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Metamaterial composed of coated nano-spheres at infrared frequencies

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Abstract. A negative index of refraction in a three-dimensional collection of coated nano-spheres is observed in the terahertz (THz) spectral region. This negative-index metamaterial is a binary composite of two different types of coated nano-spheres (SiO_2@SiC/SiO_2@AZO); in one type SiO_2 spheres coated with a semiconductor (SiC) nano-shell and in another one, nano-shells made of a plasmonic material (AZO). The effective medium parameters derived by extended Maxwell–Garnett (EMG) effective medium theory by specifying the Mie resonances of the coated nano-spheres in the binary composite. The results are supplemented with frequency band structure and transmission curves, calculated by an accurate electromagnetic multiple-scattering technique. The predictions of the EMG theory are in agreement with those of the multiple-scattering technique, providing the effective medium approximation a helpful guide for the experimental works in this field.

1 Introduction

Designed materials composed of various inclusions can show strange and unique electromagnetic properties which are not intrinsic in the individual components. These engineered composites, called metamaterials, play an important role whenever material response in the electromagnetic spectrum is restricted and also empower the fabrication of novel devices. In the past few years, metamaterials present a new class of phenomena in optics that originate from a negative index of refraction (a feature which is not found in natural materials): frequency filters, super-lensing, optical multiplexers, near-field imaging, cavity super-resonance, focusing and extending devices [1–3].

There has been so much improvement in the expansion of negative-index metamaterials within the past few years. This development for metamaterials that work at microwave frequencies has been much faster than for those at optical or infrared frequencies. The first structure with negative index of refraction [4,5] was a combination of split-ring resonators (SRRs), which prepare a negative permeability [6] and continuous metallic wires (with a negative permittivity) [7]. However, split-ring resonators cannot be easily scaled down to nanometer scale due to their complicated shape [8]. There have been some recent efforts to propose structures simpler than the common metamaterials (SRRs and wires) which can represent the negative refractive index band. For instance, a negative-index composite has been reported which consist of two different types of non-overlapping spheres [9]: one made of a polaritonic material which is responsible for negative permeability and the other from a Drude-like material that prepares negative permittivity. The magnetic activity in such composite, which occurs within the region of the Mie resonance of a single sphere, is a result of the enhancement of the displacement current inside each sphere which, in turn, gives rise to a macroscopic magnetization of the composite. The required electric response is also induced by a material with a Drude-type permittivity such as noble metals or doped semiconductors in the infrared.

In this paper, we report on the design of a negative-index metamaterial in the form of a binary composite which comprises two sets of coated nano-spheres in the air matrix. All the nano-spheres have similar cores (SiO_2) but they are covered by different nano-shells; in one set, the nano-shells are made of a semiconductor (SiC) that possesses negative magnetic permeability in certain frequency regions and in the other set they are made of a new plasmonic material (Al:ZnO) possessing negative electric permittivity. Under suitable choice of geometric parameters (thickness of nano-shells, radius and volume filling fraction of the nano-spheres) a negative refractive index band emerges. To study this binary composite theoretically, the extended Maxwell-Garnett (EMG) effective medium theory is used. In

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addition, its electromagnetic response is studied by the use of the full multiple-scattering method. It is found, that the predictions of the EMG theory are in agreement with those of the multiple-scattering approach

The layout of this article is as follows: In sect. 2, we present the effective medium theory and show how the effective parameters for a composite arise within the context of an EMG theory. Section 3 presents the effective medium parameters calculated for the binary composite (SiO_2@SiC/SiO_2@AZO). A scattering matrix technique is then used to calculate the band structure of an equivalent periodic structure, which further verifies the results of the effective medium theory. Finally, the results are summarized in sect. 4

2 Effective medium theory

Effective medium theories are usually used for macroscopic modeling of a non-uniform medium by considering different microscopic configurations for the material. A material which consists of a composition of discrete media can be presumed as a homogeneous one only at a large observation scale. One of the intriguing topics about the heterogeneous materials (mixtures) is calculating their effective dielectric function by having appropriate information about the structural properties of the mixture, such as the permittivity of each constituent medium, percentage of each medium and their shapes [10,11]. There are many different effective medium theories used for modeling electromagnetic properties of composites. One of them is the Maxwell-Garnett model [12], which is simple and convenient for modeling due to its linearity. The MG model assumes that the mixing materials are in separated phases. Constituent particles are assumed to have much smaller dimensions than the wavelength of the incident radiation. However, they must be large enough to provide their own electromagnetic response. MG supposes a two-material mixture so that the particles of one material are dispersed in the continuous host of another material.

We consider the case of small spherical particles having a complex dielectric constant ε_i, which are embedded, at a filling fraction f, in a host medium having a real dielectric constant ε_h. The effective dielectric constant ε_{eff} of a suspension of small spherical particles embedded in a host medium of dielectric constant ε_h is given by the Clausius-Mossotti equation

$$\frac{\varepsilon_{eff}-\varepsilon_h}{\varepsilon_{eff}+2\varepsilon_h}=f\frac{\varepsilon_i-\varepsilon_h}{\varepsilon_i+2\varepsilon_h}\,. \tag{1}$$

Therefore, the MG effective dielectric function can be obtained through the relation

$$\varepsilon_{eff}=\varepsilon_h\frac{\varepsilon_i(1+2f)+2\varepsilon_h(1-f)}{\varepsilon_i(1-f)+\varepsilon_h(2+f)}\,. \tag{2}$$

The MG model includes interactions between the particles only through the Lorentz field, which limits its use to only small filling factors [13]. It usually describes an isotropic matrix containing spherical inclusions that are isolated from each other, such as the metal particles dispersed in a surrounding host matrix.

The quasi-static extension of the Maxwell-Garnett formula, also known as the extended Maxwell-Garnett formula, can be obtained by the Mie theory [14,15]. An incident plane wave, represented by the magnetic field $H_{inc}=H_0\exp(ik_0z)\hat{y}$ and $k_0=w/c$, is incident on a single isolated sphere of radius r_0 and relative permittivity $\varepsilon_i=n^2$. The scattered magnetic field can be decomposed into multipole terms; the proportionality constant of the 2^m-pole term is

$$b_m=\frac{\psi_m(nx)\psi'_m(x)-n\psi_m(x)\psi'_m(nx)}{\psi_m(nx)\xi'_m(x)-n\xi_m(x)\psi'_m(nx)} \tag{3}$$

and the 2^m-pole coefficients of the scattered electric field are

$$a_m=\frac{n\psi_m(nx)\psi'_m(x)-\psi_m(x)\psi'_m(nx)}{n\psi_m(nx)\xi'_m(x)-\xi_m(x)\psi'_m(nx)}\,. \tag{4}$$

Here $x=k_0r_0$, and ψ_n and ξ_n are the Riccati-Bessel functions, and the primes indicate differentiation with respect to the argument. b_m and a_m are also known as Mie coefficients. $m=1$ corresponds to the dipole oscillation while $m=2$ is associated with the quadrupole oscillation and so on. Only the b_1 coefficient, which is the strength of the magnetic dipole response, will be of interest when considering the effective permeability; only the a_1 term will be needed to find the effective permittivity. This yields the following extended MG formula:

$$\varepsilon_{eff}=\frac{k_0^3+4i\pi Na_1}{k_0^3-2i\pi Na_1}\,, \tag{5}$$

where N is the volume density of the dipoles. The filling fraction f of the composite is $f=4\pi Nr_0^3/3$, and should be kept to modest values. The optical properties of a suspension of small spheres may depend not only on the

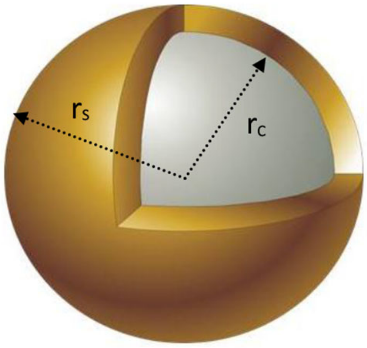

Fig. 1. A coated sphere.

effective dielectric constant ε_{eff}, but also on the effective relative magnetic permeability μ_{eff}. The latter quantity can be different from unity, even when the inclusion and host materials are non-magnetic. This happens when the Mie coefficient b_1, which represents the magnetic dipole term, is not negligible [16]. In analogy with the derivation of (5), the effective permeability can be shown to be given by

$$\mu_{eff} = \frac{k_0^3 + 4i\pi N b_1}{k_0^3 - 2i\pi N b_1}\,. \tag{6}$$

The effective refractive index $n_{eff} = \sqrt{\varepsilon_{eff}\mu_{eff}}$ can be substituted in the Snell law to calculate the refraction of incident waves.

Now we consider the scattering by a homogeneous sphere coated with a homogeneous layer of uniform thickness [17]. Suppose that the electromagnetic wave is incident on a coated sphere with inner radius r_C and outer radius r_S (fig. 1). The appropriate Mie coefficients are (n_C and n_S are the refractive indices of the core and coating)

$$b_m = \frac{n_S\psi_m(y)[\psi'_m(n_S y) - B_m\chi'_m(n_S y)] - \psi'_m(y)[\psi_m(n_S y) - B_m\chi_m(n_S y)]}{n_S\xi_m(y)[\psi'_m(n_S y) - B_m\chi'_m(n_S y)] - \xi'_m(y)[\psi_m(n_S y) - B_m\chi_m(n_S y)]}\,, \tag{7}$$

$$B_m = \frac{n_S\psi_m(n_C x)\psi'_m(n_S x) - n_C\psi_m(n_S x)\psi'_m(n_C x)}{n_S\chi'_m(n_S x)\psi_m(n_C x) - n_C\psi'_m(n_C x)\chi_m(n_S x)}\,, \tag{8}$$

$$a_m = \frac{\psi_m(y)[\psi'_m(n_S y) - A_m\chi'_m(n_S y)] - n_S\psi'_m(y)[\psi_m(n_S y) - A_m\chi_m(n_S y)]}{\xi_m(y)[\psi'_m(n_S y) - A_m\chi'_m(n_S y)] - n_S\xi'_m(y)[\psi_m(n_S y) - A_m\chi_m(n_S y)]}\,, \tag{9}$$

$$A_m = \frac{n_S\psi_m(n_S x)\psi'_m(n_C x) - n_C\psi'_m(n_S x)\psi_m(n_C x)}{n_S\chi_m(n_S x)\psi'_m(n_C x) - n_C\chi'_m(n_S x)\psi_m(n_C x)}\,, \tag{10}$$

where $x = k_0 r_C$, $y = k_0 r_S$, $\chi_m(z) = -zy_m(z)$ and $y_m(z)$ is the spherical Bessel function of the second kind. To find the effective media values for coated spheres, we simply substitute these equations into eqs. (5) and (6).

The effective permittivity, ε_{eff}, of a composite comprising two different kinds of solid spheres embedded in a matrix (binary composite) is given by

$$\sum_{j=A,B} C_j \frac{\varepsilon_h - \varepsilon_{eff} - \frac{3i}{2x_j^3}a_{1,j}f_{AB}(2\varepsilon_h + \varepsilon_{eff})}{\varepsilon_h + 2\varepsilon_{eff} - \frac{3i}{x_j^3}a_{1,j}f_{AB}(\varepsilon_h - \varepsilon_{eff})} = 0 \tag{11}$$

and the permeability, μ_{eff} by

$$\sum_{j=A,B} C_j \frac{\mu_h - \mu_{eff} - \frac{3i}{2x_j^3}b_{1,j}f_{AB}(2\mu_h + \mu_{eff})}{\mu_h + 2\mu_{eff} - \frac{3i}{x_j^3}b_{1,j}f_{AB}(\mu_h - \mu_{eff})} = 0, \tag{12}$$

where f_{AB} is the total filling factor of the spheres and the respective C_A and $C_B = 1 - C_A$ are the concentrations for the spheres A and B. In deriving (11) and (12) it has been assumed that the respective random unit-cell volumes of spheres A and B are proportional to the A and B sphere radii. Equations (11) and (12) provide a generalization of the earlier expressions for the effective permittivity and permeability, (5) and (6), to the two-sphere composite [18].

The above-mentioned effective medium theory can be used to study all the optical properties of the multiphase composites. We consider a binary composite structure that consists of two different types of coated nano-spheres which are embedded in a host medium. Such composites with effective parameters that obey the previous expressions can show a negative index of refraction at infrared frequencies which absolute value can be adjusted by controlling the filling fraction and size parameters.

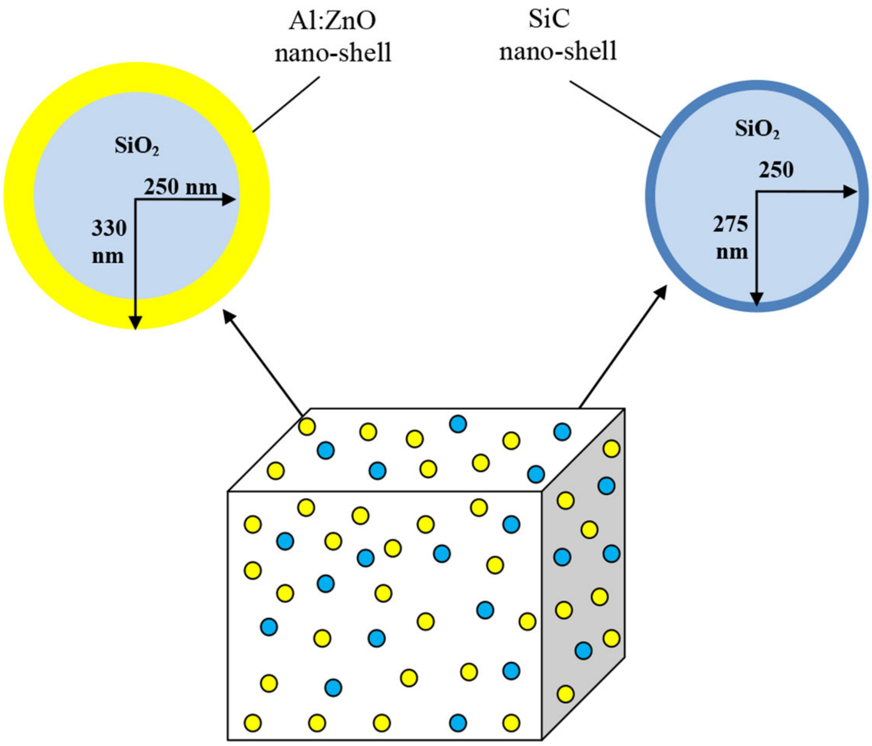

Fig. 2. Modeling of a typical binary composite with two types of coated nano-spheres.

3 Composite of coated spheres

3.1 Negative index of refraction

Recent progresses in production methods have made almost possible the realization of artificial metamaterials with remarkable properties that are not observed in natural materials. The usual metamaterial with negative refractive index (NRI), is a combination of a sublattice of SRRs (exhibits negative permeability) with a sublattice of thin metallic wires (possesses negative permittivity). This and similar forms of metamaterials have shown negative refractive index in the microwave regime [19,20].

SRRs have a complicated shape that makes it difficult to scale down to nanometer sizes. Therefore SRRs and wires cannot operate well in optical and infrared regions. Theoretical suggestions have been made during the past years for metamaterials with a strong magnetic response that consist of a simpler scattering object than SRRs, such as two-dimensional arrays of ferroeletric [21] or polaritonic [22] cylinders and also three-dimensional arrays of spheres [9, 23]. In order to achieve a (NRI) one has to combine the three-dimensional (3D) magnetic arrays of spheres with a lattice of plasmonic spheres (which provide negative ε) [24,26]. Yannopapas has reported NRI in a superlattice that consists of semiconductor spheres coated with a metal shell in the UV regime [27,28]. In this case the core of the coated spheres is responsible for magnetic activity while the required electric response is induced in superlattice by metal shell.

In this paper we are particularly interested in designing a negative-index metamaterial with strong electric and magnetic responses in infrared regions. For this purpose, we consider a case of binary composite materials consisting of coated nano-spheres. The respective binary composite is a 3D array of two different types of non-overlapping coated nano-spheres. The core of all nano-spheres are the same but they are covered by different nano-shells; in one type the nano-shell made from a semiconductor (ionic) material and in the other type the nano-shell made from a Drude-like or plasmonic material. Consider that the chosen materials as well as geometric parameters can supply a desired metamaterial. Figure 2 shows a schematic of the binary composite which consists of two types of coated nano-spheres. The core material is assumed to be SiO_2 with the refractive index of 1.45 and a radius of about 250 nm.

We choose silicon carbide (SiC) as the semiconductor nano-shell. SiC is the only group IV compound semiconductor, and it has more than 200 polytypes. The wide band gap makes SiC a very attractive semiconductor to make devices for applications in high-power, high-frequency and high-temperature environment. During the past decade, SiC is also becoming more and more active in optoelectronics thanks to the progress in materials growth and nano-fabrication [29].

Table 1. Sign of the real value of effective permittivity, permeability and refractive index in terms of frequency changes.

Binary composite	(0.995–0.9975)	(0.9975–0.9978)	(0.9978–0.9979)	(0.9979–0.9983)	(0.9983–1.000)	(1.000–1.002)
$(SiO_2@SiC)/$ $(SiO_2@AZO)$	$\mathrm{Re}(\varepsilon_{eff}) > 0$	$\mathrm{Re}(\varepsilon_{eff}) < 0$	$\mathrm{Re}(\varepsilon_{eff}) < 0$	$\mathrm{Re}(\varepsilon_{eff}) < 0$	$\mathrm{Re}(\varepsilon_{eff}) < 0$	$\mathrm{Re}(\varepsilon_{eff}) < 0$
	$\mathrm{Re}(\mu_{eff}) > 0$	$\mathrm{Re}(\mu_{eff}) > 0$	$\mathrm{Re}(\mu_{eff}) > 0$	$\mathrm{Re}(\mu_{eff}) < 0$	$\mathrm{Re}(\mu_{eff}) > 0$	$\mathrm{Re}(\mu_{eff}) > 0$
	$\mathrm{Re}(n_{eff}) > 0$	$\mathrm{Re}(n_{eff}) > 0$	$\mathrm{Re}(n_{eff}) < 0$	$\mathrm{Re}(n_{eff}) < 0$	$\mathrm{Re}(n_{eff}) < 0$	$\mathrm{Re}(n_{eff}) > 0$

Since many of ionic materials, like SiC, exhibit phonon-polariton excitations within the near-infrared regime, they would be very good choices for these nano-structures. Such materials are usually optically active ionic crystals that support phonon-polaritons, *i.e.*, simultaneously excited modes of the material polarization and the EM field. Usually, the phonon-polaritons are excited when infrared radiation impinges on such a material inducing coupled oscillations of the polarization and the EM field. The dielectric function in such materials is provided by the Drude-Lorentz model [30]

$$\varepsilon(\omega) = \varepsilon_\infty \left(1 + \frac{\omega_L^2 - \omega_T^2}{\omega_T^2 - \omega^2 - i\omega\gamma}\right), \tag{13}$$

where ε_∞ is the asymptotic value of the dielectric permittivity at high frequencies, γ is the loss factor, and the respective ω_T and ω_L are the transverse and longitudinal optical phonon frequencies. For simplicity, the damping constant will be neglected for this study.

We take silicon carbide (β-SiC) with a zinc blende crystal structure whose permittivity is given by (13) having $\omega_T = 23.8 \times 10^{12}$ Hz, $\omega_L = 29.06 \times 10^{12}$ Hz, and $\varepsilon_\infty = 6.51$. As mentioned above, losses are ignored in ionic materials therefore we set $\gamma = 0$ for SiC. The radius of the nano-spheres is $R_{SiC} = 275$ nm,the thickness of the nano-shell is 25 nm and the volume filling fraction occupied by this type of nano-spheres is $f_{SiC} = 0.21$.

Previously mentioned that, in order to have negative refractive index, what is required is a collection of nano-spheres with negative ε_{eff} within all or part of the negative μ_{eff} region. Such nano-spheres would have plasmonic nano-shells whose EM response can be modeled in the infrared regime by a Drude-type electric permittivity

$$\varepsilon(\omega) = 1 - \frac{\omega_p^2}{\omega^2 + i\gamma\omega}, \tag{14}$$

where ω_p is the bulk plasma frequency and γ is the loss factor. Metals are the most familiar candidate for the plasmonic materials because metals tend to have large plasma frequencies and high electrical conductivity; they have traditionally been the materials of choice for plasmonics. One of the important disadvantages of conventional metals is high loss. Conventional metals have very high carrier concentrations, which in turn makes their plasma frequencies very large. A large plasma frequency produces a large imaginary permittivity, which translates to large loss. Alternative plasmonic materials have lower carrier concentrations and, hence, smaller losses.

Recently, we have presented a new set of nano-structured composites which can exhibit a phenomenon known as surface plasmon resonance in a broad frequency range from the deep infrared to the terahertz region. We have proposed a doped semiconductor (n-type Ge) as a plasmonic material and the effective permittivity and refractive index of ZnS(ZnO)/Ge and $TiO_2(SiO_2)$/Ge composites have been calculated over terahertz frequencies [31,32]. Transparent conducting oxides (TCOs) such as Al:ZnO, Ga:ZnO and indium-tin-oxide (ITO) are other alternatives to noble metals which exhibit losses nearly five times smaller than that of the best metal (Ag) in the near-IR [33]. In this letter, we choose aluminum-doped zinc oxide (Al:ZnO, herein referred to as AZO) as a plasmonic nano-shell whose permittivity is described by (14) with $\omega_p = 258 \times 10^{12}$ Hz. Similar to SiC, the losses are not included for AZO permittivity and we use $\gamma = 0$. The radius of the nano-spheres is $R_{AZO} = 330$ nm, the thickness of the nano-shell is 80 nm and the volume filling fraction occupied by this type of nano-spheres is $f_{AZO} = 0.37$.

The full calculations of the effective permittivity, permeability and refractive index of the mentioned binary composite (SiO_2@SiC/SiO_2@AZO) are shown in fig. 3. For this purpose, we use eqs. (11) and (12) with $C_{SiC} = C_{AZO} = 0.5$ and the resulting complex index of refraction is given by the branch of the square root ($n_{eff} = \sqrt{\varepsilon_{eff}\mu_{eff}}$) that yields a non-negative imaginary part of n_{eff} along the positive real frequency axis.

It is evident that all the effective medium parameters exhibit strong resonant behaviour which principally stems from the resonances of the electric (magnetic)-dipole components of coated nano-spheres. In fig. 3(a) one identifies a small area where all $\mathrm{Re}(\varepsilon_{eff})$, $\mathrm{Re}(\mu_{eff})$ and $\mathrm{Re}(n_{eff})$ become negative within the same, more or less, spectral region. AZO coating is designed to have the electric-dipole resonance on its outer surface while SiC coating provides the magnetic one. However, the evanescent field within the coating can tunnel into the SiO_2 cores, which improve the resonance conditions leading to a negative refractive index metamaterial. Signs of real values of effective permittivity, permeability and refractive index for the binary composite, in terms of frequency changes, are given in table 1. It is obvious that from $\omega/\omega_T = 0.9979$ to $\omega/\omega_T = 0.9983$ all the effective parameters assume negative values so the negative-index metamaterial is evident. It is worth noting that $\mathrm{Re}(\varepsilon_{eff})$ and $\mathrm{Re}(\mu_{eff})$ are not necessarily both negative within the negative refractive index region due to the imaginary part of ε_{eff} and μ_{eff} (*e.g.*, $\omega/\omega_T = 0.9983$ to $\omega/\omega_T = 1.000$).

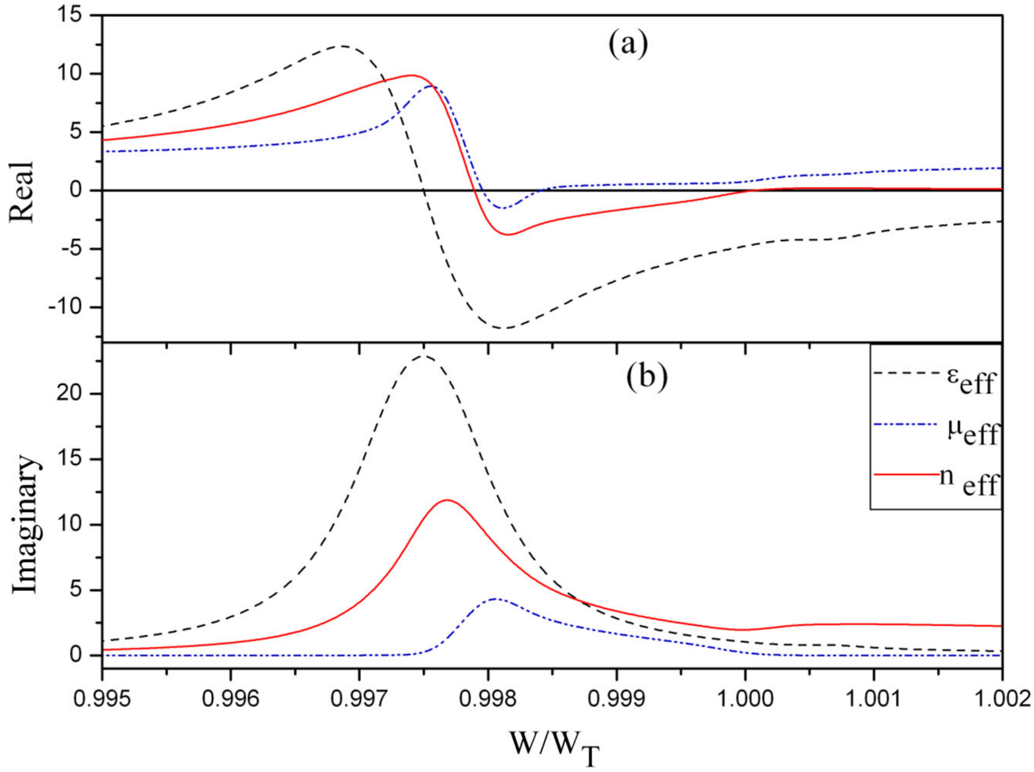

Fig. 3. (a) Real and (b) imaginary parts of ε_{eff}, μ_{eff} and n_{eff} for the binary composite (SiO_2@SiC/SiO_2@AZO) predicted by the extended Maxwell-Garnett theory.

Within the negative-index frequency region (fig. 3(b)), the imaginary parts of ε_{eff}, μ_{eff} and n_{eff} are also significant. The imaginary part of the effective index of refraction is proportional to attenuation and has moderate values in this range. The presence of loss is an inevitable consequence of the underlying resonances, but the losses are smaller at frequencies away from the center of the lines. Finally, note that the imaginary parts of both the calculated effective permittivity and permeability shown in fig. 3(b) are always positive. This indicates attenuation, either due to material losses or reactive energy storage; the structure is definitely passive.

3.2 Band calculation and verification

Classical wave propagation in periodic composite materials has attracted a lot of attention in recent decades. The study of photonic crystals with the theoretical prediction and experimental realization has taken the lead. Photonic crystals are composite materials with a periodic dielectric function that have frequency regions, known as photonic gaps, over which there can be no propagation of light in the crystal. A number of methods are suggested for the calculation of frequency band structure of photonic crystals as well as the transmission/reflection coefficient of light incident on a slab of the photonic crystal. The results of the effective medium theory (previous section) can be verified by a comparison with an accurate multiple-scattering method. To this end, we use the code MULTEM 2 to perform the band calculations of photonic crystal [34]. This program finds the complex band structure and also transmission of a three-dimensional photonic crystal which consists of non-overlapping spheres in a homogeneous medium through a scattering matrix technique. This computer program is extended in some aspects to allow one to treat coated spheres as scatterers; the individual sphere may consist of a number of homogeneous spherical shells.

A plane electromagnetic wave with wave vector k is incident on a plane of non-overlapping spheres (fig. 4). The electric-field component of the incident wave is defined by

$$E_{in}(r) = E_0 \exp(ik \cdot r). \tag{15}$$

The transmitted and reflected waves of the plane of spheres (in compact notation) are given by

$$\begin{aligned} E^{+}_{tr} &= T \cdot E^{+}_{in}, \\ E^{-}_{rf} &= R \cdot E^{+}_{in}, \end{aligned} \tag{16}$$

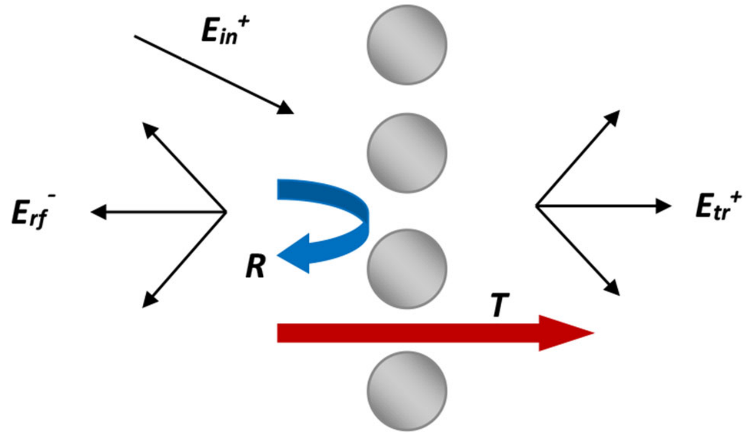

Fig. 4. A plane electromagnetic wave is scattered from a plane of non-overlapping spheres.

where T and R are transmission and reflection matrices and include all of the information about the host dielectric, sphere (core and shells) dielectric, lattice, sphere (core and shells) size and frequency. Then by using Bloch's theorem and imposing periodic boundary conditions we can also obtain the dispersion relation which determines the frequency band structure.

In order to verify the effective medium approach we consider a simple cubic lattice that consists of non-overlapping coated nano-spheres. The lattice constant is $a = 1\,\mu$m, and the (SiO_2@SiC) nano-spheres have a radius of $r_{SiC} = 0.275a$ while for the (SiO_2@AZO) nano-spheres the radius is $r_{AZO} = 0.33a$.

The band structures are calculated with the scattering matrix code for the wave vectors parallel to the ΓX, ΓM and ΓR lines (fig. 5(b)). The symmetric shape of the band structures around the origin is clear. The comparison in fig. 5(b) indicates that the modes are mostly overlapped and only at very low frequencies the curves partly deviate. This can verify the isotropic behavior of this nano-composite. The real (solid line) and imaginary (dotted line) parts of n_{eff} for the binary composite (SiO_2@SiC/SiO_2@AZO) which is described in the previous section, are calculated by the extended Maxwell-Garnett theory and are shown in fig. 5(a). It is obvious that the band structure predicts a wide photonic band gap from $\omega/\omega_T = 0.98$ to $\omega/\omega_T = 1.31$. This photonic gap nearly corresponds to the frequency region over which n_{eff} is negative, *i.e.*, from $\omega/\omega_T = 0.9978$ to $\omega/\omega_T = 1.0$. In this region the wave vector $k = \omega n_{eff}/c < 0$ leading to a negative phase velocity $\upsilon_P = \omega/k < 0$. This shows that backward waves which are defined by $\upsilon_P < 0$ are found in this range.

The negative index of the refraction region overlaps the backward wave region. This effect is a consequence of the negative-index metamaterial. Moreover, this result can guarantee the accuracy of the EMG theory.

We expect to observe a considerably weakened transmission of light within the band gap region. Figure 6(b) shows the transmittance of light on a slab of the crystal consisting of (SiO_2@SiC) and (SiO_2@AZO) coated nano-spheres as obtained by the multiple-scattering method. The transmission calculation is done for a transverse (S: the electric field parallel to the surface of the slab) incident wave. We identify a main gap where transmittance is significantly suppressed. In fig. 6(a) we show the real part of the effective refractive index n_{eff} for the binary composite (SiO_2@SiC/SiO_2@AZO) as obtained from the extended Maxwell-Garnett theory. It is clear that, within the frequency region where n_{eff} is negative, the corresponding transmittance given by the multiple-scattering approach is greatly decreased, a fact which, again, verifies the validity of the EMG theory. The real part of the effective refractive index assumes values from $\mathrm{Re}(n_{eff}) = -5.26$ at $\omega/\omega_T = 0.9979$ to $\mathrm{Re}(n_{eff}) = 10.25$ at $\omega/\omega_T = 0.9976$. We observe that, again, the region where the real part of n_{eff} is negative correlates very well with the gap of the transmittance curve. From this, we can infer that the extended Maxwell-Garnett prediction of negative n_{eff} is verified by the multiple-scattering technique. In other words, the predictions of the extended Maxwell-Garnett theory are in good agreement with those of an accurate multiple-scattering approach.

4 Summary

We have presented a metamaterial composed of two different types of coated nano-spheres which has a negative index of refraction at infrared frequencies. The respective metamaterial is a binary composite of (SiO_2@SiC) and (SiO_2@AZO) coated nano-spheres. Effective medium theory was applied to predict the effective medium parameters of the composite. The results of the negative effective index of refraction have been verified by the multiple-scattering calculations of frequency band structure and also transmission of a 3D photonic crystal that consists of the mentioned coated nano-spheres. The structure presented here is simple and more practical than SRRs. Our results also provided useful information for researchers studying NRI metamaterials.

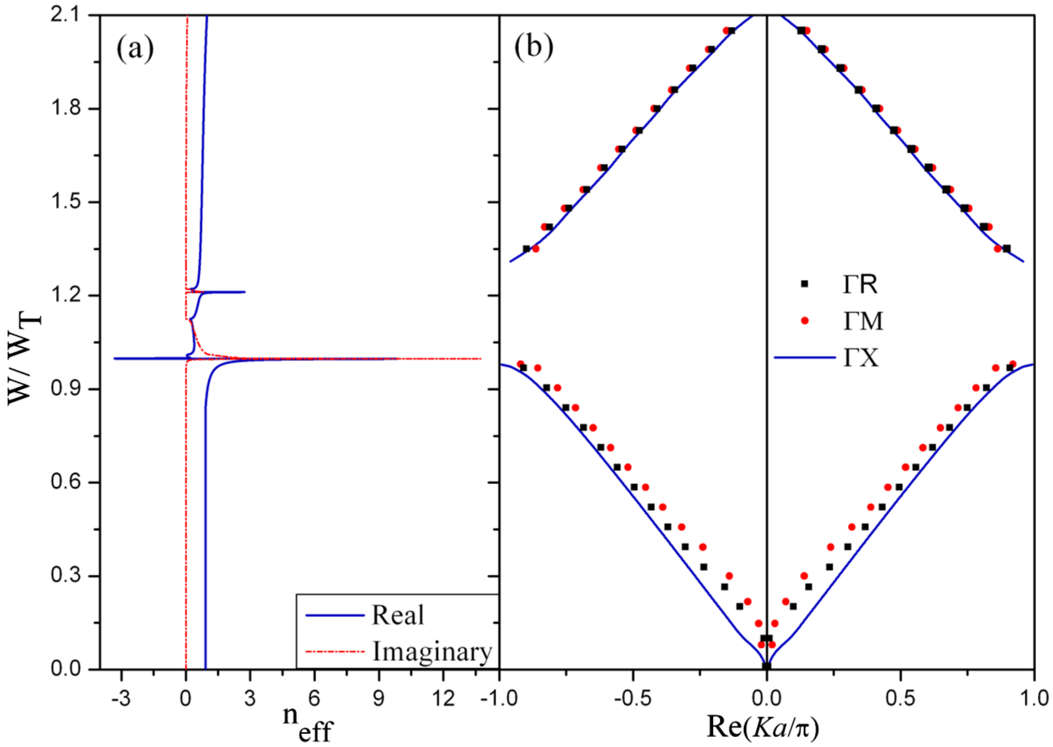

Fig. 5. (a) The real (solid line) and imaginary (dotted line) parts of n_{eff} for the binary composite (SiO_2@SiC/SiO_2@AZO) predicted by the extended Maxwell-Garnett theory and (b)The real part of the frequency band structure of a simple cubic lattice of the (SiO_2@SiC) and (SiO_2@AZO) coated nano-spheres with lattice constant $a = 1\,\mu$m. The solid line and different symbols demonstrate the results of band calculations for various values of the wave vectors.

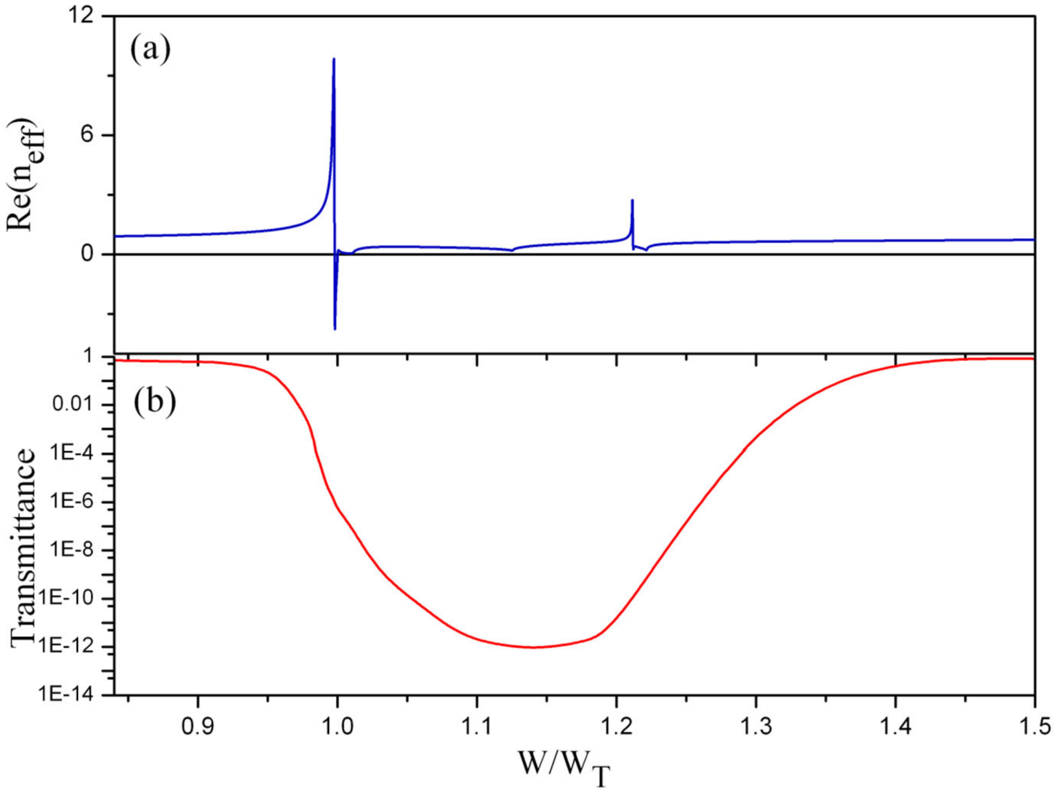

Fig. 6. (a) The real part of n_{eff} for the binary composite (SiO_2@SiC/SiO_2@AZO) predicted by the extended Maxwell-Garnett theory and (b) transmittance curve for light incident normally on a slab of the crystal consisting of (SiO_2@SiC) and (SiO_2@AZO) coated nano-spheres as calculated by the multiple-scattering method.

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References

1. V.G. Veselago, Sov. Phys. Usp. **10**, 509 (1968).
2. R.A. Silin, Opt. Spectrosc. **44**, 109 (1978).
3. J.B. Pendry, Contemp. Phys. **45**, 191 (2004).
4. D.R. Smith, W.J. Padilla, D.C. Vier, S.C. Nemat-Nasser, S. Schultz, Phys. Rev. Lett. **84**, 4184 (2000).
5. R.A. Shelby, D.R. Smith, S. Schultz, Science **292**, 77 (2001).
6. J.B. Pendry, A.J. Holden, W.J. Stewart, I. Youngs, Phys. Rev. Lett. **76**, 4773 (1996).
7. J.B. Pendry, A.J. Holden, D.J. Robbins, W.J. Stewart, IEEE Trans. Microwave Theory Tech. **47**, 2075 (1999).
8. T.J. Yen, W.J. Padilla, N. Fang, D.C. Vier, D.R. Smith, J.B. Pendry, D.N. Basov, X. Zhang, Science **303**, 1494 (2004).
9. V. Yannopapas, A. Moroz, J. Phys.: Condens. Matter **17**, 3717 (2005).
10. V.M. Shalaev, *Nonlinear Optics of Random Media: Fractal Composites and Metal- Dielectric Films* (Springer, Berlin, 2000).
11. S. Bosch, J. Ferré-Borrull, N. Leinfellner, A. Canillas, Surf. Sci. **453**, 9 (2000).
12. J.C. Maxwell-Garnett, Philos. Trans. R. Soc. London, Ser. A **203**, 385 (1904).
13. C.G. Granqvist, O. Hunderi, Phys. Rev. B **18**, 2897 (1978).
14. W.T. Doyle, Phys. Rev. B **39**, 9852 (1989).
15. R. Ruppin, Opt. Commun. **182**, 273 (2000).
16. C. Bohren, J. Atmos. Sci. **43**, 468 (1986).
17. C.F. Bohren, D.R. Huffman, *Absorption and Scattering of Light by Small Particles* (Wiley-Interscience, New York, 1983).
18. R. Luo, Appl. Opt. **36**, 8153 (1997).
19. C.G. Parazzoli, R.B. Greegor, K. Li, B.E.C. Koltenbah, M. Tanielian, Phys. Rev. Lett. **90**, 107401 (2003).
20. A.A. Houck, J.B. Brock, I.L. Chuang, Phys. Rev. Lett. **90**, 137401 (2003).
21. S. O'Brien, J.B. Pendry, J. Phys.: Condens. Matter **14**, 4035 (2002).
22. K.C. Huang, M.L. Povinelli, J.D. Joannopoulos, Appl. Phys. Lett. **85**, 543 (2004).
23. M.S. Wheeler, J.S. Aitchison, M. Mojahedi, Phys. Rev. B **72**, 193103 (2005).
24. V. Yannopapas, Phys. Rev. B **75**, 035112 (2007).
25. V. Yannopapas, Appl. Phys. A **87**, 259 (2007).
26. V. Yannopapas, Sol. State Commun. **204**, 51 (2015).
27. V. Yannopapas, Phys. Status Solid **1**, 208 (2007).
28. V. Yannopapas, J. Phys.: Condens. Matter **20**, 255201 (2008).
29. H. Ou, Y.Ou, A. Argyraki, S. Schimmel, M. Kaiser, P. Wellmann, M.K. Linnarsson, V. Jokubavicius, J. Sun, R. Liljedahl, M. Syväjärvi, Eur. Phys. J. B **87**, 58 (2014).
30. H. Ibach, H. Luth, *Solid-State Physics* (Springer, Berlin, 2003).
31. H. Sadeghi, A. Zolanvar, A. Ranjgar, R. Norouzi, Plasmonics **9**, 327 (2014).
32. H. Sadeghi, A. Zolanvar, A. Ranjgar, R. Norouzi, J. Electron. Mater. **43**, 4294 (2014).
33. G.V. Naik, J. Kim, A. Boltasseva, Opt. Mater. Express **1**, 1090 (2011).
34. N. Stefanou, V. Yannopapas, A. Modinos, Comput. Phys. Commun. **132**, 189 (2000).