Revisit of generalized Kerker’s conditions using composite metamaterials

Rfaqat Ali1,2 *

1 Instituto de Física, Universidade Federal do Rio de Janeiro, Rio de Janeiro, RJ 21941-972, Brazil
2 Photonics Research Center, Applied Physics Department, Gleb Wataghin Physics Institute, P.O. Box 6165, University of Campinas - UNICAMP, 13083-970 Campinas, SP, Brazil

E-mail: r.ali@if.ufrj.br

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Abstract
Achieving zero backward scattering (ZBS) and zero forward scattering (ZFS), i.e. the so-called first and second Kerker’s conditions respectively, by sphere spherical particles is considered to be impossible due to the unavailability of naturally occurring magnetic materials in the visible frequency range. We report theoretical modeling to design composite metamaterials that present large optical magnetic permeability in the visible frequency range by employing Mie scattering theory and extended Maxwell Garnett theory. We numerically show that a careful selection of constituents of a composite metamaterial one can obtain metamaterials with sufficiently large artificial permeability that eventually provides the Kerker’s criterion to achieve the Kerker’s conditions. By taking realistic material parameters, we demonstrate that the metamaterials exhibiting ZBS and ZFS have a smaller imaginary part of the refractive index than metallic structures that pave a path to design high-performance nanophotonic devices.

Keywords: Kerker’s conditions, effective medium theory, metamaterials

(Some figures may appear in colour only in the online journal)

1. Introduction
Optimal control of directional scattering by spherical particles in the visible frequency domain is an ultimate goal for researchers to optimize the efficiency of optical devices [1–7]. For referential examples, the desire to achieve zero backscattering plays a crucial role in optical manipulation of spherical particles [8–10] and light management structures used in photovoltaic devices [11, 12]. The Mie scattering theory provides an exact solution to the problem of light scattering by spheres [13] and the seminal work on directional scattering was presented by Kerker et al in 1983 [14]. They revealed proper combinations of relative permittivity and relative permeability of an unconventional magneto-dielectric sphere to achieve zero backward scattering (ZBS) and zero forward scattering (ZFS) that are commonly known as the first and second Kerker’s conditions, respectively. Fulfillment of the first Kerker’s condition requires a material with identical relative permittivity \( \varepsilon \) and relative permeability \( \mu \) (i.e. \( \varepsilon = \mu \)), while the second Kerker’s condition needs material with \( \varepsilon = \frac{4 - \mu}{\mu + 2} \).

Observation of these conditions for the visible light is considered to be impossible due to the unavailability of such materials that possess large relative magnetic permeability, thus, Kerker’s method cannot be applied in optical frequencies. However, in spite of these limitations, ZBS and ZFS have successfully been experimentally demonstrated by a dipolar sphere of large permittivity [15, 16]. In this case, the incident field equally excites the electric and magnetic dipoles that produce an anisotropic scattering pattern, since the anisotropic distribution of the scattered field is governed by the interference between the electric and magnetic dipoles. For instance, the in-phase and out of phase oscillations of the electric and magnetic dipoles can provide an opportunity to observe ZBS and ZFS, respectively [17–20], although these proposals are limited to the sphere of small radius \( r \) as compared to the incident wavelength \( \lambda \), such that \( r \ll \lambda \).

Nowadays, the anomalous but very fascinating scattering behavior is observed by artificially designed so-called...
metamaterials with unconventional optical constants [21, 22]. They have the ability to present unique scattering properties like optical cloaking [23], negative optical reflection [24] and efficient control on scattering directionality [10] with valuable interest in the fields of nanoantennas, nano waveguides and optical manipulations [10, 25–27]. The recent advancement in the field of metamaterials allows us to achieve the invisibility of an object through scattering cancellation by manipulating the optical permittivity and permeability of the materials [23, 28], although it is challenging to find appropriate materials to design the metamaterial that presents a strong artificial magnetism in the visible range. Recently, the ZBS has been shown by a sphere made of composite metamaterial [10] by tuning the permittivity of the sphere in such a way that electric and magnetic multipoles interfere destructively in the backward direction, thus ZBS is achieved. This proposal is neither robust against the radius nor accommodates the permeability of the sphere, while the Kerker’s method is applicable for all radii.

Motivated by the recent advancement in the field of metamaterials, in this article, we put forward a composite material platform to design composite metamaterials that exhibit strong artificial magnetic response in the optical frequencies range. Furthermore, we engineer the optical constants of the composite metamaterials to meet the Kerker’s criterion to achieve the ZBS and ZFS. In order to implement this proposal, we consider a composite metamaterial made of a dielectric host medium containing dipolar spherical inclusions of large optical permittivity [23, 29, 30]. Furthermore, we apply extended Maxwell–Garnett theory to calculate the effective optical constants of homogenized composite metamaterials in the optical frequency range by assuming that the sizes of the inclusions are sufficient to overcome the non-local effects [31, 32]. In addition, the Mie scattering theory is used to calculate the relevant directional scattering cross sections of the composite metamaterials.

The rest of the article is organized as follows. Section 2 is a methodological part, where the Mie scattering theory and extended Maxwell–Garnett (EMG) theory are briefly discussed. The main findings of the work are presented in sections 3, where the Kerker’s conditions for composite metamaterials are analyzed. Finally, we summarize our findings in section 4.

2. Methodology

2.1. Mie theory

Consider a plane wave with vacuum wavelength $\lambda_0$ scattering by a spherical particle of radius $r$, refractive index $n = \sqrt{\varepsilon / \mu}$ embedded in a non-absorbing host medium with refractive index $n_1 = \sqrt{\varepsilon_1 / \mu_1}$, where $\varepsilon$ and $\mu$ are the relative permittivity and relative permeability of the sphere (host medium). The scattering and absorption of incident light by the sphere is described by the Mie scattering theory in terms of normalized scattering efficiency $Q_{sca}$, absorption efficiency $Q_{abs}$, extinction efficiency $Q_{ext}$ and mathematically defined as [13, 17, 18, 33]

\[ Q_{sca} = \frac{2}{X^2} \sum_{\ell=1}^{\infty} (2\ell + 1) |a_\ell|^2 + |b_\ell|^2, \]

\[ Q_{abs} = \frac{2}{X^2} \sum_{\ell=1}^{\infty} (2\ell + 1) (\text{Re}[a_\ell] - |a_\ell|^2 + \text{Re}[b_\ell] - |b_\ell|^2), \]

\[ Q_{ext} = Q_{abs} + Q_{sca}, \]

where $a_\ell$ and $b_\ell$ are the Mie scattering coefficients corresponding to the transverse magnetic and transverse electric modes, respectively. The index $\ell$ is used to denote $\ell^{th}$ order spherical harmonic channel and $x = kr$ is size parameter with $k = 2\pi / \lambda$. The Mie scattering coefficients can be derived by employing the boundary conditions on the surface of the scatterer and written as [13]

\[ a_\ell = \frac{\mu_\psi (mx) \psi_\ell(x) - \mu_\xi (mx) \psi_\ell(x)}{\mu_\psi (mx) \psi_\ell(x) - \mu_\xi (mx) \psi_\ell(x)}, \]

\[ b_\ell = \frac{\mu_\psi (mx) \psi_\ell(x) - \mu_\xi (mx) \psi_\ell(x)}{\mu_\psi (mx) \psi_\ell(x) - \mu_\xi (mx) \psi_\ell(x)}, \]

where $\psi, \xi$ are Riccati–Bessel functions [34], and $m = n / n_1$ is the relative refractive index. In order to study the directional scattering pattern, the expressions for differential scattering efficiencies in forward ($\theta = 0$) and backward ($\theta = \pi$) directions are given as [13]

\[ Q_{sca}|_{\theta=\pi} = \frac{1}{X^2} \sum_{\ell=1}^{\infty} (2\ell + 1)(-1)^\ell (a_\ell - b_\ell)^2, \]

\[ Q_{sca}|_{\theta=0} = \frac{1}{X^2} \sum_{\ell=1}^{\infty} (2\ell + 1)(a_\ell + b_\ell)^2. \]

It can be seen in equation (6), the first Kerker’s condition (i.e. $Q_{sca} = 0$) can be achieved by providing $a_\ell = b_\ell$ and this can occur for identical identical permittivity and permeability of the sphere (see equations (4) and (5)). On the other hand, the second Kerker’s condition can be satisfied by achieving $Q_{sca} \approx 0$, provided that $a_1 \approx -b_1$, which occurs when the condition $\epsilon = \frac{4\pi}{2\mu + 1}$ is satisfied in the quasi-static limit.

It is worth mentioning that the optical theorem imposes a severe condition to achieve ZFS. This theorem of optics relates the extinction cross section $\sigma_{ext}$ to the forward scattering amplitude $s_0(0,0)$ and can be expressed as $\sigma_{ext} = \frac{\lambda^2}{4\pi} \text{Im}[s_0(0,0)]$ [35, 36]. In practice, $Q_{ext} = 0$ (at 2nd Kerker’s condition) becomes negligible without necessarily making both of them zero [14], which not only vanishes the forward scattering but also nullifies the total extinction cross section (one may refer to equation (3) in quasi-static approximation). Subsequently, the non zero $a_1$ and $b_1$ imply that the overall scattering in all other directions may still be significantly different from zero, which seems to be a contradiction to the fact that the extinction cross section vanishes. However, the usual approximation for the dipole
coefficients fail to satisfy energy conservation requirements \[35\]. A more accurate approximation for those coefficients, taking into account radiative corrections, shows that the forward scattering is not exactly zero, relaxing the contradiction. Therefore, one cannot completely suppress the forward scattering, it may be possible to achieve near zero forward scattering (NZFS) at 2nd Kerker’s condition. In order to demonstrate these conditions, we use the effective medium theory that will provide an environment to satisfy Kerker’s conditions.

2.2. Extended Maxwell–Garnett theory

When electromagnetic field propagates through a heterogeneous medium comprises of non-absorbing host medium of permittivity \(\varepsilon_h\), and small spherical inclusions of permittivity \(\varepsilon_i\), radius \(b\), such that \(b \ll \lambda\). There is a variety of effective medium theories for homogenization of the heterogeneous medium \[37, 38\]. The collective optical response of such a homogenized medium can be discussed by defining the effective constants provided by one of the effective medium theory. Due to the best performance, we use the EMG theory \[23, 39–43\], which provides the effective permittivity and permeability as functions of volume filling fraction \(f\), size of the inclusions, permittivity and permeability of the host medium as \(n_{\text{eff}} = \sqrt{\varepsilon_{\text{eff}} / \mu_{\text{eff}}}\), where

\[
\varepsilon_{\text{eff}} = \varepsilon_h \frac{y^3 + 3i\beta_1}{y^3 - \frac{3}{2}i\beta_1},
\]

and

\[
\mu_{\text{eff}} = \mu_h \frac{y^3 + 3i\alpha_1}{y^3 - \frac{3}{2}i\alpha_1},
\]

Here \(y = \sqrt{\varepsilon \omega b / c}\), is the size parameter inside the host medium, where \(\omega\) is the frequency, \(c\) is the speed of light, \(\alpha_1\) and \(\beta_1\) are dipolar Mie coefficients of the inclusions. The size parameter must be \(y \ll 1\) \[43\], otherwise higher order multipoles will contribute to effective permittivity and permeability and spoil the validity of EMG theory.

3. Results and discussions

We begin our numerical analysis by considering a composite material made of \(\text{SiO}_2\) as the host medium with relative permittivity \(\varepsilon_h = 2.1\) \[44\] and dielectric spherical inclusions of radius 20 nm, bulk relative permittivity \(\varepsilon_i = 169\) \[23, 29\] and volume filling fraction \(f\). The effective optical constants of the composite material are calculated by EMG theory (section 2.2), since the inclusions have a large refractive index so that the incident field can generate displacement currents inside the inclusions and produces tiny magnetic dipoles. The collective effect of these spherical magnetic dipoles generates a remarkable magnetic response in the composite metamaterials. The effective optical response of the composite metamaterials is measured in terms of effective permittivity and effective permeability by using equations (8) and (9) that have external degrees of freedom, like volume filling fraction, permittivities and size of inclusions. One can perform fine
tuning of these parameters to tailor the resonance at a chosen wavelength for operation.

In figure 1(a), the real and imaginary parts of the effective permittivity (blue) and permeability (purple) of the composite metamaterial are displayed as a function of incident wavelength. It is clearly seen that the composite metamaterial shows remarkable magnetic response with a spectral region that indeed lies in our domain of interest. For instance, the composite metamaterial shows an identical effective permeability and index of refraction at the wavelength of 560 nm with relatively small imaginary parts. According to Kerker’s method, this is an ideal scenario to satisfy the first Kerker’s condition for all sized spherical particles. Figure 1(b) shows a color map of backward scattering efficiency by the composite sphere as a function of the incident wavelength and radius of the sphere. It is clearly seen that at $\lambda = 560$ nm the backward scattering is completely suppressed which occurs due to the in-phase oscillations of the electric and magnetic multipoles with the same amplitudes such that $a_\ell = b_\ell$, provided by identical $\varepsilon_{\text{eff}}$ and $\mu_{\text{eff}}$ as shown in figure 1(a). Therefore, the overall backward scattering efficiency defined in equation (6) is reduced to zero regardless of the sizes of the sphere. The drastic reduction of backward scattering by the composite sphere of radius 500 nm is shown in figure 1(c) by calculating the backward scattering efficiency versus wavelength. It is clearly shown that the dramatic reduction appears at $\lambda = 560$ nm and backward scattering amplitude reduces to zero, thus satisfying the 1st Kerker’s condition.

In contrast, the 2nd Kerker’s condition requires the condition $\varepsilon_{\text{eff}} = \frac{4 - \mu_{\text{eff}}}{\mu_{\text{eff}}}$, which surprisingly also occurs at a wavelength of 500 nm. In figure 1(d), we calculate the forward scattering efficiency by a composite sphere of radius 70 nm as a function of the incident wavelength. The drastic reduction of forward scattering appears at $\lambda = 500$ nm and satisfies the 2nd Kerker’s condition. It is worth mentioning that the second Kerker’s condition can be achieved by providing $a_\ell = -b_\ell$, which only implies to the dipolar particles (i.e. for $\ell = 1$). Otherwise, this effect might be spoiled for a large sphere due to higher-order multipole contributions to the forward scattering efficiency. Thus, provides nearly zero forward scattering $Q_f \approx 0$ at $\lambda = 500$ nm for a dipolar sphere (of radius 70 nm).

Now we extend our proposal to design metamaterials by using naturally occurring materials that will present a significant magnetic response to satisfy the Kerker’s conditions. The immediate question that arises, is there any material that possesses large permittivity in the visible region? The answer is yes, for instance, a carefully designed core–shell system shows a very high permittivity due to plasmonic resonances with strong spectral dependence and in the quasi-static approximation these core–shell nanoparticles can be used as inclusions into a host medium. For simplicity, we begin with an internal homogenization approach introduced by Chettiar and Engheta [30] that provides effective permittivity of core–shell nanoparticles by homogenizing it to an effective sphere of the same dimension. Through this approach, the effective permittivity of a core–shell nanoparticle is calculated by equating the polarizabilities of the core–shell to a homogenized
spherical nanoparticles. In order to implement this proposal, we consider metallic nanosphere as a core of radius $b$ embedded in dielectric spherical shell of radius $a$, forming a core–shell nanoparticle [45–48]. Assuming that the core is silver (Ag) and we may use Drude formula to define it is permittivity, $\epsilon_c = \frac{\omega_p^2}{\omega^2 + \im \omega \Gamma}$, with $\omega_p = 9.2$ eV, $\Gamma = 0.0212$ eV, $\omega_\infty = 5.0$, where 1 eV = 241.8 THz [49] and $\omega$ is the angular frequency of incident light. Furthermore, the core–shell is homogenized to an effective sphere of radius $a$ with permittivity $\epsilon_{MG}$ provided by equation (10) and displayed in figure 2(b) at fixed radii ratio at $\frac{b}{a} = 0.84$. It has been verified that in the quasi-static limit, the effective sphere has a good agreement to exact Mie solution of a core–shell nanoparticles of the same dimension [50] and indeed it can be used as inclusion into a host medium to fabricate the structures of desired optical and physical properties [30, 50–52].

Let us design a bulk composite medium comprises of randomly arranged effective (homogenized) nanospheres with permittivity $\epsilon_{MG}$ and radius $a$ embedded in a host medium with volume filling fraction $f$. In figure 2(c), we calculate the effective permittivity (blue) and permeability (purple) of newly designed composite metamaterial by means of EMG theory at fixed $f = 0.25$ as a function of $\lambda$. In this configuration, we consider radii of the effective nanosphere about 14 time less than the incident wavelength, which is a safe zone for the validation of the effective medium theory [43].

It can be seen in figure 2(c) that the composite metamaterial not only presents large effective permeability but also provides identical permittivity and permeability with week imaginary parts at a wavelength of 650 nm. Therefore, according to Kerker’s criterion, it must present zero backward scattering at this wavelength. We now calculate the directional scattering by the composite sphere of radius $r$ with permittivity given in figure 2(c). The backward scattering efficiency and forward scattering efficiency as a function of incident wavelength are displayed in figures 3(a) and (b), respectively. It is clearly seen that the backward scattering efficiency is drastically reduced to zero at $\lambda = 650$ nm which is due to the fact that equal electric and magnetic response. On the other hand, the NZFS appears at a wavelength of 620 nm due to out of phase oscillations of electric and magnetic dipoles, thus satisfying the 2nd Kerker’s condition.

4. Conclusions

In this paper, we have theoretically modeled composite metamaterials using Mie scattering theory and effective medium theory. Our numerical results have shown that the composite metamaterials present strong artificial magnetic permeability in the optical frequency domain that appears due to the displacement currents inside the inclusions. Our findings show that a fine tuning of the parameters appears in the EMG theory, where one can create a spectral region with identical permittivity and permeability not only for bulk homogeneous composite sphere but also for a regularly arranged nanospherical particles. Altogether, this proposal not only paves a path to design new possible structures with artificial magnetic response but also allows us to achieve Kerker’s conditions for low refractive index materials in the visible frequency range. These structures with unique optical properties have much lower Im$(\mu_{eff})$ than a metallic structure, which makes our proposal a good candidate to design high-performance optical antennas, metamaterials and other novel nanophotonic devices.

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ORCID ID

Rfqaat Ali https://orcid.org/0000-0003-2612-4020

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