Intrinsic abnormal electromagnetic medium based on polar lattice vibration

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A new class of media with abnormal electromagnetic parameters has been attracting increasing attention because of its exotic properties and potential application. Currently, typical metamaterials are mainly composed of artificially designed metallic periodic structures. However, due to the limitations of available fabrication technologies, physical size and material effects, it is difficult to realize these abnormal properties by these artificial structures in the high-frequency regime. Therefore, it is important to find materials with intrinsic abnormal electromagnetic responses. In this field, a new mechanism based on the interaction between polar lattice vibrations and electromagnetic waves has been proposed. In this paper, we review progress in this field.

abnormal electromagnetic medium, polar lattice vibration, intrinsic property, infrared, indefinite medium

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Abnormal electromagnetic (EM) medium refer to a kind of material system with abnormal permittivity and permeability (negative or less than unity), which is typically represented by left handed material (LHM) with negative permittivity and permeability at the same time. Abnormal EM medium is normally realized via metamaterials based on metallic periodic structures [1–12]. The abnormal properties in the metamaterial usually originate from the artificial structure, and not from the materials themselves. These materials attracted much attention not only because of their scientific significance, but also their potential for enlargement of the performance scale, and design of new functional materials. Therefore, the initial investigations of LHMs and metamaterials have been honored as breakthroughs in 2003 and 2006, respectively, by *Science* magazine [13,14]. Also, they were elected to be one of the top ten breakthroughs in material science by the journal, *Materials Today* [15].

Conventional metamaterials are based on highly engineered artificial structure units, normally with electric or magnetic resonances, such as wires and split ring resonators (SRRs) [2,4–9]. In the infrared and optical regions, metamaterials are showing more and more shortcomings for the current fabrication technologies, characteristic physical size and material effects of metals in mesoscopic scale. However, intrinsic EM responses in nature existed materials provide another choice. For example, ferromagnetic resonances in ferrites can provide tunable negative permeability at microwave frequency [16–18], and the electric resonance in ferroelectric can be used to achieve a negative permittivity [19,20]. However, because of their high frequency cut-offs, these EM resonances cannot be extended to the infrared or optical regions, restricting the development of intrinsic abnormal media. Therefore, it is a challenging problem to realize intrinsic abnormal EM properties in the high frequency regime. Recently, abnormal EM media based on polar lattice vibrations in the infrared region have attracted

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increasing attention, and significant discoveries have been made. In this paper, we focus on the latest researches and give a review of the field.

1 Polar lattice vibration and abnormal permittivity

In the 1950s, Born et al. [21] considered the coupling between a polar lattice vibration mode and EM wave, and quantitatively described the dielectric dispersion relation. For a single oscillator, the dielectric dispersion relation of polaritonic materials can be expressed as

$$\varepsilon = \varepsilon_{\infty} + \frac{(\varepsilon_0 - \varepsilon_{\infty})\omega_T^2}{\omega_T^2 - \omega^2 + i\gamma\omega}, \quad (1)$$

where ε_{∞} is the high-frequency dielectric constant; ε_0 is the static permittivity; ω_T is the transverse optical phonon frequency; and γ is the damping coefficient.

Figure 1 shows the theoretical dielectric dispersion curve of a single vibration mode. The curves can be divided into two parts, one is the region of high permittivity and the other is the region of abnormal permittivity. When the frequency is a little lower than ω_T , the real part of permittivity is high. However, when the frequency is a little greater than ω_T and satisfies that $\omega_T < \omega < \omega_L$, the real part of the permittivity is less than zero. ω_L is the longitude optical phonon frequency, which usually corresponds to zero in the dispersion curve. When the frequency is greater than ω_L , the permittivity is less than one. Actually, the region ($\varepsilon < 1$) is the abnormal permittivity region we focus on. In addition, ω_T and ω_L satisfy the Lyddane-Sachs-Teller (LST) relation $\omega_L / \omega_T = \sqrt{\varepsilon_0 / \varepsilon_{\infty}}$, which implies that a much wider abnormal permittivity region can be gained using materials with high ε_0 and low ε_{∞} . If there are $1/\nu_a$ ion pairs in unit volume, the value of ω_T depends on the composition of the crystal, and is given by:

Figure 1 Dielectric dispersion curve for a single vibration mode.

$$\omega_T = \frac{2}{3} \left(\frac{\pi}{\bar{M} \nu_a (\varepsilon_0 - \varepsilon_{\infty})} \right)^{1/2} Ze(\varepsilon_{\infty} + 2), \quad (2)$$

where $\bar{M}$ is the reduced mass in a primitive cell and Ze is the effective charge of the ions. ω_T is related to the properties of the anion and cation, including mass and electronegativity. In another word, by changing the composition of the material, the value of ω_T can be controlled. Furthermore, this can yield a tunable abnormal permittivity. Similarly, it can also be extended into other crystals with complex structures. In this case, the dielectric dispersion is modeled by the sum of independent damped harmonic oscillators [21,22]:

$$\varepsilon = \varepsilon_{\infty} + \sum_i \frac{\Delta\varepsilon_i \omega_{iT}^2}{\omega_{iT}^2 - \omega^2 + i\gamma_i \omega}, \quad (3)$$

where the oscillator parameters $\Delta\varepsilon_i$, ω_{iT} , and γ_i are the frequency, oscillator strength, and damping coefficient of the i th transverse vibration mode, respectively.

The abnormal EM medium can be realized both in regions of high permittivity and abnormal permittivity via the interaction between polar lattice vibrations and EM waves. We will give detailed descriptions of these two mechanisms.

2 Infrared abnormal EM media with tunable permittivity

The abnormal permittivity can be realized by the polar lattice vibration, and regions with abnormal properties have relations with the composition of the material as described in Section 1. Therefore, a tunable abnormal permittivity can be achieved by changing the composition of the crystal using the methods of solid solution or doping. A series of $\text{Ca}_{1-x}\text{Sr}_x\text{F}_2$ solid solutions were chosen. Their room temperature reflectance spectra were measured at near-normal incidence in the infrared region. The dielectric properties were measured using the relation between the permittivity and reflectivity, $R = \left| \frac{\sqrt{\varepsilon} - 1}{\sqrt{\varepsilon} + 1} \right|^2$, for non-magnetic

materials. The dielectric dispersion curves retrieved from the analysis of the reflectivity data are shown in Figure 2. Because of the interaction between the polar lattice vibrations and the EM waves, the real parts of the permittivity show a resonance mode and the position of the dielectric resonance shifts towards lower frequency as the composition of the material changes from CaF_2 to SrF_2 . For the pure crystals, the dielectric resonance is sharp. The resonance strength of SrF_2 is a little stronger than that of CaF_2 , because the electronegativity of Sr ions is larger than that of Ca ions. However, for the solid solution, the partial substitution of Ca ions with Sr ions will cause distortions in the CaF_2 lattice, which may enhance the damping factor during

Figure 2 (a) Real and (b) imaginary parts of the dielectric dispersion curves of $\text{Ca}_{1-x}\text{Sr}_x\text{F}_2$ with different x values. Modified from [23].

the vibration and result in a larger dissipation. As a result, the resonance strength of the solid solutions decreases significantly. When the composition is close to 1:1, the resonance strength reaches its minimum.

Figure 3 presents the permittivity of $\text{Ca}_{1-x}\text{Sr}_x\text{F}_2$ as a function of composition at three typical frequencies, 330, 480, and 620 cm^{-1} . It shows that as the molar fraction x , varies from 0 to 1, the variation range of the permittivity can cover part of the negative region, the whole of the ultra-low (zero to one) region, and reaches permittivity of one. Additionally, it can be seen from Figure 3 that when the frequency is close to the transverse optical frequency, the dielectric loss is enhanced. The large dielectric loss will cause a strong absorption of EM waves in the material, which is unfavorable for the application. Therefore, the working frequency should be far from the resonance frequency when using the polar lattice vibration to realize abnormal permittivity.

3 Abnormal EM media composed of high-permittivity particles based on Mie theory

In the paper published by Born et al. [24] in 1908, they derived a rigorous solution for plane EM wave diffraction by spherical particles. Combining Mie theory with effective medium theory, Lewin [25] investigated the EM responses

Figure 3 (a) Real and (b) imaginary parts of the permittivity of $\text{Ca}_{1-x}\text{Sr}_x\text{F}_2$ at 400, 480, and 620 cm^{-1} . Modified from [23].

of the composite with dielectric or magnetic spherical particles dispersion in another background matrix, and derived the effective permittivity and permeability:

$$\varepsilon_{\text{eff}} = \varepsilon_h \left[1 + \frac{3\nu}{\frac{F(\theta) + 2K_e}{F(\theta) - K_e} - \nu} \right], \quad (4)$$

$$\mu_{\text{eff}} = \mu_h \left[1 + \frac{3\nu}{\frac{F(\theta) + 2K_m}{F(\theta) - K_m} - \nu} \right], \quad (5)$$

where $K_e = \varepsilon_h / \varepsilon_p$ and $K_m = \mu_h / \mu_p$, ε_p and μ_p are the permittivity and permeability of particles, respectively. ε_h and μ_h are the permittivity and permeability of the matrix, respectively. ν is the volume fraction of the spherical particles; $F(\theta) = 2(\sin\theta - \cos\theta) / [(\theta^2 - 1)\sin\theta + \theta\cos\theta]$; $\theta = k_0 r \sqrt{\varepsilon_p \mu_p}$; r is the radius of the particles; and k_0 is the free-space wavenumber. It can be seen that, $F(\theta)$ becomes negative infinite when its denominator nears zero, which results in either a negative permittivity or permeability. Figure 4 shows the effective EM parameters for the composite.

Figure 4 Effective EM parameters of the composite. $\varepsilon_p=100$; $\mu_p=\varepsilon_h=\mu_h=1$; $v=0.064$.

For Mie resonances, particles with a large permittivity or permeability are required. However, the permittivity and permeability of most materials will decrease as the frequency increases and cannot maintain a high level. The large permittivity present during the lattice vibrations provides a promising way to realize Mie resonances at a high frequency. Thus, it allows for an abnormal EM medium in the infrared region. Wheeler et al. [26] proposed a scheme to obtain negative effective permeability using a collection of polaritonic spheres. Based on the polar lattice vibration of LiTaO_3 crystal, which result in a high permittivity at infrared frequency, the effective parameters were calculated and the results are shown in Figure 5.

Later, Wheeler et al. [27] also proposed a core-cell structure, with a LiTaO_3 sphere core and a coating on its surface whose dispersion relation follows the Drude model. Using this structure they achieved an isotropic LHM in far infrared region. Schuller et al. [28] experimentally observed the abnormal EM properties of SiC crystal whiskers based on Mie resonance. They also proposed that it could be possible to obtain a LHM by combining the TE_{011} and TM_{011} modes using SiC particles of two sizes.

In addition, by selecting suitable polaritonic crystals, this kind of abnormal EM medium can be achieved in other infrared regions.

4 All-dielectric LHM at infrared frequency

LHM is a kind of metamaterials, whose permittivity and permeability are both less than zero. Similarly, theoretical and experimental researches primarily focus on the artificial periodic metallic structures, which are typically represented by the composition of SRRs and metallic wires [4–9,29–33]. In terms of intrinsic properties of materials, a common method is to use the polariton as an electric inclusion to provide negative permittivity. This can be combined with dielectrics which possess negative permeability. In this case, an all-dielectric LHM can be achieved without metallic structures.

Figure 5 Effective (a) permeability and (b) permittivity of LiTaO_3 spheres. Reprinted figure with permission from [26].

Based on the cation- TiO_3 lattice vibration mode, which is located at about 110 cm^{-1} , to provide a negative permittivity, our group selected CaTiO_3 (CTO) rods as electric inclusion. By combining the CTO rods with LiTaO_3 (LTO) spheres based on Mie magnetic resonance, we realized an all-dielectric LHM at 3.5 THz [34]. The EM transmission behaviors were simulated using the CST microwave studio based on the finite-difference time-domain (FDTD) method. The effective EM parameters, which were retrieved according to effective medium theory, are shown in Figure 6. There is a passband, which is marked with a blue arrow, near the overlap of the forbidden bands of the LTO spheres and the CTO rods. The passband is a left-handed band, in which the effective permittivity, permeability, and the refractive index of the composite are all less than zero. The effective region with negative index is 3.53–3.64 THz. The minimum value, which is about -1 , is achieved at 3.54 THz.

The electric and magnetic inclusions are not isolated and they interact with each other. Using EM simulation, the coupling effects between them were investigated [34]. The results show that when lattice vibrations occur in the CTO rod, a displacement current forms inside the CTO rod, which results in an annular induced magnetic field. There is an interaction between the induced magnetic field and the magnetic resonance field in the LTO sphere, which destroys the magnetic resonance and results in a composite without left-handed (LH) properties. Therefore, the distance between the

Figure 6 Effective EM parameters of the CTO/LTO composite. Reprinted by permission of [34].

electric and magnetic inclusions, the intrinsic dielectric resonance frequency of the electric inclusions, and the propagation direction of the EM wave all affect the LH properties of the LTO/LTO composite [34].

5 Indefinite medium based on anisotropic lattice vibrations

Negative refraction is a fantastic phenomenon that when an EM wave is incident from a medium to another and the incident light and refracted light are located on the same side of normal direction. For a long time, the negative refraction has been considered as a unique characteristic of LHM [1,3–7], until Lindell et al. [35] investigated the optical properties of anisotropic media and proposed a kind of indefinite medium, a non-LH negative refraction was realized. Indefinite medium is a kind of abnormal EM medium. It is also a kind of uniaxial and anisotropic medium, in which only one of the four components of ϵ and μ is negative, while the others are positive [35–37]. If only consider the e light with a wave vector $\mathbf{k}$ propagating in the xz plane, and the non-magnetic medium satisfies $\epsilon_x > 0$ and $\epsilon_z < 0$, then the dispersion relation in the medium is as follows:

$$\left(\frac{\omega}{c}\right)^2 = \frac{k_z^2}{|\epsilon_x|} - \frac{k_x^2}{|\epsilon_z|}, \quad (6)$$

where the subscript x or z refers to the components of the vector in the corresponding direction. In this case, the equi-frequency surface (EFS) in the crystal has a hyperbolic dispersion relation. The refracted directions of the wave vector and Poynting vector do not lie in the same side of the normal direction, thus a negative refraction of light. This theory has already been confirmed by experiments and simulations [36]. The angles of refraction for the wave vector and Poynting vector are given by $\theta_k = \tan^{-1}(k_x/k_z)$ and $\theta_s = \tan^{-1}(S_x/S_z) = \tan^{-1}(k_x \epsilon_x / k_z \epsilon_z)$, respectively. At present, the indefinite medium is still achieved by artificial structures, such as metal/dielectric multilayer [37] and metal nanowire arrays [36]. Although many existing materials, such as a YVO_4 twin structure [38], can also show negative refraction, this negative refraction can only be achieved over a small region of incident angles (about 0° – 12°). Thus, it is not an all-angle negative refraction. Therefore, we proposed an indefinite medium based on the strong anisotropy of polar lattice vibrations in uniaxial crystals. This will provide a promising method for the realization of all-angle LH properties using intrinsic properties of materials [39].

In a uniaxial crystal, because the parameters of the lattice are different in the two directions, parallel and perpendicular to optical axis, the lattice vibration frequencies for the same vibration mode will be different. This will result in an indefinite permittivity with a strong anisotropy, which is shown in Figure 7.

Figure 7 Dielectric dispersion in a uniaxial crystal with a simple structure.

We used α - Al_2O_3 single crystal to serve as an example and measured its reflectivities with different polarizations [39]. The retrieved permittivities are shown in Figure 8. Four indefinite permittivity regions are present, and they are indicated by the shaded regions in the figure. In these regions, α - Al_2O_3 can be considered as an indefinite medium and is in the family of abnormal EM media.

The magnetic field component in the y -direction demonstrates a negative refraction when the 500 cm^{-1} EM wave is refracted at the interface between the α - Al_2O_3 and vacuum, which is shown in Figure 9. It can be seen that after the refraction of the EM wave at the surface, the wave vector $\mathbf{k}$, which simply considered as the propagation direction of the peaks and crests, is refracted positively; however, the Poynting vector, $\mathbf{S}$, is refracted negatively. Additionally, the large losses of the permittivity cause a strong absorption. Therefore, the refractive wave can only propagate for several wavelengths in the crystal. This is why the negative refraction is hard to detect.

Figure 10 shows the calculated results of the refracted θ_k and θ_s at different incident angles at 500 cm^{-1} . It can be seen that an all-angle negative refraction is achieved using

Figure 8 Dielectric dispersion curves of α - Al_2O_3 single crystal. Reprinted with permission from [39], Copyright [2010], American Institute of Physics.

Figure 9 Simulation results for 500 cm^{-1} TM light at incident angles of 45° (a) without and (b) with considering the loss of the permittivity. The c axis is the optical axis and is perpendicular to the interface. The source is a TM Gaussian beam with a width of $20\text{ }\mu\text{m}$. $\varepsilon_{\perp} = 7.56 + 1.90i$ and $\varepsilon_{//} = -3.14 + 1.30i$.

Figure 10 Calculated results of θ_k and θ_s at 500 cm^{-1} .

the intrinsic indefinite medium. When the dielectric loss increases in the material, θ_s and the absolute value of the refractive index will both decrease.

Although we used α - Al_2O_3 single crystal as an example, and its region with indefinite permittivity only covers part of the range $400\text{--}900\text{ cm}^{-1}$. However, by selecting other uniaxial crystals with different lattice vibration frequencies, such as SiC [40], this kind of indefinite medium can be achieved in other infrared regions. Thus, negative refraction can be achieved on a much wider scale.

6 Summary

The theory of coupling effects between polar lattice vibrations and EM waves has already been formed over a period of more than half a century. However, the application of this interaction mechanism in functional materials is rarely reported. It is actually a promising method for designing optical abnormal media based on intrinsic properties. The variety of crystal structures in nature provides abundant resources for abnormal dielectrics. By selecting suitable compositions and structures, it is possible to extend the abnormal EM properties over a wide frequency region.

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