

# Isotropic magnetic response of silicon carbide microspheres at mid-infrared wavelength

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## ABSTRACT

Base on the Lorentz–Lorenz formula, magnetic response in metamaterial composed of randomly dispersive SiC microspheres is demonstrated theoretically around 13  $\mu\text{m}$  mid-infrared wavelength. By adjusting the microspheres volume filling fraction the permeabilities both less than one and more than one can be obtained. Through parameters optimization, isotropic negative permeability is represented. Electromagnetic simulation shows the mechanism for magnetic effects and negative permeability in SiC microspheres.

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## 1. Introduction

There has been enormous interest in exploiting novel electromagnetic properties in the artificial engineered materials which were composed of subwavelength units arranging regularly or randomly. Magnetic effects are difficult to be exploited at optical frequency due to the intrinsic limitations of natural optical materials whose magnetic response is very weak. As a necessary composite part of negative refractive index metamaterial [1], negative permeability is urgently required in the optical frequency due to the fact negative permittivity can be obtained in metal material. Up to now, magnetic resonance response and even negative permeability have been realized experimentally in split resonant rings (SRRs) or similar structures at microwave [2–6] and decreased size SRRs and metal–dielectric voids at optical frequency [7–13]. However, the structures are very complex to design and are confined to two dimensions; moreover, the fabrication process is very hard due to complicated craft and expensive top-down physical approaches such as electron beam lithography which is fairly difficult to obtain large samples.

The permittivity and the permeability with spatial tailored properties provide a scheme to design invisible cloaking devices based on transformation optics and conformal mapping [14,15]. At visible and infrared frequency, spatial variational permittivities could be realized using the anisotropic dielectrics composite

material. The polarized light realizing invisibility is not depended on the permeability due to the lack of magnetic effects [16–18]. Tailored permeabilities are required necessarily to future designs of invisible device for all directional polarized light.

Recently, magnetic resonance and negative permeability at high frequency which arises from Mie resonance of dielectric spheres with large permittivity was proposed theoretically [19,20]. Experiments have demonstrated negative permeability effect in the composite structure with ceramic spheres embedded conducting wire frame at microwave frequency [21,22] and nonmagnetic rods made of an ferroelectric  $\text{SrTiO}_3$  in Terahertz frequency [23].

In this paper, silicon carbide (SiC) is chosen to investigate magnetic effects in the material composed of randomly dispersed SiC microspheres nearby 13  $\mu\text{m}$  mid-infrared wavelength range. Firstly, effective permeabilities less than one are achieved for small volume filling fraction. Then, continuous varying permeabilities are realized through adjusting volume filling fraction at 13  $\mu\text{m}$ . Finally, negative permeability around 13  $\mu\text{m}$  is achieved through larger volume fraction. Due to the random arrangement of microspheres, three-dimensional large area sample can be obtained easily for proposed magnetic metamaterial.

## 2. Theory

When a plane wave with wavelength  $\lambda$  is incident to the single dielectric sphere with radius  $R$  and the long wavelength limit condition ( $\lambda \gg R$ ) is satisfied, the scattering electromagnetic field

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is equivalent to a radiated dipole. The effective magnetic polarizability  $\alpha_m$  and effective electric polarizability  $\alpha_e$  are derived from Ref. [24]

$$\begin{aligned}\alpha_m &= 6\pi b_1 / k_h^3 \\ \alpha_e &= 6\pi a_1 / k_h^3\end{aligned}\quad (1)$$

where  $k_h = 2\pi n_h / \lambda$  is the wave number in ambient medium (air in this work  $n_h = 1$ ). The first order Mie scattering coefficients  $b_1$  and  $a_1$  which correspond to the strength of magnetic and electric dipole response are given as follows [25]:

$$\begin{aligned}b_m &= \frac{\psi_m(nx)\psi'_m(x) - n\psi_m(x)\psi'_m(nx)}{\psi_m(nx)\xi'_m(x) - n\xi_m(x)\psi'_m(nx)} \\ a_m &= \frac{n\psi_m(nx)\psi'_m(x) - \psi_m(x)\psi'_m(nx)}{n\psi_m(nx)\xi'_m(x) - \xi_m(x)\psi'_m(nx)}\end{aligned}\quad (2)$$

where  $\psi_m$  and  $\xi_m$  are the Riccati–Bessel functions and  $m$  is the index term. The primes indicate differentiation with respect to the argument. The relative refractive index  $n$  is the ratio of the refractive index of dielectric sphere to the refractive index of the ambient medium. The size parameter  $x$  is  $k_h R$ .

The efficiency factor  $Q_{sca}$  of scattering cross section has following forms [25]

$$Q_{sca} = \frac{2}{x^2} \sum_{m=1}^{\infty} (2m+1)(|a_m|^2 + |b_m|^2) \quad (3)$$

In the long wavelength condition, the first order term dominates the scattering effect, so, high order terms can be ignored in the calculations. The efficiency factor is simplified as follows:

$$Q_{sca} \approx \frac{6}{x^2} (|a_1|^2 + |b_1|^2) \quad (4)$$

Using the Lorentz–Lorenz formula which averages the response of electrical dipoles and magnetic dipoles in composite material composed of the collection of randomly dispersed subwavelength spheres, the effective permeability  $\mu_{eff}$  and effective permittivity  $\epsilon_{eff}$  can be described as follows [20]:

$$\mu_{eff} = \mu_h \frac{3 + 2N\alpha_m}{3 - N\alpha_m} \quad (5)$$

$$\epsilon_{eff} = \epsilon_h \frac{3 + 2N\alpha_e}{3 - N\alpha_e} \quad (6)$$

where  $\mu_h$  and  $\epsilon_h$  are the permeability and the permittivity of surrounding material respectively, for the air they are  $\mu_h = \epsilon_h = 1$ .  $f$  is the volume filling fraction and  $N$  is the volume density of spheres ( $N = 3f/4\pi R^3$ ).

### 3. Results and analysis

At mid-infrared wavelength, the permittivity of SiC exhibits a sharp resonance near  $12.6 \mu\text{m}$  due to excitation of transverse optical phonons. At the low frequency side of this resonance, the permittivity is large with moderate damping. Under this condition, large permittivity of spherical particles similar to resonant magnetic dipole can generate strong magnetic resonant effects at frequencies near phonon polaritonic resonance. The permittivity of SiC can be described as follows:

$$\epsilon_{SiC} = \epsilon_{\infty} \left[ 1 + \frac{\omega_L^2 - \omega_T^2}{\omega_T^2 - \omega^2 - i\Gamma\omega} \right] \quad (8)$$

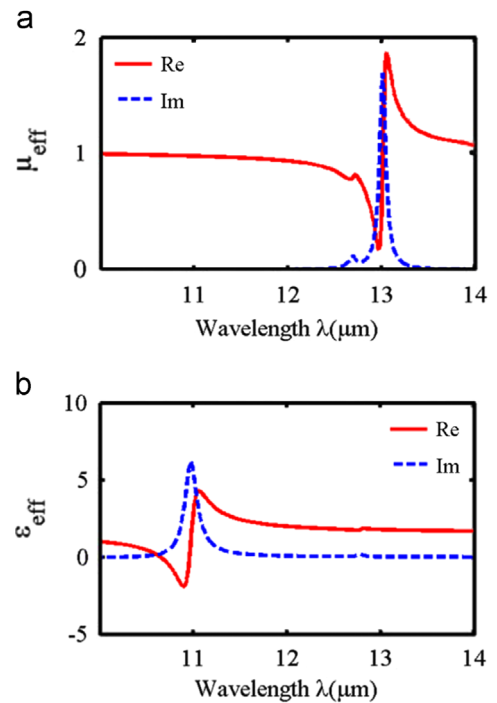
where  $\omega$  is the incident angular frequency,  $\omega_L = 2\pi \times 29.07 \text{ THz}$  is the longitudinal optical phonon frequency,  $\omega_T = 2\pi \times 23.79 \text{ THz}$  is the transverse optical phonon frequency,  $\Gamma = 2\pi \times 0.1428 \text{ THz}$  is the damping frequency, and  $\epsilon_{\infty} = 6.7$  is the permittivity at high-frequency limit. All above data are taken from Ref. [26].

#### 3.1. Magnetic resonance

Firstly, we consider the electromagnetic resonant effects of SiC microspheres. The calculated results are shown Fig. 1(a) shows that the effective permeability produces resonant peak around  $13 \mu\text{m}$ . The real part of  $\mu_{eff}$  is less than one that behaves diamagnetic properties. The minimal  $\text{Re}(\mu_{eff})$  is 0.2. In short wavelength side of resonant peak, material represents diamagnetic property corresponding to  $\text{Re}(\mu_{eff})$  less than one. The effective permittivities tend to a fixed value at the same wavelength range. In contrast to effective permeability, the peak for effective permittivity yields resonant peak around  $11 \mu\text{m}$ , as shown in Fig. 1(b). Negative permittivity with small value appears. For example,  $\text{Re}(\epsilon_{eff})$  is  $-1$  at  $10.8 \mu\text{m}$ , that provides an opportunity to design superlens at corresponding wavelength. The differences of peaks for effective permeability and effective permittivity arise from the distinct electric and magnetic scattering properties corresponding to different peak positions indicated in Fig. 2(a) and (b).

#### 3.2. Continuously varying permeability

The calculated effective permeabilities of SiC microspheres can be adjusted by changing the volume filling fraction  $f$  at  $13 \mu\text{m}$  as shown in Fig. 3. It is seen from Fig. 3(a) that the real part of  $\mu_{eff}$  for  $0.9 \mu\text{m}$  decreases from 1 to 0 as the  $f$  increases from 0 to 0.4. However, when  $R = 0.7 \mu\text{m}$ , the real part of  $\mu_{eff}$  increases from 0 to 2 as  $f$  increases from 0 to 0.4 as shown in Fig. 3(b). In natural existing material, due to the absence of magnetic responses, the permeability is the same as vacuum. Here, we demonstrate that, through choosing different microsphere radius, effective permeability either less than one or more than one can be achieved. Modest imaginary parts also appear which means the existence of definite energy dissipation. The wide-range tunable permeability will provide convenient approaches to design novel devices based on the theory of transformation optics.



**Fig. 1.** The real part and imaginary part for (a) effective permeability and (b) effective permittivity of randomly dispersed SiC microspheres. The radius is  $R = 0.8 \mu\text{m}$  and volume filling fraction is  $f = 0.2$ .

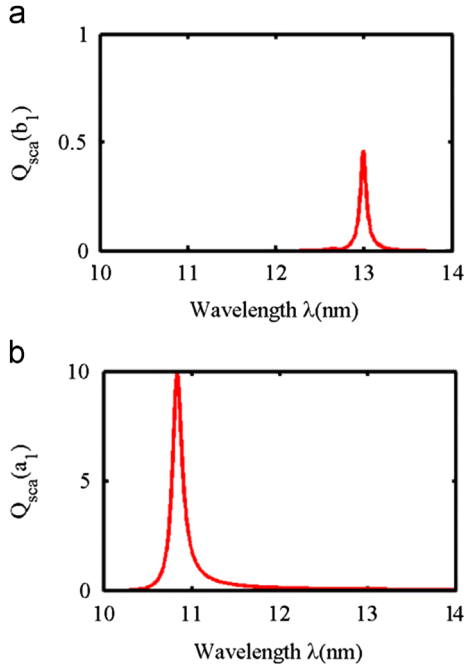


Fig. 2. (a) Magnetic efficiency factor  $Q_{sca}(b_1)$  and (b) electric efficiency factor  $Q_{sca}(a_1)$ .

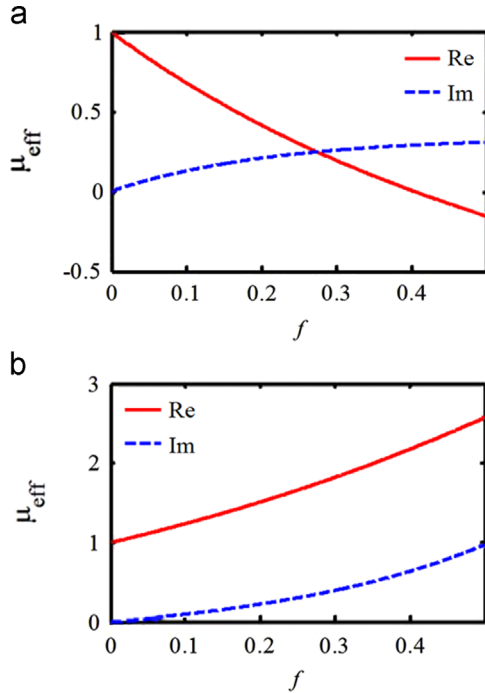


Fig. 3. Calculated effective permeabilities for (a)  $R=0.9 \mu\text{m}$  and (b)  $R=0.7 \mu\text{m}$  at  $13 \mu\text{m}$ .

### 3.3. Negative permeability

To optimize microspheres radius for magnetic resonance, the  $Q_{sca}$  is calculated as a function of radius  $R$  which is indicated in Fig. 4. We can see that  $Q_{sca}$  reaches maximum value when  $R$  approaches  $0.8 \mu\text{m}$ . Under these parameters (radius  $R=0.8 \mu\text{m}$  and volume filling fraction  $f=0.35$ ) we calculated effective permeability which is shown in Fig. 5. It shows that the maximal negative permeability  $-0.4$  is achieved at  $13 \mu\text{m}$ . The negative  $\text{Re}(\mu_{eff})$  mainly results from induced strong magnetic resonance of large permittivity of SiC microspheres. The resonant wavelength should

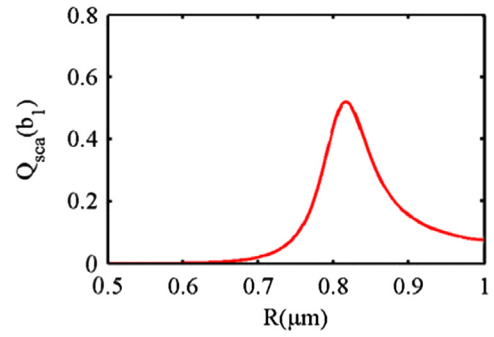


Fig. 4. The magnetic efficiency factor  $Q_{sca}(b_1)$  as the function of SiC microsphere radius  $R$ . The wavelength is  $13 \mu\text{m}$ .

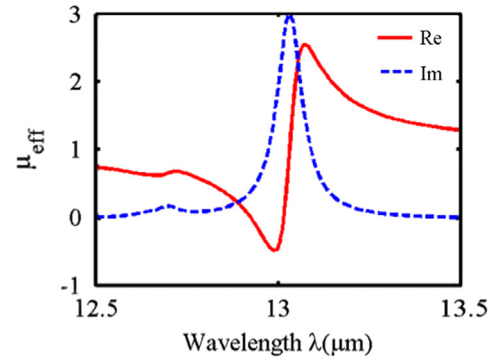
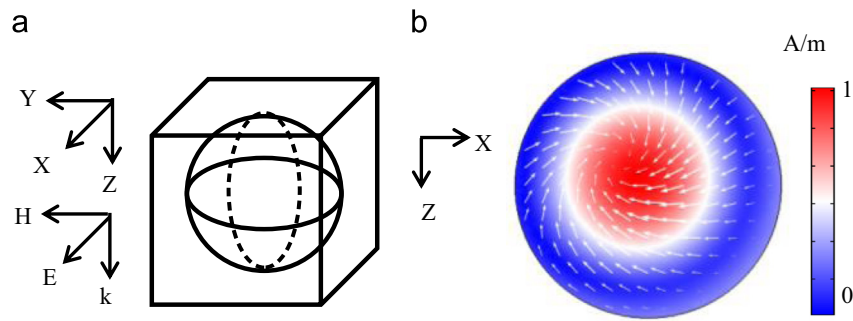


Fig. 5. Permeability for SiC sphere dispersed in air.

satisfy the formula  $\lambda_{res} = 2R\sqrt{\epsilon_{res}}$ , where  $\epsilon_{res}$  is the permittivity of SiC for resonant wavelength. The two peaks located at  $12.7 \mu\text{m}$  and  $13 \mu\text{m}$  agree well with the result calculated by resonant formula. Although the high frequency peak around  $12.7 \mu\text{m}$  agrees with resonant condition, the permeability is still positive which is because  $12.7 \mu\text{m}$  is close to the polaritonic resonance peak of SiC induced mainly by the inherent excitation of optical phonon of silicon carbide microspheres. The total effective dipole resonance from large permittivity of SiC is suppressed severely due to the combination of the polaritonic phonon resonance at the same wavelength. Consequently, negative permeability is only achieved at narrower wavelength range around  $13 \mu\text{m}$ .

For explaining the origination of diamagnetic and negative permeability in SiC spheres, electromagnetic simulation is performed using finite element method. As shown in Fig. 6(a), a three-dimensional cubic cell is selected and a plane electromagnetic wave at  $13 \mu\text{m}$  is normally incident to the cell. As shown in Fig. 6(b), incident magnetic field along  $y$  direction induces the closed displacement currents in SiC microsphere which produce the induced magnetic field along opposite- $Y$  direction. As a result, the strong magnetic dipole resonance is achieved which is just the origin of diamagnetic effects and negative permeability in SiC microspheres.

In the preceding sections, the Mie theory is used to deal with the scattering of spherical particles which is electrically neutral. J. Klaacka et al. extends Mie theory to deal with the electromagnetic scattering of charged spherical particles [27–29]. In this theory, parameter  $g$  is introduced to modify corresponding Mie scattering coefficients  $b_m$  and  $a_m$ . When  $g=0$ , scattering theory for charged particle will be equivalent to the Mie theory. For radius  $R=0.5\text{--}1 \mu\text{m}$  selected in this work we find that charged SiC spherical particles cannot influence the scattering properties compared with neutral SiC spherical particles. Because corresponding size parameter  $x$  is less than 1 and more than 0, the parameters  $g$  is approximate the order of  $10^{-4}$  which is very close



**Fig. 6.** (a) Basic cubic cell for electromagnetic simulation. Periodic boundary conditions are used along X and Y directions. Cell constant and SiC sphere radius are  $a=2\text{ }\mu\text{m}$  and  $R=0.8\text{ }\mu\text{m}$  respectively. The incident wavelength is  $13\text{ }\mu\text{m}$  and (b) Normalized magnetic field distribution in X–Z plane. White arrows denote the electric displacement direction.

to 0. In addition, Ref. [27,29] demonstrate that the scattering would be affected for surface charged particles when  $x < 0.01$  or  $x > 1$ . Consequently, scattering theory of charged particles is a more powerful tool that can be used to explore the electromagnetic response for smaller and larger size particles in infrared and visible range.

#### 4. Conclusions

In this work, we have investigate magnetic response in metamaterial composed of randomly dispersive silicon carbide microspheres in the mid-infrared wavelength range. The continuous varying permeabilities either less than one or more than one are achieved for different microsphere radius by adjusting the volume fraction. Through parameters optimization negative permeability is realized. The permeability less than one can be used to exploit the diamagnetic properties in artificial structured material. For specific wavelength, continuous varying permeabilities could be used in the designs of cloak devices base on transformation optics. Finally, isotropic negative permeability provides a feasible way for the application of magnetic plasmon and the realization of isotropic negative refractive index material. The magnetic response in random silicon carbide micropowder has been demonstrated experimentally at mid-infrared wavelength [30]. Furthermore, we found that the silicon carbide microspheres can be fabricated by the sol–gel method [31]. So, the permeability in silicon carbide microspheres either less than one or negative demonstrated in this paper will be investigated experimentally in the next step of work.

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