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Loss induced negative refraction and super-prism effect at highly absorptive interface*

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It is well-established that waves are inhomogeneous in a lossy isotropic medium, and the validation of the classical Snell's law is still questionable for light refraction at the dissipative and dispersive interface. With high absorption, direct experimental investigation is rather difficult due to the extremely short penetration depth; i.e., the skin depth. In this paper, a simple and unified description of this issue is proposed, which can be applied to both materials with anomalous dispersion and in the Drude region. The gradient $\nabla_k \omega$ is found to be incident angle θ_i-dependent, and the direction of the group velocity may deviate significantly from the phase velocity due to the loss induced permittivity structure. The physics behind the negative refraction effect is explained, and a novel loss induced super-prism effect is also predicted.

Keywords: Snell's Law, Drude model, anomalous dispersion, lossy isotropic media

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1. Introduction

Beyond the optically transparent dielectric materials, the validation of the classical Snell's law is questionable for dispersive and dissipative media, where the refraction index $\tilde{n}(\omega) = n + \mathrm{i}k$ becomes complex and frequency ω-dependent. The wave transmitted into these media is, in general, inhomogeneous. Its phase velocity differs greatly from the velocity of the energy flow, while the latter may even lack precise meaning.[1] There are still many controversies about the light propagation in these absorbing materials, including negative refraction,[2,3] superluminal light velocity,[4] Goos–Hänchen effect,[5,6] and so on. Even regarding the seemingly simple issue, such as the reflection and refraction of electromagnetic wave at a planar lossy interface, it is still ambiguous—in spite of the fact that there are already many serious theoretical and experimental studies devoted on this subject. Born and Wolf pointed out that the group velocity may exceed c and may even be negative due to the anomalous dispersion. They also proposed a set of typical equations to describe the refraction of light entering from dielectric to metal.[7] Some argued against this theory by pointing out that these equations are not reversible when applied to the opposite situation of light entry; i.e. from metal to dielectric.[8–10] Other generalized versions of the Snell's laws are based on complex or real valued boundary conditions, and quite different results are obtained.[11–15] Moreover, the experimental verification is extremely difficult because the associated heavy absorption results in opacity and a very short penetration depth. Chen's group carried out a series of experiments to measure the light refraction at a pure Ag/air or Au/air interface, and a negative refraction effect was observed.[9,16,17] They attributed this observation to the group refractive index $n_g = n + \omega(\mathrm{d}n/\mathrm{d}\omega)$, which can be negative due to the dispersive properties of the refractive index n in the Drude region. However, to reduce the parallel displacement, which is induced by the non-uniform absorption, the wedge angles of the prism like metallic samples in their experiments are extremely small. Consequently, it is difficult to study the influence of incident angle θ_i on the light refraction.[7,11,15]

In this work, we study the light refraction at a planar lossy interface, which can be applied to either Drude medium or anomalous dispersion medium.[18] According to the Krames–Kronig relation, the real part of the permittivity exhibits rapid variation in the neighborhood of heavy absorption, which leads to a peculiar equifrequency contour in the wave vector space. This is the physics behind the loss-induce negative refraction effect in this paper. Based on the real valued boundary conditions, we calculate the incident angle dependent equifrequency contours to determine the propagation direction of the refracted beam in absorptive materials. The negative refraction effect is explained based on the dispersive feature of the dielectric function and the θ_i-dependent wave vector k. A novel

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loss induced super-prism effect is also predicted at the boundary between positive and negative refraction regions. This work may provide further understanding to the negative refraction effect at a metal/dielectric interface, which is widely used in various optoelectronic devices in the fields of plasmonics and metamaterials.

2. Modified Snell's law

In an absorptive material, the permittivity ε is generally a complex function of angular frequency ω, and so is the wave vector $\widetilde{\boldsymbol{k}} = \boldsymbol{k}_\mathrm{r} + \mathrm{i}\boldsymbol{k}_\mathrm{i}$, which is also complex. Then, the solution for a plane wave becomes

$$\boldsymbol{E}(\boldsymbol{r},t) = \boldsymbol{E}\mathrm{e}^{\mathrm{i}\widetilde{\boldsymbol{k}}\cdot\boldsymbol{r}-\mathrm{i}\omega t} = \boldsymbol{E}\mathrm{e}^{-\boldsymbol{k}_\mathrm{i}\cdot\boldsymbol{r}}\mathrm{e}^{\mathrm{i}\boldsymbol{k}_\mathrm{r}\cdot\boldsymbol{r}-\mathrm{i}\omega t}. \tag{1}$$

These inhomogeneous plane waves have planar surfaces of constant amplitude and phase, which are no longer parallel to each other. We distinguish the imaginary part of $\widetilde{\boldsymbol{k}}$, which is in fact related to the attenuation constant α. We also determine that the waves travel along the direction of group velocity, which in the three-dimensional space is given by

$$\boldsymbol{v}_\mathrm{g} \propto \boldsymbol{\nabla}_{\boldsymbol{k}}\omega. \tag{2}$$

Now let us consider the light refraction at an absorptive interface, where a propagating ray is incident onto a medium with complex dielectric permittivity $\widetilde{\varepsilon}_2$, as plotted in Fig. 1. Both the wave vectors of incidence and refraction lie entirely at the incidence plane, which can be divided into parallel and normal components, $\boldsymbol{k}_\parallel$ and $\boldsymbol{k}_\perp$, to the interface. In the lossy material, we denote the complex wave number as $\widetilde{\boldsymbol{k}}_\mathrm{t}$, and its real part as $\boldsymbol{k}_\mathrm{t}$. The relation between the above mentioned wave vectors are clearly plotted in Fig. 1.

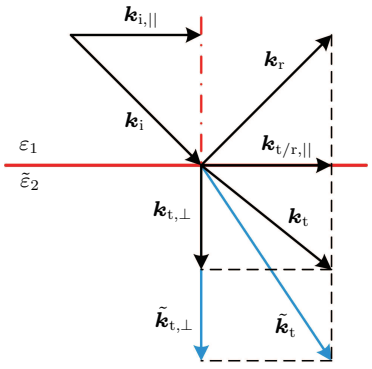


Fig. 1. Orientations of the wave vectors at the interface, where it is complex. The black vectors denote real phase vectors, while the blue vectors are complex. The subscripts $\perp$ and $\parallel$ define the perpendicular and parallel components to the interface, respectively.

In the following section, we will show that the real refracted wave number $\widetilde{k}_\mathrm{r}$ is θ_i-dependent, which is the key idea of this paper. To determine the refraction direction via Eq. (2), the contour plot of $\omega(k_\parallel, k_\perp)$ in the k-space will be obtained, which is usually a smooth convex surface for a specific ω. Based on the well-known theorem of vector calculus, the propagation direction is along the normal $\boldsymbol{\nabla}_k\omega$ of these curves. The exact direction for refraction points in the direction of increasing ω and away from the interface.[19]

Applying the real valued boundary conditions, it follows that

$$\boldsymbol{k}_{\mathrm{i},\parallel} = \boldsymbol{k}_{\mathrm{r},\parallel} = \boldsymbol{k}_{\mathrm{t},\parallel} = \boldsymbol{k}_\parallel, \tag{3}$$

in which the parallel components of incident wave vector is θ_i-dependent; i.e.,

$$k_\parallel = \omega\sqrt{\varepsilon_1\mu_1}\sin\frac{\theta_\mathrm{i}}{c_0}. \tag{4}$$

This is the direct result that follows the conservation of momentum. Since the total wavevector $\widetilde{k}_\mathrm{t}$ in the lossy media satisfies the dispersion equation $\widetilde{k}_\mathrm{t}^2 = \widetilde{\varepsilon}_2\mu\frac{\omega^2}{c_0}$, we can deduce from Eq. (3) that only its normal component is complex. One can see from Fig. 1 that the normal component of $\widetilde{\boldsymbol{k}}_\mathrm{t}$ includes real and imaginary parts, such as

$$(\widetilde{\boldsymbol{k}}_\mathrm{t})_\perp = \boldsymbol{k}_{\mathrm{t},\perp} + \mathrm{i}\boldsymbol{k}_{\mathrm{t},\perp}, \tag{5}$$

where the attenuation vector $\boldsymbol{k}_{\mathrm{t},\perp}$ is always perpendicular to the interface. Therefore, we can make a simple remark on the refracted planar wave that its planes of constant amplitude are always parallel to the interface.[7] We determine that the direction of phase velocity is determined only by the real $\boldsymbol{k}_\mathrm{t}$, which can point towards the interface in case of negative refraction.

Figure 2 schematically shows the light refraction on a lossy surface, where the equifrequency surfaces of the dielectric and lossy media are plotted above and below the $k_\parallel$ axis, respectively. In the homogenous and lossless dielectric, the equifrequency surface is a simple circle with radius $r = k_\mathrm{i}(\omega) = \sqrt{\varepsilon_1\mu_1}\frac{\omega}{C_0}$. In contrast, the equifrequency diagrams of absorptive medium are quite different from the simple circles, and depend on the exact dispersive property of the permittivity. So we need to make a contour plot of $\omega(k_\parallel, \pm k_\perp)$ based on $\widetilde{\varepsilon}_2$, where the plus-minus sign is due to the fact that the phase velocity can point towards or away from the interface. According to Fig. 1, the term $k_\perp$ is given by

$$k_\perp = \mathrm{Re}\left[\sqrt{\frac{\omega\widetilde{\varepsilon}_2\mu_2}{c_0^2} - k_\parallel^2}\right]. \tag{6}$$

Note that the $k_\parallel$ is a function of incident angle θ_i, so both $k_\perp$ and $k_\parallel$ in the absorptive material are θ_i-dependent.

Figure 2 separately plots the upper contour for the incident medium and the lower contour for the transmitted medium corresponding to a particular frequency ω for clarity. The incident angle θ_i corresponds to a specific wave vector $\boldsymbol{k}_\mathrm{i}$ on the incident contour. Since the $k_\parallel$ is conserved across the interface, we draw a dashed line through the incident $\boldsymbol{k}_\mathrm{i}$ and perpendicular to the interface (here, the $k_\parallel$ axis). So the equifrequency surface is for the energy conservation and the

black-dashed line is for the conservation of momentum. The fixed-$k_{\parallel}$ line intersects the transmitted contour in two points, which yields the two possible phase velocities $\boldsymbol{v}_{\rm p1}$ and $\boldsymbol{v}_{\rm p2}$. To further determine the refraction direction, we need to eliminate one intersection whose group velocity points towards the interface.

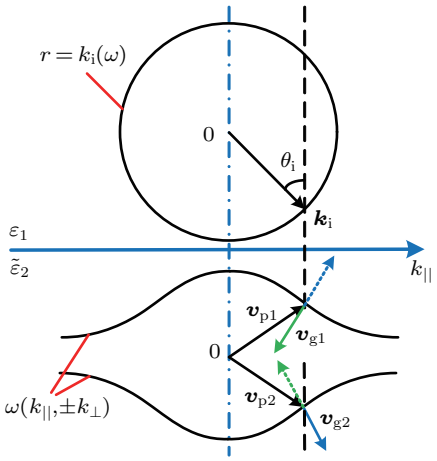


Fig. 2. Typical diagram of equifrequency surface for light refraction at a lossy interface. The black-dashed line is normal to the interface and illustrates the conservation of $k_{\parallel}$. The black arrows show the possible wave vector, i.e., the phase velocity direction, while the colored arrows show the possible group velocity directions. The green solid arrow corresponds to negative refraction, while the blue solid arrow denotes the positive refraction.

If the size of the equifrequency surface $\omega(k_{\parallel},\pm k_{\perp})$ decreases as the frequency ω increases, then the group velocities of the two intersected points are along the inward normal to these surfaces. In this case, the group velocities correspond to the two green arrows in Fig. 2, however, only the solid green arrow $\boldsymbol{v}_{\rm g1}$ is physically reasonable. In the opposite case where the equifrequency surface expands as frequency ω increases, the group velocities of the two points are shown as the blue arrows in Fig. 2, but only $\boldsymbol{v}_{\rm g2}$ represents a distinct refracted wave. Note that $\boldsymbol{v}_{\rm g1}$ and $\boldsymbol{v}_{\rm g2}$ illustrate negative and positive refraction, respectively. Finally, we can obtain the modified Snell's law via $\tan\theta_{\rm t} = \pm(\mathrm{d}k_{\parallel}/\mathrm{d}k_{\perp})$, which takes the form

$$\tan\theta_{\rm t} = \pm\frac{\sqrt{\varepsilon_1\mu_1}\sin\theta_{\rm i}}{\mathrm{Re}[\sqrt{\tilde{\varepsilon}_2(\omega)\mu_2 - \varepsilon_1\mu_1\sin^2\theta_{\rm i}}]}, \tag{7}$$

where the positive and negative signs denote the positive and negative refraction, respectively. If the imaginary part of $\tilde{\varepsilon}_2(\omega)$ is zero, then equation (7) will be reduced to the well-known expression of the classical Snell's law automatically. It is widely accepted that the criterion of the negative refraction requires both negative real parts of ε and μ for an isotropic medium, but based on the proposed theory above, the negative refraction may occur if the size of the equifrequency surface shrinks as the frequency ω increases, which may be referred as loss-induced negative refraction.

3. Anomalous dispersion

If the optical dispersion of a material is anomalous, then its optical refractive index decreases when the excitation frequency increases. Based on the Kramers–Kronig relations, anomalous dispersion usually occurs simultaneously with heavy absorption.[1] Let us start with the classical Lorentz oscillator model for the dielectric constant, and consider a typical dielectric function which can be cast into[18]

$$\varepsilon(\omega) = 1 + \sum_i \frac{S_i\omega_i^2}{\omega_i^2 - \omega^2 - \mathrm{i}\omega\gamma_i}, \tag{8}$$

where ω_i is the resonance frequency of the ith absorption line, and S_i and γ_i represent the strength and the damping constant, respectively. Figure 3 plots a concrete example of this model with only two absorption lines ω_1 and ω_2, and the dark areas denote the anomalous dispersion regions.

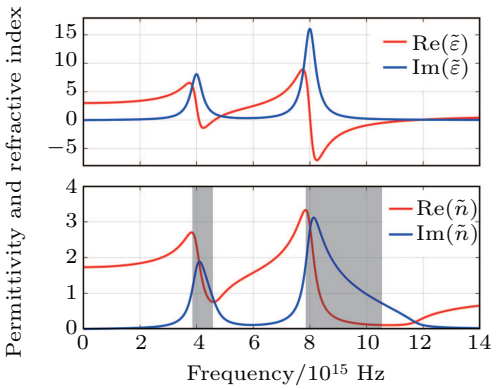


Fig. 3. Frequency dependent permittivity and refractive index of anomalous dispersion at the two resonance frequencies. The parameters are $S_1 = S_2 = 1$, $\gamma_1 = \gamma_2 = 0.5\times10^{15}$ s^{-1}, and $2\omega_1 = \omega_2 = 8\times10^{15}$ s^{-1}.

Let us examine if the negative refraction effect can take place at the interface of a medium with anomalous dispersion. Therefore, we plot the isofrequency contours in the $(k_{\parallel},k_{\perp})$ plane around the first absorption line ω_1.

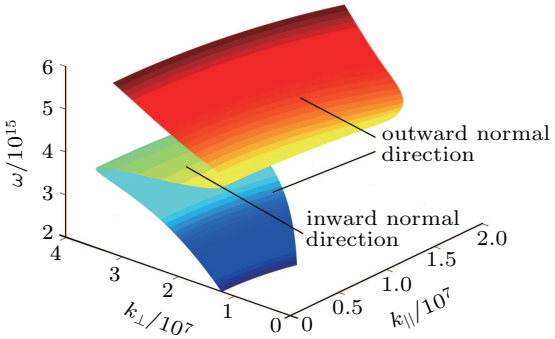


Fig. 4. The isofrequency surface between 2×10^{15}–6×10^{15} s^{-1} in the medium with anomalous dispersion.

For the sake of clarity, we only plot the upper half branch of the isofrequency surface $\omega(k_{\parallel},k_{\perp})$. It is clear from Fig. 4 that as the frequency ω increases, some part of the surface moves outward, which indicates that the group velocity is

along the outward normal to these surfaces. Therefore, positive refraction occurs in the corresponding frequency ranges. The other part of the surface moves inward as ω increases, which corresponds to the negative refraction. It is clear from Fig. 4 that the negative refraction does exist around the anomalous dispersion region due to the absorption. Next, we will show that the criterion for negative refraction in this model is incident angle dependent, while the group velocity index defined by $n_g = n + \omega(dn/d\omega)$ is not applicable to this case. There is a simple way to determine whether the refraction is positive or negative. At a fixed frequency ω, $k_{\parallel}$ is also fixed, so the corresponding criterion for negative refraction can be simply written as

$$\frac{dk_{\perp}}{d\omega} < 0. \tag{9}$$

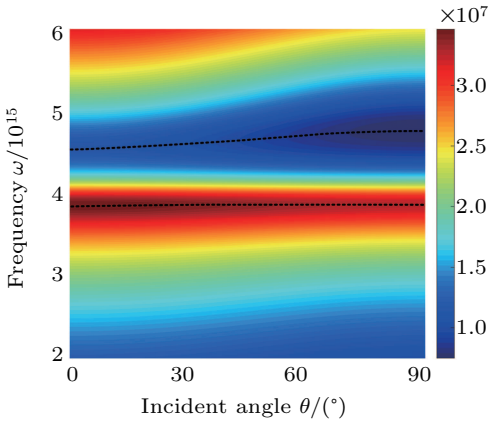


Fig. 5. $|k_{\perp}|$ as a function of the incident angle θ and angular frequency ω. The area between these two dotted lines denotes the negative refraction region $dk_{\perp}/d\omega < 0$, which is incident angle dependent.

Figure 5 demonstrates that the criterion for negative refraction in fact depends on the incident angle θ_i, which does not coincide with the anomalous dispersion region. One more thing that needs to be mentioned is that super-prism effect can occur as the frequency transits from negative refraction to positive refraction. The super-prism effect refers to the phenomenon that the refracted angle changes enormously due to a small change in frequency.[19] Suppose a light beam with a finite frequency width $2\Delta\omega$ is incident on the interface, and the boundary frequency ω_b between the positive and negative refraction happens to locate in the middle of the frequency range. Usually, the contours corresponding to ω_b locate at the most inside or outside in the nearby frequency range. Consider a simple example that the contour of ω_b is the outermost curve, as shown in Fig. 6(b). So for $\omega \in [\omega_b - \Delta\omega, \omega_b]$, positive refraction takes place, while for $\omega \in [\omega_b, \omega_b + \Delta\omega]$, negative refraction occurs. Consequently, half of the light beam will be positively refracted, while the other half part will be negatively refracted, and the angle between the positively and negatively refracted beams is dependent on the gradient of the contours. When such a beam is incident on the interface, we will have two refracted beams instead of one, and a schematic diagram is plotted in Fig. 6. In summary, a novel super-prism effect will take place when the boundary frequency ω_b is included in the incident bandwidth. This effect will be more clear when applied to the next example of noble metal described by Drude model.

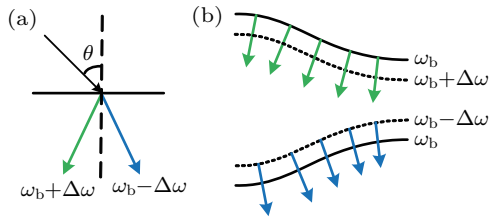


Fig. 6. (a) Super-prism effect around the frequency ω_b at the edge between the positive and negative regions, where the equifrequency contour of ω_b locates at the most outside. (b) Equifrequency contours of the medium with anomalous dispersion, and ω_b corresponds to the boundary frequency with the biggest size.

4. Drude model

In this section, we will treat the Drude free electron model for metal. We only consider the noble metals of silver and gold, which are the two most important metals in plasmonics and metamaterials. Based on the Drude model, the frequency dependence of the dielectric function of the free electrons in metal is

$$\varepsilon(\omega) = \varepsilon_{\infty} - \frac{\omega_p^2}{\omega^2 + i\Gamma\omega}, \tag{10}$$

where ω_p is the volume plasma frequency, ε_{∞} is a constant offset, and Γ is the damping constant. Figure 7 plots the equifrequency diagram for silver, where the wavelength ranges from 377 nm to 1.88 μm. It can be seen from Fig. 7 that the equifrequency surfaces move inward as the frequency increases, so negative refraction occurs at the silver surface for all visible regions.

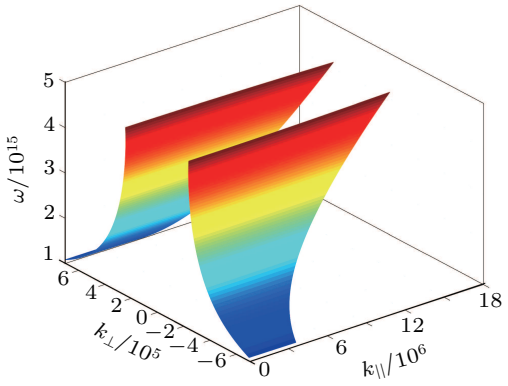


Fig. 7. Equifrequency diagram for silver, where the parameters are $\varepsilon_{\infty} = 5$, $\omega_p = 9.2$ eV, and $\Gamma = 0.021$ eV.[17]

For some noble metals like gold, the interband effect already starts in the visible range. To take in the contribution from the bound electrons, the corresponding Lorentz term should be added[18]

$$\varepsilon_{\rm ib}(\omega) = 1 + \frac{\omega_{\rm l}^2}{\omega_0^2 - \omega^2 - {\rm i}\Gamma\omega}, \tag{11}$$

where ω_0, $\omega_{\rm l}$, and ω denote the oscillation frequency, density, and damping of bound electrons, respectively. The overall dielectric function of gold contains both the Drude term in Eq. (10) and the $\varepsilon_{\rm ib}$ term in Eq. (11). Figure 8 plots the equifrequency diagram for gold, where the surfaces move inward, outward, and inward again as the frequency increases. Therefore, both negative refraction and super-prism effects can take place when a light beam is incident on the gold interface. The results are in accordance with the experimental research done by Chen's group, in which the light refraction goes from negative to positive in the visible region at the pure air/Au interface.[16,17] It is also clear in the neighborhood of the boundary frequency between the positive and negative refraction that if the frequency of the incident beam varies across this boundary frequency, the refracted beam will switch between the positive or negative refraction. Consequently, a very small change in the incident frequency may result in a large variation of the refracted angle, due to the above mentioned switch. This is the reason for the loss induced super-prism effect.

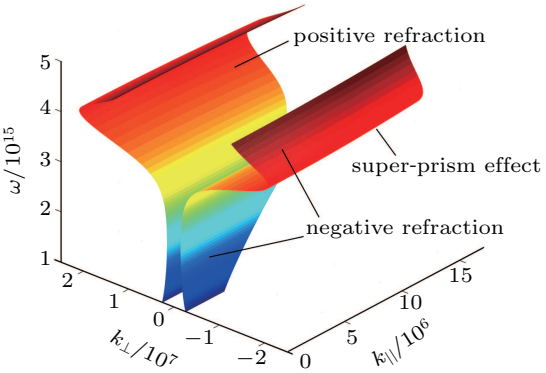


Fig. 8. Equifrequency diagram for gold, where the parameters are $\varepsilon_\infty = 7.8$, $\omega_{\rm p} = 9.1$ eV, $\Gamma = 0.072$ eV, $\Gamma_0 = 2.8$ eV, $\omega_{\rm l} = 3$ eV, and $\gamma = 0.6$ eV.[18]

5. Conclusion

The classical Snell's law is extended to the light refraction at the interface of the lossy isotropic media, in which the waves are inhomogeneous. Instead of using the conventional group index definition $n_{\rm g}$, which is incident angle independent, we apply the equifrequency diagram in the k-space to determine the refraction direction. A new criterion for negative refraction effect at the lossy interface is proposed, which is related to the dependence of equifrequency surface on the frequency. A novel super-prism effect is also predicted, which is likely to occur at the boundary frequency between the negative and positive refraction.

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