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Negative refraction: An intrinsic property of uniaxial crystals

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We theoretically and experimentally show that negative refraction can be realized at the surface of conventional uniaxial crystals by orientating the crystals with their optic axes at a certain angle θ_0 to the normal of the light incoming surface. The concept of negative refraction can be extended to be an intrinsic property of all uinaxial crystals. It is revealed that the angular range for incident light to yield negative refraction attains its maximum that only depends on the difference of two indices of refraction $|n_e\text{-}n_0|$ when $\tan^2\theta_0=n_0/n_e$. The careful experiments on positive uniaxial crystal YVO_4 and negative unaxial crystal calcite ($CaCO_3$) give results in good agreement with the calculated ones. It should be noted that the negative refraction reported here differs from that occurring in metamaterials or photonic crystals in that the wave vector of light k in conventional crystals does not form a left-handed triplet with electromagnetic field E and H.

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Negative refraction (NR) is a phenomenon where light is refracted to propagate along the same side as the incident light with respect to the normal of the interface, contrary to the normal light refractions. It can be used to realize the "superlens" with a resolution smaller than the wavelength and many other optic applications.[1–5] Veselogo[6] first theoretically explored the possibility of NR and concluded that it can be achieved at an interface of two materials, one possessing both positive permittivity ε and permeability μ and another both negative ε and μ that necessarily lead to a negative index of refraction $n<0$.[3] The first experimental observation[1] was realized by using a metamaterial composed of a two-dimensional array of repeated unit cell of copper strips ($\varepsilon<0$) and split ring resonators ($\mu<0$) at microwave frequencies. The subsequent experiments and numerical simulations confirmed the existence of NR.[7–12] Photonic crystals (PC), another kind of artificial materials, usually are made of dielectrics or metals with smaller cells comparable to the light wavelengths. They were recently reported to exhibit negative refraction at microwave[2,13,14] and infrared frequencies,[15] though these materials have both a local positive ε and μ. The physical mechanism[13,16–19] that is different from the one for the metamaterials, arises from the dispersion relation in PCs. The band structure calculations[14,19] showed that the wave vector k_t inside the PCs can form a left-handed triplet with electromagnetic field E and H at a certain frequency range, leading to the Poynting vector S_t which is antiparallel to k_t, i.e., $S_t \cdot k_t<0$.

For a long time, all known naturally occurring materials exhibit positive refraction. NR at the visible light wavelengths was recently realized in a twinning structured interface associated with two uniaxial YVO_4 crystals.[20] What is shared here with metamaterials and PCs is that the incident and refracted lights are on the same side with respect to the normal of the interface. The mechanism in the uniaxial crystal case is the components of the Poynting vector and wave vector parallel to the interface (the x axis, for example) satisfy $S_{tx} \cdot k_{tx}<0$, without requiring $S_t \cdot k_t<0$. In fact, for a light traveling in a uniaxial crystal, $S_t \cdot k_t<0$ cannot happen, and therefore, a left-handed triplet with electromagnetic field E and H will not be expected. That is not the kind of negative refraction originally proposed by Veselogo.[6] Liu and co-workers[21] theoretically made a generalization of the negative refraction phenomenon happened at this kind of artificial interface. They predicted that the NRs can occur at a wide variety of interfaces associated with two uniaxial media, in particular, at the interface formed by an isotropic medium with a uniaxial one. Yau *et al.*[22] demonstrated that a NR phenomenon at a visible light wavelength can occur at the surface of a uniaxial crystal $CaCO_3$. In this paper, we extend the concept of NR in conventional crystals by showing NR is an intrinsic property for uniaxial crystals. This means that NR can be observed in all uniaxial crystals including these with the tetragonal, hexagonal and trigonal symmetries with their optic axes orientated at certain angles to the interface normals. It is revealed that the incident angular range to yield NR and the corresponding refracted angular range directly relate to the index of refraction of the unaxial crystals. The results obtained by experiments on both positive and negative uniaxial crystals support this conclusion.

The crystals used in our experiments are YVO_4 and calcite ($CaCO_3$) with tetragonal and trigonal symmetry, respectively. Both are uniaxial axis and have relative large birefringence $|n_e\text{-}n_0|$. The former is a positive-axis crystal with $n_e=2.2154$ and $n_0=1.9929$ (at 630 nm),[23] while the later a negative-axis one with $n_e=1.4852$ and $n_0=1.6557$ (at 630 nm).[24] Now consider a rectangular slab with a size of 15 mm $\times$ 10 mm $\times$ 20 mm cut from a YVO_4 crystal in such a way to make its optic axis (the crystallographic axis c) parallel to a pair of surfaces while at an angle $\theta_0=45°$ to the normal of another pair of surfaces (light entering surface) of the slab. We choose a coordinate system to let one surface of the former pair lie within the x-z plane and simultaneously one of the latter pair within the x-y plane. Then the normal of the light entering surface will be along the z axis, see Fig. 1(a). If an incident light whose electric field E is polarized along the y direction, this is just as the case for the ordinary light to travel through in the crystal. The refraction of the

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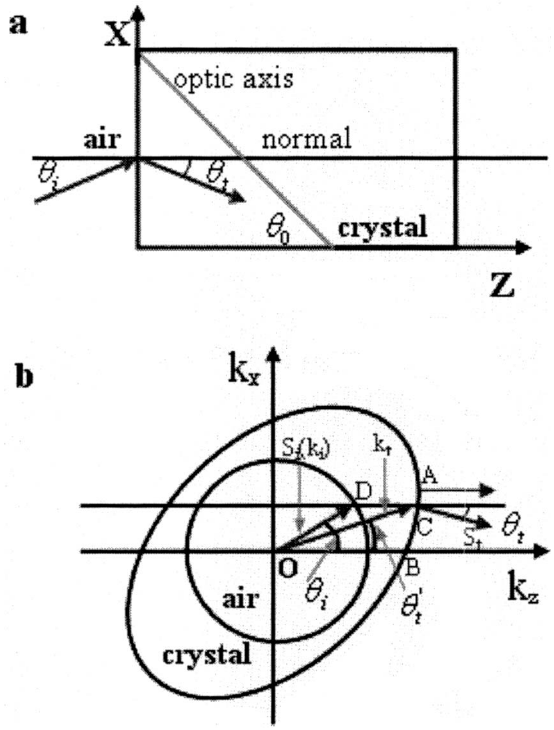

FIG. 1. (a) The orientation of a rectangular slab crystal in the chosen orthogonal set of axes. The optic axis (in green) is parallel to the x-z plane while at an angle θ_0 to the normal of the light entering surface of the crystal which lie within the x-y plane. (b) Representative diagrams of the dispersion relation equations for incident light (circle) and refracted light (ellipse). θ_i, θ_t, and $\theta_{t'}$ are incident angle, refracted angle and the angle between the k_t wave vector for the refracted light and the normal line of crystal. Point A is where the normal line for the ellipse is parallel to the z axis, namely, $\partial k_{tx}/\partial k_{tz}=0$, leading to S_t parallel to the z axis.

beam occurring at the surface will obey Snell's law at whatever incident angle. So we only consider the extraordinary light case, where its E field lies in the x-z plane. The dispersion relation can be obtained by solving the Maxwell's equation for refracted light in terms of the chosen coordinate system

$$\frac{(k_{tx}\cos\theta_0 + k_{tz}\sin\theta_0)^2}{n_e^2} + \frac{(k_{tx}\sin\theta_0 - k_{tz}\cos\theta_0)}{n_0^2} = \frac{\omega^2}{c^2} \tag{1}$$

if we only consider the non-magnetic crystals. The dispersion relation for the incident light is then reduced as

$$k_{ix}^2 + k_{iz}^2 = \frac{\omega^2}{c^2} \tag{2}$$

considering $n_e=n_0=1(\approx 1)$ for light in vacuum (in air). Where ω is the circular frequency and c is the light velocity in vacuum, $k_{ix}(k_{tx})$ and $k_{iz}(k_{tz})$ are the x and z components of the wave vectors for the incident (transmitted) beams.

Figure 1(b) graphically shows Eqs. (1) and (2) with the crystal orientation angle being θ_0. For the incident light, the time-averaged pointing S_i, which stands for the direction of propagation of light θ_i, coincides with the wave vector k_i. For the refracted ray, S_t, however, deviates from the direction θ_t' of the wave vector k_t due to the dispersion for the extraordinary ray. The refracted angle θ_t can be geometrically determined from the k-ε dispersion ellipse. According to the derivations from Maxwell's equation, the components of wave vectors parallel to the interface conserve for the incident and refracted ray, i.e., $k_{ix}=k_{tx}$ (Snell's law). For an incident light OD (k_i) with an angle θ_i, the angle θ_t' (OC) is easily found by applying the boundary condition. The S_t is in turn determined since it is invariably parallel to the direction of the normal line at the corresponding point C on the ellipse. We expect that a relation $S_{ix}S_{tx}<0$ will hold if light is incident at a certain angle such as OD shown in Fig. 1(b). This means that the incident and refracted beams are on the same side with respect to the normal of the light incoming surface, i.e., the negative refraction phenomenon, contrary to the case with positive refractions where the two beams are on the opposite side with respect to the normal.

The incident angular range that yields the negative refraction can be analyzed by utilizing Fig. 1(b). We note that there always exists a special point A on the ellipse shown in Fig. 1(b) at which the normal line is parallel to the z axis, namely, $\partial k_{tx}/\partial k_{tz}=0$, leading to S_t parallel to the z axis. The angle θ_i is usually not equal to zero unless $\theta_0=0$ or 90°. The corresponding angles for $k_i(S_i)$ and k_t vectors are now denoted as θ_{im} and θ_{tm}', respectively. If the incident angle satisfies $0<\theta_i<\theta_{im}$, then the angle of k_t satisfies $0<\theta_t'<\theta_{tm}'$, the relation $S_{ix}S_{tx}<0$ will always hold. That is, at the incident angular range $0<\theta_i<\theta_{im}$ negative refraction occurs. If incident angle θ_i is outside the above angular range, relation $S_{ix}S_{tx}>0$ will hold, suggesting that the positive refraction occurs. So θ_{im} determines the incident angular range to observe negative refraction for a uniaxial crystal.

The angle θ_{im} is dependent on the orientation angle θ_0. We found that at point A the following relation holds:

$$k_{tx} = \frac{(n_e^2 - n_0^2)\sin\theta_0\cos\theta_0}{(n_e^2\sin^2\theta_0 + n_0^2\cos^2\theta_0)^{1/2}}. \tag{3}$$

Then,

$$\tan\theta_{im} = \frac{k_{ix}}{k_{iz}} = \frac{k_{tx}}{\sqrt{1-k_{tx}^2}} \tag{4}$$

considering $k_{ix}=k_{tx}$.

From Eqs. (3) and (4), it is easily obtained that θ_{im} arrives at its maximum values through the following relation:

$$\tan\theta_{im} = \frac{1}{\sqrt{\dfrac{1}{(n_e-n_0)^2}-1}} \tag{5}$$

when $\tan^2\theta_0=n_0/n_e$. The maximum refracted angle θ_{tm} will be observed if $\theta_i=0$ [OB in Fig. 1(b)], which is represented by the following formula:

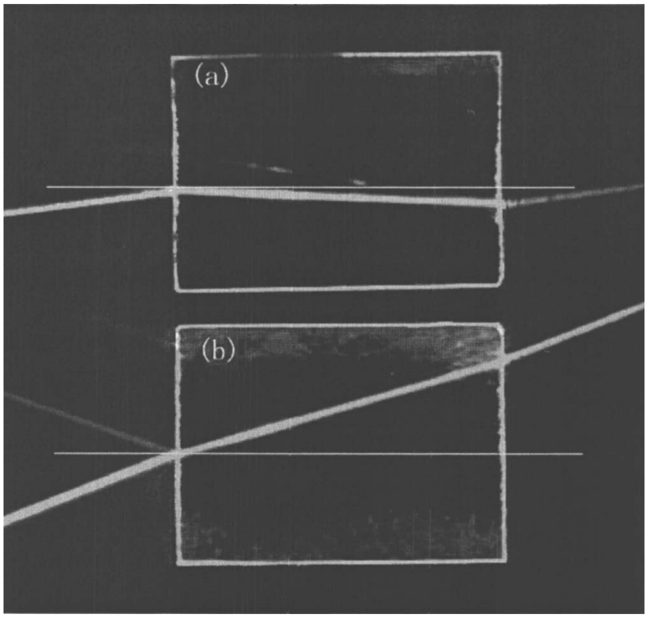

FIG. 2. Photograph of refractions at the surfaces YVO_4 crystal slabs by 530 nm laser beam from air. The white lines are the normal of the light entering surface of the slabs plotted for eye guide. (a) Negative refraction, the incident angle θ_i is about 8.3° and the refracted angle θ_t is about −2.5°. Note that the incident and the refracted lights are on the same side of the normal. (b) The positive refraction, the incident angle θ_i is about 20.1° while the refracted angle θ_t is about 17.2°. Note that the incident and the refracted lights are on the opposite side of the normal.

$$\tan \theta_{tm} = \frac{\left(\frac{n_e^2}{n_0^2} - 1\right)\left(\frac{n_e}{n_0}\right)^{1/2}}{1 + \frac{n_e^3}{n_0^3}}. \qquad (6)$$

We carefully examined the refractions that occurred at the surface of the YVO_4 crystal slab by measuring the incident and the refracted angles. The experiment was conducted by using a polarized 630 nm laser. The angles were measured over a wide angular range by a goniometer with the aid of a digital power meter. Both the positive and the negative refraction were observed. As an example, Figs. 2(a) and 2(b) clearly show that the NR indeed can occur at the surface of the YVO_4 crystal under a certain incident angle, apart from the positive refraction that we are familiar with by using a 530 nm laser for the demonstration purpose. We found that the incident angle range to yield negative refraction is from 0° to 12.8°. The refracted beam lies in the angular range from −6.02° to 0°, see Fig. 3(a). Outside this incident angular range, positive refractions occur. The down-right inset gives the schematic of angular ranges of incident angle θ_i and refracted angle θ_t with respect to the normal for NR (in yellow color). The calculations of the incident and the refracted angles were also performed based on combining the k-ε dispersion relations for incident and refracted beams. The remarkable agreement is obtained for both positive and negative refractions.

To further confirm the above arguments, we conducted

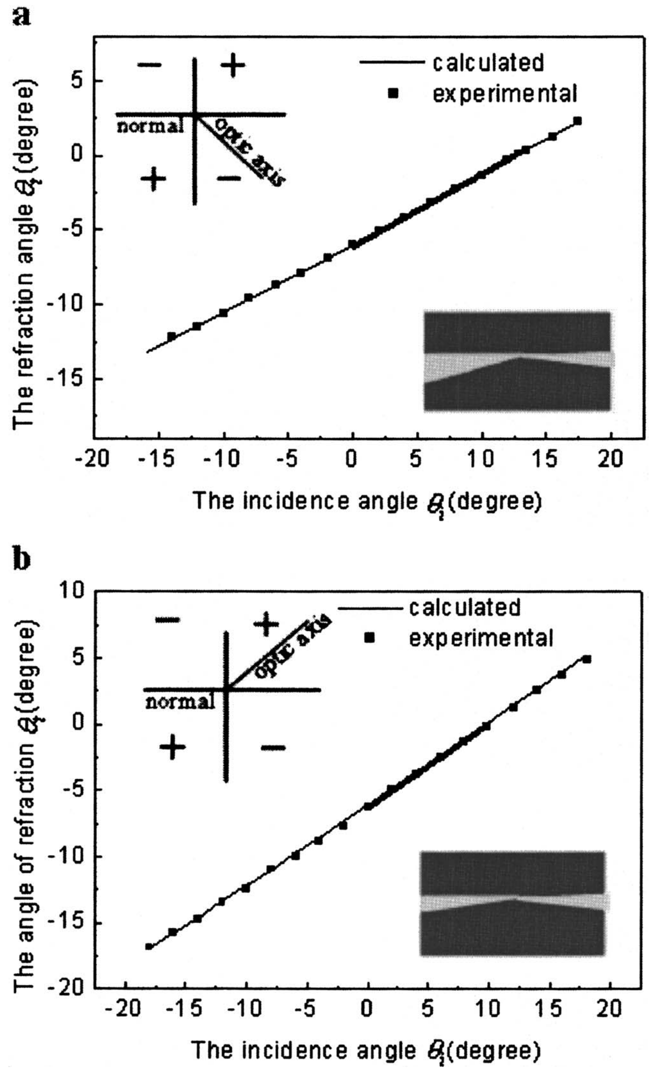

FIG. 3. The comparison between the experimental and calculated light propagation directions for (a) YVO_4 crystal and (b) $CaCO_3$ crystal. The data points are measured with a 630 nm laser beam. The curves are calculated with the index of refraction given in text. The red parts of the curves indicate the occurrences of negative refractions. The upper left insets indicate the sign of incident and refracted angles, and the optic axis directions (in orange). The lower right inset gives the schematic of angular ranges of incident angle θ_i and refracted angle θ_t with respect to the normal for negative refraction. Note the incident angular range for YVO_4 is wider than that for calcite.

similar experiments by using the negative axis crystal calcite. The sample was cut and polished into a rhombohedron with the size of 13 mm × 10 mm × 18 mm. The orientation angle θ_0 was set as 46°. The incident angular range that yields the NR was measured to be from 0° to 9.83°, while the corresponding refracted angle from −6.15° to 0°, see Fig. 3(b). Outside this incident angular range, positive refractions occur. The lower right inset indicates the incident and refracted angular ranges for NRs (in yellow color). Again results of good agreement between the measured and calculated angles are obtained for both positive and negative refractions.

From Eqs. (5) and (6) we can conclude that the NR may occur in all uniaxial crystals since their index of refraction n_0 is always different from n_e, leading to both nonzero θ_{im} and θ_{tm}. The maximum angular range θ_{im} for incident light to yield NR depends on the quantity $|n_e\text{-}n_0|$, while the maximum refracted angular range θ_{tm} on the quantity n_e/n_0 if the optic axis of crystals is orientated at an angle $\theta_0 = \arctan(n_0/n_e)^{0.5}$ to the normal of the light entering surface. For most uniaxial crystals, θ_{im} and θ_{tm} are quite small values according to Eqs. (5) and (6) since their $|n_e\text{-}n_0|$ and n_e/n_0 values are quite close to zero and 1, respectively, at the visible light wavelengths. Even so, the NR can be regarded as an intrinsic property for uniaxial crystals since it is only associated with their indices of refraction. The underlying mechanism for this intrinsic property is that the Poynting vector for refracted light S_t can possesses an opposite sign to that for the incident light in their components along the surface at a certain angular range under proper orientations of crystals.

The results reported here do not violate any existing fundamental physical law. The Poynting vector S and electromagnetic field E and H form a right-handed triplet inside the uniaxial crystals, contrary to the cases for metya materials and PCs. However, they are common in controlling light propagations: light is refracted to propagate along the same side as the incident light with respect to the normal. NRs are also expected to observe in biaxial crystals including those with orthorhombic, monoclinic and triclinic symmetry. In these crystals, two optic axes exist. There will be more light entering surfaces to choose to produce the negative refractions. The NR produced by only one piece of optic anisotropic crystal, without the twinning structure formed by direct bonding of two crystals will make the device fabrication much easier. These results have broadened our knowledge of physical properties for crystals. Moreover, this intrinsic property of the crystals may provide many new experimental opportunities to investigate physical phenomena associated with the NR.

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