illuminated at normal incidence over its entire area and the intensity emitted from the output slit was monitored (Fig. S1 A). Due to interference at the output-slit, between light transmitted along the waveguide and light transmitted directly through the top Ag cladding layer, a periodic modulation in the emitted intensity is obtained as a function of $d$ (Fig. S1 B). At a given wavelength $\lambda_0$, the effective mode index of the waveguide is given by $n_2 = 2\lambda_0 / P$, where $P$ is the period of the modulation.

3. Effect of thickness and edge angle of Au/Si$_3$N$_4$/Ag prism

We characterize in detail the refractive properties of a Ag/Si$_3$N$_4$/Au waveguide within its frequency interval of negative refractive index. The result of refraction from a single prism of Ag/Si$_3$N$_4$/Au waveguide (W1), of dielectric-core thickness $t_1 = 75$ nm, into a thick Ag/Si$_3$N$_4$/Ag waveguide (W2), of dielectric core thickness $t_2 = 500$ nm, is shown in Fig. S2 A, at two closely-spaced wavelengths of $\lambda_0 = 501$ nm and $\lambda_0 = 488$ nm, respectively. Given an incident angle of $\varphi_1 = 7^\circ$ at the W1/W2 interface, substantially different refraction angles $\varphi_2 = -31.2^\circ$ and $-27.1^\circ$ are obtained in each case. Applying Snell’s law (Eq. S1) using these angles of refraction and measured values $n_1 = 0.65$ and 0.57, respectively (Fig. S1 B), we obtain effective refractive indices $n_2 = -2.72$ and $-2.09$, respectively. Weak transmission of a positively-refracted spot can also be observed, hinting that 75-nm-thick constrictions can support a strongly attenuated positive-index photonic mode in addition to the negative-index SPP mode. As the waveguide thickness is increased to $t_1 = 150$ nm, the positively refracted spot increases in intensity: photonic mode transmission competes with the SPP mode (Fig. S2 B). While the negatively-refracted spot size is slightly reduced with respect to the input slit length, the positively-refracted spot is enlarged. Such variations in spot size likely result from focusing or diffraction effects dependant on both the sign and the magnitude of $n_1$.

To confirm the applicability of Snell’s law to the bimetallic waveguide structures, we vary the angle of incidence $\varphi_1$ by fabricating W1 prisms ($t_1 = 150$ nm.) with edge angles of $\theta = 5^\circ$ and $7^\circ$ respectively. Refraction results at $\lambda_0 = 501$ nm are shown in Fig. S2 B. A distinct negatively-refracted spot is obtained in both cases, implying a
Fig. S1. Experimental determination of refractive index $n_2$ of thick Ag/Si$_3$N$_4$/Ag waveguide. (A) Cross-sectional diagram of interferometer used to determine $n_2$. The structure is a modified version of the device of Fig 2B, in which the optically opaque metal cladding on both sides of the dielectric core is replaced with a 150-nm-thick layer of Ag. This semi-transparent cladding allows for interference, at the output-slit position, between the transmitted waveguide mode and the partially transmitted incident illumination. Periodic modulation of the light emerging from the output slit as a function of slit-slit distance $d$ yields the effective mode index of the waveguide ($S2$). (B) Measured output-slit intensity as a function of $d$ for wavelengths within the predicted negative index-region. Distance $d$ is varied from 1 to 3 $\mu$m in 25-nm increments. The waveguide index is given by $n_2 = 2\lambda_0 / P$, where $P$ is the period of modulation. For free-space wavelengths $\lambda_0 = 476$, 488, 496, 501, and 514 nm, the resulting indices in the Ag/Si$_3$N$_4$/Ag waveguide are $n_2 = 0.40$, 0.57, 0.50, 0.65, and 0.82, respectively.
**Fig. S2.** Detailed exploration of refractive properties of Au/Si3N4/Ag waveguide in the negative index region. (A) Output-spot position for the single prism geometry of Fig. 2C as a function of wavelength. The core thickness and interface angle of the prism are held constant at \( t = 75 \text{ nm} \) and \( \theta = \phi_i = 7^\circ \), respectively. The angle of refraction \( \varphi_2 \) varies from \(-31.2^\circ\) to \(-27.1^\circ\) as \( \lambda_0 \) is decreased from 501 to 488 nm. (B) Output-spot position for prism angles of \( \theta = \phi_i = 7^\circ \) and \( 5^\circ \). Here, the core thickness and excitation wavelength are held constant at \( t = 150 \text{ nm} \) and \( \lambda_0 = 501 \text{ nm} \), respectively. The angle of the negatively-refracted spot shifts from \(-28.8^\circ\) (for \( \phi_i = 7^\circ \)) to \(-20.0^\circ\) (for \( \phi_i = 5^\circ \)), corresponding to a constant prism index \( n_i = -2.5 \) and confirming the applicability of Snell’s Law to such waveguides operating in the negative index region. Note that in both A and B, bands of positive and negative refraction be observed, corresponding to transmission of a positive-index photonic mode and a negative-index plasmonic mode, respectively.
refraction angle of $\varphi_2 = -20.0^\circ$ when $\varphi_1 = 5^\circ$ and of $\varphi_2 = -28.8^\circ$ when $\varphi_1 = 7^\circ$. The corresponding ratio $\sin(\varphi_2)/\sin(\varphi_1) = n_1/n_2$ is $-3.82$ and $-3.88$, respectively. The constancy of this ratio as a function of edge angle implies that Snell’s law indeed applies to the present structures operating in the negative index region.

References