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Negative refraction by a planar Ag/SiO_2 multilayer at ultraviolet wavelength to the limit of silver

J. Zhao,[1,2] J. Gao,[1,a] Y. Deng,[2,3] H. Liu,[1] and X. Wang[1]
[1]*Key Laboratory for Optical System Advanced Manufacturing Technology, Changchun Institute of Optics, Fine Mechanics and Physics, Chinese Academy of Sciences, 3888 Dongnanhu Road, Changchun, Jilin, 130033, P. R. China*
[2]*University of Chinese Academy of Sciences, Beijing, 100039, China*
[3]*State Key Laboratory of Luminescence and Applications, Changchun Institute of Optics, Fine Mechanics and Physics, Chinese Academy of Sciences, 3888 Dongnanhu Road, Changchun, Jilin, 130033, P. R. China*

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For planar structured hyperbolic metamaterial, the shortest wavelength achievable for negative refraction is often limited by dielectric layers, which are usually wide band gap semiconductors that absorb light strongly at wavelength shorter than their absorption edge. Here we proposed that using SiO_2 may break such limitation based on effective medium theory. Through calculation and simulation we demonstrated broad angle negative refraction by a planar Ag/SiO_2 layered structure at wavelength down to 326 nm. Its imaging and focusing abilities were also presented. The lower limit of wavelength here is defined by the property of silver, whose permittivity turns positive below 324 nm. *© 2014 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution 3.0 Unported License.* [http://dx.doi.org/10.1063/1.4873156]

Manipulating light in the ultraviolet regime with subwavelength resolution can promote the development of photolithography, ultraviolet optoelectronic devices and fluorescence imaging. The "left-handed" material, which processes negative permittivity ($\varepsilon < 0$) and negative permeability ($\mu < 0$) simultaneously, would generate negative refractive index that is critical for subwavelength resolution.[1,2] People have demonstrated artificial two or three dimensional periodic structures for negative refraction from microwave to near-infrared regime.[3,4] However, scaling down these structures into sub-micrometer scale for visible and ultraviolet light is restricted by current nanofabrication techniques.[5] To release such constriction, Pendry pointed out that, since electric and magnetic fields decouple in the near field, for transverse magnetic (TM) polarized wave, only $\varepsilon < 0$ is required for negative refractive index.[6] Metals have $\varepsilon < 0$ in the visible and ultraviolet regime, thus a periodic one dimensional metal-dielectric layered structure has been proposed to realize negative refraction, with efficient transmission through evanescent wave coupling between nearby layers.[7] There are two configurations of such "hyperbolic metamaterial",[8] as shown in Figs. 1(a) and 1(b). The planar structure (Fig. 1(a)) has highly practical significance, since it is readily achievable by current thin film deposition techniques.

This planar structure, however, places additional requirements on material selection. The dispersion relation for hyperbolic metamaterial is given by:

$$\frac{k_x^2}{\varepsilon_z} + \frac{k_z^2}{\varepsilon_x} = k_0^2 \tag{1}$$

where k_0 is the wave vector in free space; k_x and k_z are wave vectors in the metamaterial along x and z directions respectively; ε_x and ε_z are the effective permittivities of the metamaterial along x and z directions respectively. Negative refraction could occur only if $\varepsilon_z \varepsilon_x < 0$,[9] which corresponds

[a]Author to whom correspondence should be addressed. Electronic mail: gaojs@ciomp.ac.cn

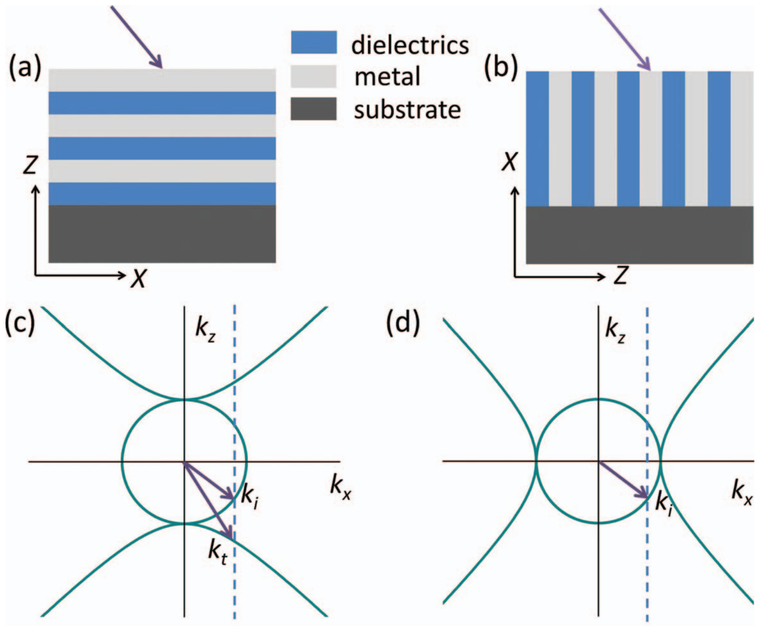

FIG. 1. (a) Planar and (b) vertical structured hyperbolic metamaterial. (c) Hyperbolic dispersion relation with $\varepsilon_x > 0$, $\varepsilon_z < 0$. (d) Hyperbolic dispersion relation with $\varepsilon_x < 0$, $\varepsilon_z > 0$. The circles in (c) and (d) represent the dispersion relation of free space.

to a hyperbola that "sits" on either k_z or k_x axis, as shown in Figs. 1(c) and 1(d) respectively. But to realize the planar structure, which means the light can be bent negatively when incident on the metamaterial's surface parallel to k_x, the hyperbola needs to "sit" on k_z axis (Fig. 1(c)) to satisfy the conversation of k_x. Otherwise, as the case of Fig. 1(d), incident light will be bifurcated into two refractive waves propagating into opposite directions along the k_x axis to conserve k_x.[10,11] When $\varepsilon_x > 0$ and $\varepsilon_z < 0$, the hyperbola "sits" on k_z axis. According to the effective medium theory,[9] when the layer is much thinner than the incident wavelength (<1/10 λ), ε_x and ε_z can be expressed as follows:

$$\varepsilon_x = f\varepsilon_m + (1-f)\varepsilon_d \tag{2}$$

$$\varepsilon_z = \frac{1}{f\varepsilon_m^{-1} + (1-f)\varepsilon_d^{-1}} \tag{3}$$

where f is the filling ratio of metal, defined by the ratio between the total thickness of metal to the total thickness of the metamaterial; ε_m and ε_d are the real part of permittivity of metal and dielectric, respectively. Considering that $0 < f < 1$, to achieve $\varepsilon_x > 0$ and $\varepsilon_z < 0$, the choice of metal and dielectric must meets $\varepsilon_d\varepsilon_m < 0$ and $-\varepsilon_m < \varepsilon_d$.

Normally the minus real part of permittivity of noble metals $-\varepsilon_m$ (e.g. Ag, which has relatively low loss from visible to ultraviolet wavelength range) is much larger than the permittivity of commonly available dielectric materials ε_d in visible frequency, leading to the dispersion relation in Fig. 1(d). This may explain why negative refraction in visible range usually relies on the vertical structure as in Fig. 1(b).[12,13] For Ag, the value of $-\varepsilon_m$ decreases from visible to ultraviolet wavelength, providing the opportunity for dielectric materials with high permittivity ($\varepsilon_d > 5$) to satisfy $-\varepsilon_m < \varepsilon_d$. That's why planar structures are usually realized in ultraviolet regime.[14–17] However, in real situation, many of those dielectric materials with high permittivity such as Zinc Oxide or Titanium Dioxide are wide band gap semiconductors that they have strong absorption at wavelengths shorter than their absorption edge (commonly longer than 340 nm), which defines the lower limit of wavelength of effective negative refraction.[14] Therefore, to realize negative refraction at even shorter wavelength is still a challenge.

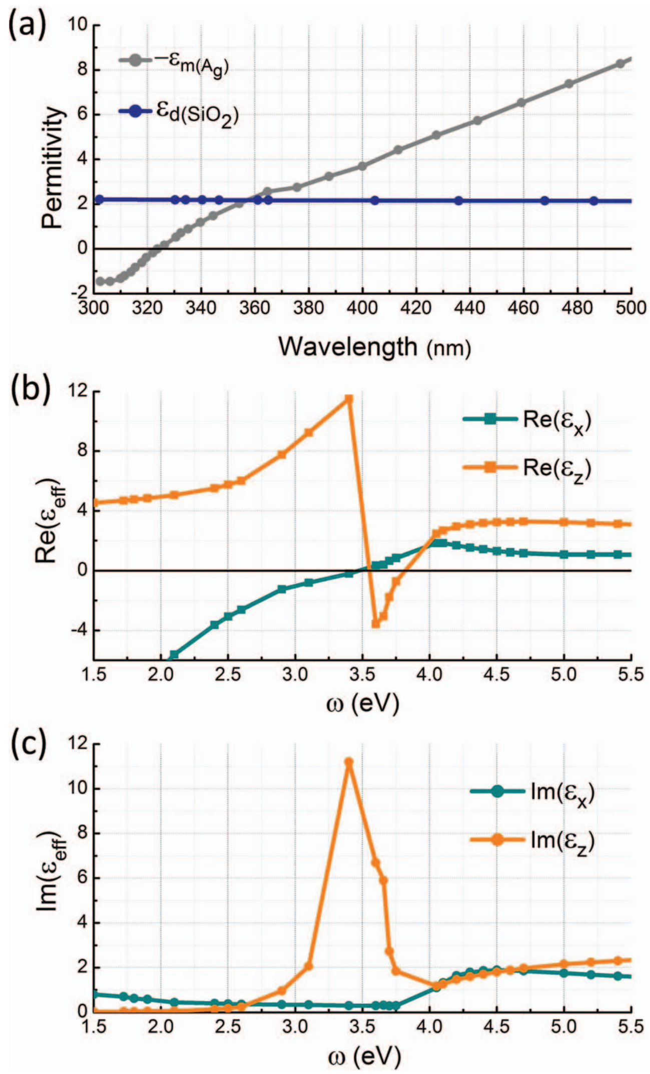

FIG. 2. (a) The real part of permittivity of SiO_2 (ε_{SiO_2}) and Ag ($-\varepsilon_{Ag}$) as a function of different wavelengths. (b) The real part and (c) the imaginary part of permittivity of ε_x and ε_z of a planar Ag/SiO_2 layered structure with $f = 0.5$ versus photo energy (eV).

Here we try to explore the possibility of using SiO_2 as the dielectric layer, which though has much smaller permittivity, is transparent to ultraviolet light at wavelengths down to 160 nm. The dependence of the real part of permittivity of SiO_2 (ε_{SiO_2}) and Ag ($-\varepsilon_{Ag}$) on the wavelength are shown in Fig. 2(a), calculated from their respective refractive index data edited by Palik.[18] With the wavelength decreases from 500 nm to 300 nm, ε_{SiO_2} is around 2.1, but ε_{Ag} progressively decreases and becomes smaller than ε_{SiO_2} at the wavelength around 360 nm. Therefore, negative refraction is possible for Ag/SiO_2 system at wavelengths below 360 nm. It should be noted that ε_{Ag} becomes positive at wavelengths below 324 nm, which sets the lower limit of negative refraction with Ag as the metal layer.

Now we consider a planar Ag/SiO_2 layered structure with $f = 0.5$. The real parts of ε_x and ε_z calculated from Eqs. (2) and (3) are shown in Fig. 2(b). Because of limited data, the plot is dispersive. But we can still find that the situation of $\mathrm{Re}(\varepsilon_x) > 0$ and $\mathrm{Re}(\varepsilon_z) < 0$ exists roughly between 3.55 eV and 3.80 eV, indicating that negative refraction may be obtained at wavelengths ranging from 326 to 350 nm. The imaginary parts of ε_x and ε_z calculated from Eqs. (2) and (3) are shown in Fig. 2(c). $\mathrm{Im}(\varepsilon_x)$ increases with the photon energy from 3.75 eV, suggesting increased transverse absorption at wavelengths shorter than 330 nm. On the other hand, $\mathrm{Im}(\varepsilon_z)$ increases rapidly as the photon energy

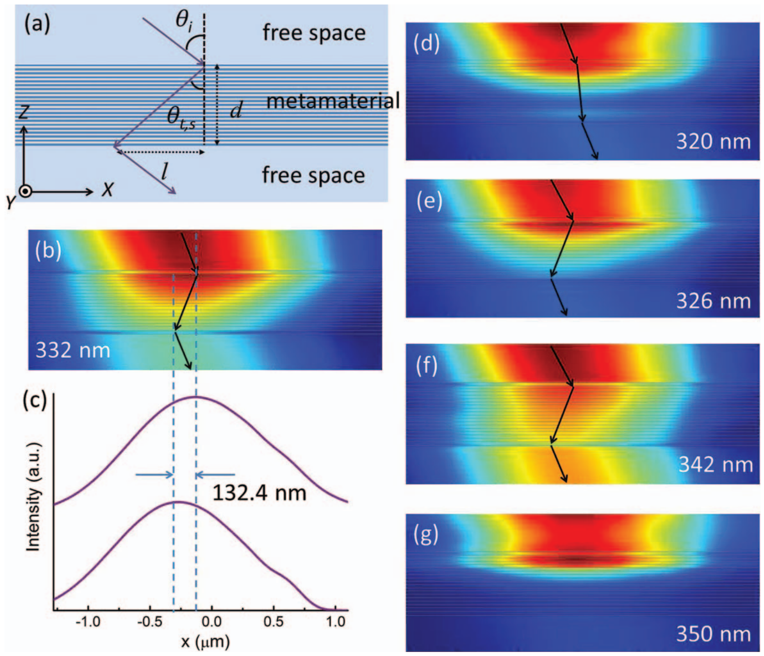

FIG. 3. (a) Simulation configuration where the structure is composed of planar Ag/SiO_2 periodic layers with each layer of 5 nm thick and totally 20.5 periods. A Gaussian TM polarized light propagating through the structure with a negative refraction is illustrated. (b) Simulated result of electric field intensity distribution for incident light with wavelength of 332 nm. (c) The 332 nm beam's intensity profiles on the upper and lower surfaces of the metamaterial. (d-g) Simulated results of electric field intensity distribution for incident light with wavelengths of 320 nm, 326 nm, 342 nm and 350 nm.

decreases from 3.7 eV, thus the longitudinal transmission loss is getting much higher at even longer wavelengths.

The negative refraction ability of the structure is invesitigated by finite-difference time-domain method. The slab for simulation contains alternate layers of Ag and SiO_2 with each layer of 5 nm thick and totally 20.5 periods (Fig. 3(a)). The number of periods chosen is to demonstrate obvious negative refraction effect (with sufficient light beam shift but less loss), and other number of periods works as well. The light source is a Gaussian TM polarized wave with an incident angle of 40°. The results are shown in Figs. 3(b)–3(g). At the wavelength of 320 nm, the refraction is positive (Fig. 3(d)), becasue the permitivity of Ag is positive here. At 350 nm, the light cannot propogate through the metamaterial (Fig. 3(g)), as the longitudinal loss is high. From 326 nm to 342 nm, negative refraction is observed (Figs. 3(b), 3(e), and 3(f)), agrees well with our estimation. The tranmission at 326 nm is much lower than that at 332 nm and 342 nm, which is contributed to the high transverse loss at 326 nm as we have pointed out. In following sections, we choose 332 nm as the wavelength of the light source.

Figure 3(c) shows the beam's intensity profiles on the two interfaces of the metamaterial in Fig. 3(b). It is observed that the incident light can be shifted by −132.4 nm. In addition, the metamaterial has a total thickness of 205 nm and the incident angle is 40°. Therefore, according to Snell's law, we can obtain the effective refractive index n_{eff} of the metamaterial, which is −1.19. Based on the same calculation, we further changed the incident angle from 20° to 50° by an interval of 5° and we obtained different n_{eff}, as are plotted in Fig. 4 by the squares.

The relationship between n_{eff} and θ_i can also be theoretically derived from the dispersion relation and Snell's law:

$$n_{eff} = \frac{\sin(\theta_i)}{\sin\left(-\tan^{-1}\left(\sin(\theta_i)\sqrt{\frac{\varepsilon_x}{\varepsilon_z(\varepsilon_z-\sin^2(\theta_i))}}\right)\right)} \quad (4)$$

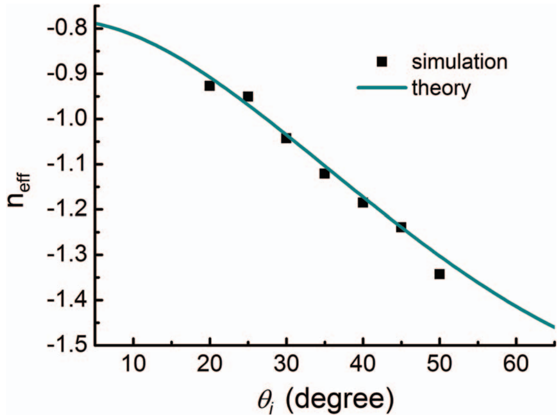

FIG. 4. The relationship between incident angle θ_i and effective refractive index n_{eff} obtained from theory (solid line) and simulation results (squares).

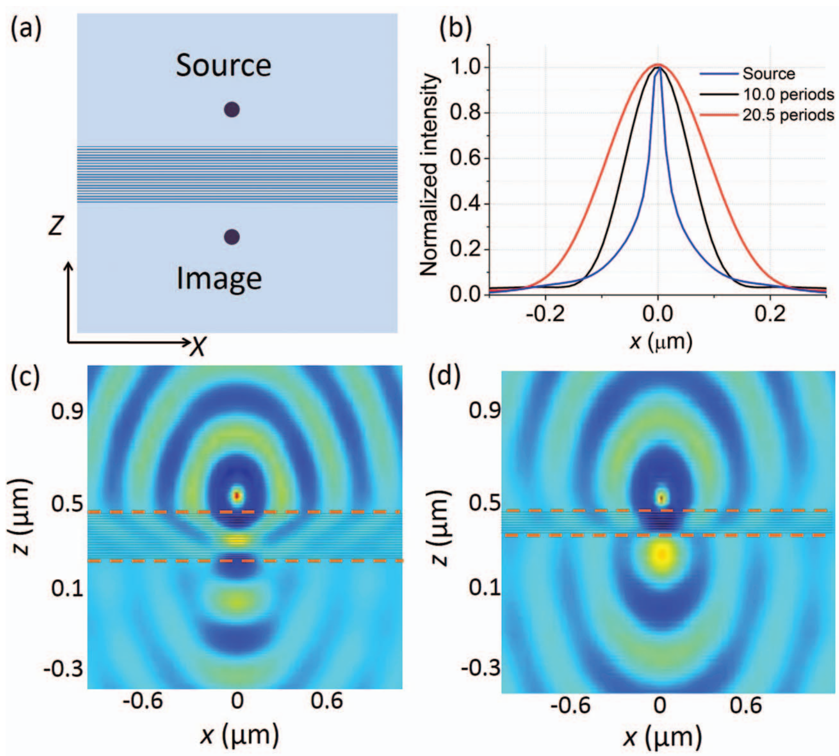

FIG. 5. (a) Illustration for the imaging of the metamaterial slab. (b) $|H_y|^2$ distribution in the source plane and in the image plane of metamaterial slabs with 10 and 20.5 periods. (c) The simulated result of phase wave front of H_y distribution with a point source placed 100 nm above a metamaterial slab with 20.5 periods. (d) The simulated result of phase wave front of H_y distribution with a point source placed 50 nm above a metamaterial slab with 10 periods.

which is also drawn in Fig. 4 by the solid line. We can see that the theoretical plot fits well with the simulation results. The metamaterial realizes effective negative refraction over a broad incident angle from 20° to 50°, with n_{eff} increases from −0.93 to −1.34.

A metamaterial with n_{eff} around −1 is desirable for imaging and focusing.[6,14,19] In Fig. 5(a), a point source is placed 100 nm above the metamaterial slab. From the distribution of phase wave front of H_y as shown in Fig. 5(c), we can see that an image is formed on the other side of the slab. The distance between source and image is measured to be 486 nm, which is close to $2d$ (410 nm). This

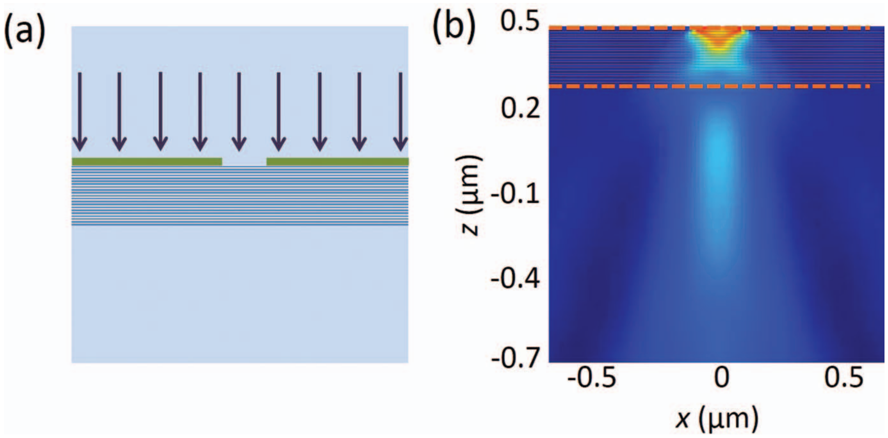

FIG. 6. (a) A TM polarized plane wave incident onto the slab with 20.5 periods through a 165 nm wide aperture on Cr mask. (b) The simulated result of power distribution.

further support that the refractive index of our metamaterial is near −1.[20] If we move the position of source, the image will move accordingly, keeping their distance to be near 2*d*, but the imaging quality would not change.

It should be noted that the feature size of the image is larger than the source spot. This is because when using real material (Ag and SiO_2 in our case) to construct negative refraction metamaterial, the evanescent wave that contributes to super-resolution is eliminated because of the high loss of real material.[21] Similar results are obtained elsewhere using real material as well.[13,22,23] If we reduce the thickness of metamaterial to 10 periods, the absorption loss is reduced and the imaging ability is improved (Fig. 5(d)). As is shown in Fig. 5(b), the full width half maximum (FWHM) is reduced from 0.61λ of 20.5 periods to 0.39λ of 10 periods, realizing superdiffraction limit imaging.[13]

Furthermore, when applying a TM polarized plane wave onto the slab through a 165 nm wide aperture on a Chromium mask (Fig. 6(a)), a focus below the slab is observed from the power distribution (Fig. 6(b)), showing the flat focusing ability of the metamaterial. The elongation of the focus point results from the angle-dependent n_{eff} (Fig. 4).[12]

In summary, we have realized effective ultraviolet negative refraction by a planar Ag/SiO_2 layered structure at wavelength down to the limit of Ag, whose permittivity turns positive below 324 nm. Because of the transparency of SiO_2 in ultraviolet regime, the previous wavelength limitation that using wide band gap semiconductors as the dielectric layers has strong absorption effect is circumvented. Theoretical calculation and numerical simulation show broad angle negative refraction by the structure for TM-polarized light from 342 nm to 326 nm with an effective refractive index around −1. The imaging and focusing abilities have also been demonstrated. To realize negative refraction at even shorter wavelength, alternative metals need to be considered. This work may lead to a clear direction for choosing real materials to constitute the desirable hyperbolic metamaterial.

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