Research article

SeokJae Yoo[a], Suyeon Lee[a], Jong-Ho Choe and Q-Han Park*

Causal homogenization of metamaterials

https://doi.org/10.1515/nanoph-2019-0069
Received March 2, 2019; revised April 17, 2019; accepted April 20, 2019

Abstract: We propose a homogenization scheme for metamaterials that utilizes causality to determine their effective parameters. By requiring the Kramers-Kronig causality condition in the homogenization of metamaterials, we show that the effective parameters can be chosen uniquely, in contrast to the conventional parameter retrieval method which has unavoidable phase ambiguity arising from the multivalued logarithm function. We demonstrate that the effective thickness of metamaterials can also be determined to a specific value by saturating the minimum-error condition for the causality restriction. Our causal homogenization provides a robust and accurate characterization method for metamaterials.

Keywords: metamaterials; effective medium theory; parameter retrieval method; causality; homogenization.

1 Introduction

Metamaterials composed of artificial subwavelength structures, namely meta-atoms, can exhibit extraordinary optical properties not found in natural materials. The basic characterization of such metamaterials is the homogenization of the meta-atoms such that the effective parameters of the homogenization describe the overall optical response of the particular collection of meta-atoms [1–3]. When the size of the meta-atoms is sufficiently small compared to the wavelength of light, such a homogenization of the metamaterial well describes its optical properties. The Nicholson-Ross-Weir (NRW)-type parameter retrieval methods, which are easy to implement for the numerical design of metamaterials, are now used as the standard to determine effective metamaterial parameters [3–10].

Homogenization of the real-world metamaterials, however, occasionally results in noncausal artifacts in the resulting effective parameters. The magnetic permeability, for example, sometimes exhibits antiresonant behavior [11–13], which was criticized to be inconsistent with the Kramers-Kronig (KK) causality relation for passive media [14]. Efforts have been made to avoid the inconsistency by considering the effects of spatial dispersion and higher order multipoles [15, 16], generalizing the Ladau-Lifshitz's modification of the KK relation [17, 18] or asserting the physical validity of antiresonance [19]. Even though metamaterials do not exhibit antiresonances, the finite size a of meta-atoms also introduces the off-resonant noncausality to the effective parameters because the homogenization procedure of metamaterials necessarily brings up noncausal time advance of the order $\sim a/c$ [20].

In addition to the noncausal artifacts in the homogenization scheme, the conventional NRW-type parameter retrieval method poses another technical problem, which leads to inconsistency with the KK relation. The method has difficulty fixing parameters uniquely for two reasons: (i) the real part of the refractive index Re(n) obtained from the multivalued complex logarithm function possesses integer-labeled multiple branches and the rules needed to choose a specific branch are absent, and (ii) the effective thickness of the homogenized slab is not uniquely determined because of the lack of a well-defined boundary for inhomogeneous meta-atom collections and the thickness is therefore usually selected according to the researcher's own taste. These ambiguities are likely to exaggerate or underestimate the effective refractive index of the metamaterial but have been recognized as an inevitable limitation of an otherwise effective parameter retrieval method.

Various methods have been proposed to overcome the parameter ambiguity caused by the multiple branches. The most prevalent method is choosing the branch that makes Re(n) continuous in the spectral range of interest [3, 21]. This method can determine the refractive index without discontinuity points in the spectrum, but there still remain infinite sets of continuous Re(n) resulting

[a]**SeokJae Yoo and Suyeon Lee:** These authors contributed equally to this work.
***Corresponding author: Q-Han Park,** Department of Physics, Korea University, Seoul 02841, Korea, e-mail: qpark@korea.ac.kr. https://orcid.org/0000-0002-6438-7123
SeokJae Yoo: Department of Physics, Korea University, Seoul 02841, Korea; and Department of Physics, University of California, Berkeley, CA 94720, USA
Suyeon Lee and Jong-Ho Choe: Department of Physics, Korea University, Seoul 02841, Korea

from simultaneous constant shifts of the chosen branches to another set of branches. Sometimes the continuity of Re(n) cannot be restored even by a selective choice of m, given the interfering multiple resonances resulting in sharp Fano resonances. Another method of avoiding parameter ambiguity is demanding that the effective refractive index n_{eff} satisfies the KK causality relation [8, 22]. However, the refractive index n_{eff} does not generally satisfy the KK relation, and this method cannot be applied to metamaterials, which usually have strong electromagnetic responses.

In this paper, we propose a causal homogenization method for metamaterials that determines their effective parameters without ambiguity. We impose the causality requirement on the effective permittivity ε_{eff} and permeability μ_{eff}. This places a strict restriction on the refractive index n_{eff} and the impedance z_{eff} of the metamaterials and removes the branch ambiguity of the real part of the refractive index Re(n_{eff}). We also demonstrate that the ambiguity surrounding the effective thickness d_{eff} can be resolved by minimizing the spectrally averaged branch error δm that arises when fixing the real part of the refractive index Re(n_{eff}). Our homogenization method enables an algorithmic determination of the effective parameters and can be used as a general method for metamaterial homogenization.

2 Results

2.1 Deterministic causal homogenization of metamaterials

In the standard method of metamaterial homogenization (Figure 1) [3], we retrieve the effective refractive index n_{eff} and the impedance z_{eff} from the measured reflection coefficient r and the transmission coefficient t using the relations

$$n_{\text{eff}} = n_0 + \frac{2m\pi}{k_0 d_{\text{eff}}} = \left(\text{Re}\,n_0 + \frac{2m\pi}{k_0 d_{\text{eff}}}\right) + i\,\text{Im}\,n_0, \tag{1}$$

$$n_0 = \frac{1}{ik_0 d_{\text{eff}}} \ln\left\{\frac{t}{1 - r(z_{\text{eff}} - 1)/(z_{\text{eff}} + 1)}\right\}, \tag{2}$$

$$z_{\text{eff}} = \pm\sqrt{\frac{(1+r)^2 - t^2}{(1-r)^2 - t^2}} = \text{Re}\,z_{\text{eff}} + i\,\text{Im}\,z_{\text{eff}}, \tag{3}$$

where the integer m is the branch, n_0 is the effective refractive index of the zeroth-order branch ($m=0$), k_0 is the wavenumber of the incident light, and d_{eff} is the effective thickness of the homogenized slab. In Eqs. (1)–(3), the undetermined integer branch m makes the effective parameters non-unique although the passivity condition requires that Re(z_{eff}) > 0 and Im(n_{eff}) > 0 [3]. Therefore, the homogenization procedure in metamaterial research admits multiple solutions for the homogenized permittivity and permeability.

In this work, we suggest that electromagnetic causality can determine the effective parameters uniquely. Causality is the most fundamental principle in physics because a physical action can occur only after its cause. In electromagnetics, the displacement field $\mathbf{D}(\boldsymbol{r}, \tau')$ at the position $\boldsymbol{r}$ and time τ is determined only by the electric field $\mathbf{E}(r, \tau')$ for time $\tau' \le \tau$. For a linear medium, whose optical response is described by its permittivity $\varepsilon(\omega)/\varepsilon_0$ relative to vacuum, the constitutive relation $\mathbf{D}(\boldsymbol{r}, \omega) = \varepsilon(\omega)\mathbf{E}(\boldsymbol{r}, \omega)$ determines the connection between $\mathbf{D}$ and $\mathbf{E}$ at the single frequency ω. If the constitutive relation of the fields $\mathbf{D}$ and $\mathbf{E}$ obeys causality, then the relative permittivity $\varepsilon(\omega)/\varepsilon_0$ will have analyticity in the upper half ω plane [23]. This mathematical condition for $\varepsilon(\omega)/\varepsilon_0$ gives rise to the KK relations

$$\text{Re}\{\varepsilon(\omega)/\varepsilon_0\} = 1 + \frac{1}{\pi} P\int_{-\infty}^{\infty} \frac{\text{Im}\{\varepsilon(\omega')/\varepsilon_0\}}{\omega' - \omega} d\omega', \tag{4}$$

$$\text{Im}\{\varepsilon(\omega')/\varepsilon_0\} = -\frac{1}{\pi} P\int_{-\infty}^{\infty} \frac{\text{Re}\{\varepsilon(\omega)/\varepsilon_0 - 1\}}{\omega' - \omega} d\omega'. \tag{5}$$

We may expect the same relation for the permeability $\mu(\omega)/\mu_0$. However, it was noted that the KK relation applied to the permeability $\mu(\omega)/\mu_0$, given by Eqs. (6) and (7), is not consistent with diamagnetism at zero frequency if the imaginary part Im{$\mu(\omega)/\mu_0$} is positive for all ω [17]. Recently, it was argued that the imaginary part of permeability can be negative without violating physical laws [19]. Metamaterials with negative refractive index have

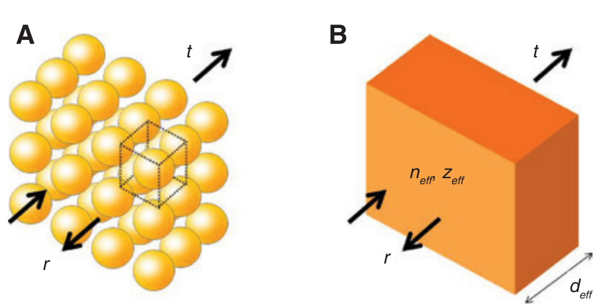

Figure 1: Homogenization of metamaterials.
(A) Schematic of a gold nanoparticle array. (B) The homogenized slab with the effective parameters. r and t are the reflection and transmission coefficients, respectively.

effective parameters showing resonant and anti-resonant behaviors; that is, the imaginary part of permittivity is positive and that of permeability is negative near resonance [14]. On the other hand, we note that the induced magnetization is determined by the applied field B so that the causality should be applied to μ^{-1}. Nevertheless, the analytic properties of μ are also possessed by μ^{-1} [17], i.e. μ^{-1} is analytic in the upper half-plane if μ is analytic and has no zeros in the upper half-plane. Thus we adopt the same KK relation for the permeability $\mu(\omega)/\mu_0$, namely

$$\mathrm{Re}\{\mu(\omega)/\mu_0\} = 1 + \frac{1}{\pi} P\int_{-\infty}^{\infty} \frac{\mathrm{Im}\{\mu(\omega')/\mu_0\}}{\omega' - \omega} \mathrm{d}\omega', \tag{6}$$

$$\mathrm{Im}\{\mu(\omega')/\mu_0\} = -\frac{1}{\pi} P\int_{-\infty}^{\infty} \frac{\mathrm{Re}\{\mu(\omega)/\mu_0 - 1\}}{\omega' - \omega} \mathrm{d}\omega'. \tag{7}$$

Here, we require the effective parameters of homogenized metamaterials to obey the causality and show that this requirement uniquely fixes the branch without ambiguity, thereby providing a deterministic parameter retrieval method. To impose the KK relation, we use the effective permeability $\mu_{\mathrm{eff}} = n_{\mathrm{eff}} z_{\mathrm{eff}}$ because the real and imaginary parts of n_{eff} and z_{eff} themselves do not obey the causality relation [24].

To obtain the causal condition for the branch $m(\omega)$ at the frequency ω, we first express the real and imaginary parts of the effective permeability in terms of the refractive index [Eq. (1)] and the impedance [Eq. (3)]. They are, respectively, given by

$$\mathrm{Re}\ \mu_{\mathrm{eff}} = \mathrm{Re}\ n_0 \mathrm{Re}\ z_{\mathrm{eff}} - \mathrm{Im}\ n_0 \mathrm{Im}\ z_{\mathrm{eff}} + \frac{2m\pi}{k_0 d_{\mathrm{eff}}} \mathrm{Re}\ z_{\mathrm{eff}}, \tag{8}$$

$$\mathrm{Im}\ \mu_{\mathrm{eff}} = \mathrm{Re}\ n_0 \mathrm{Im}\ z_{\mathrm{eff}} + \mathrm{Im}\ n_0 \mathrm{Re}\ z_{\mathrm{eff}} + \frac{2m\pi}{k_0 d_{\mathrm{eff}}} \mathrm{Im}\ z_{\mathrm{eff}}. \tag{9}$$

Plugging Eqs. (8) and (9) into the KK relation in Eq. (4), we obtain the Fredholm integral equation of the second kind for the integer branch $m(\omega)$ as follows:

$$m(\omega) = g(\omega) + P\int_0^{\infty} K(\omega', \omega) m(\omega') \mathrm{d}\omega', \tag{10}$$

where the functions $g(\omega)$ and $K(\omega', \omega)$ are given by

$$\begin{aligned} g(\omega) = {} & \frac{d}{\lambda \mathrm{Re}\{z_{\mathrm{eff}}(\omega)\}} [1 + \mathrm{Im}\{n_0(\omega)\} \mathrm{Im}\{z_{\mathrm{eff}}(\omega)\} \\ & - \mathrm{Re}\{n_0(\omega)\} \mathrm{Re}\{z_{\mathrm{eff}}(\omega)\}] \\ & + \frac{2}{\pi} \frac{d}{\lambda \mathrm{Re}\{z_{\mathrm{eff}}(\omega)\}} P\int_0^{\infty} \frac{\omega'}{\omega'^2 - \omega^2} [\mathrm{Re}\{n_0(\omega')\} \mathrm{Im}\{z_{\mathrm{eff}}(\omega')\} \\ & + \mathrm{Im}\{n_0(\omega')\} \mathrm{Re}\{z_{\mathrm{eff}}(\omega')\}] \mathrm{d}\omega', \end{aligned} \tag{11}$$

$$K(\omega', \omega) = \frac{2}{\pi} \frac{\mathrm{Im}\{z_{\mathrm{eff}}(\omega')\}}{\mathrm{Re}\{z_{\mathrm{eff}}(\omega)\}} \frac{\omega}{\omega'^2 - \omega^2}. \tag{12}$$

Equation (10) is the key result of this work. Previous works have applied the KK relation to the effective refractive index n_{eff} [8, 22]. However, the refractive index is not a physical response function respecting causality and there is no reason to require the KK relation [24]. Since Eq. (10) is a Fredholm integral equation of the second kind, it can be numerically solved. Discretizing the frequency by the N steps $\Delta\omega'$, Eq. (10) is converted to the matrix equation [25]

$$\begin{pmatrix} m(\omega_1) \\ \vdots \\ m(\omega_N) \end{pmatrix} = \left[\mathbf{I} - \Delta\omega' \begin{pmatrix} 0 & \cdots & K(\omega_N, \omega_1) \\ \vdots & \ddots & \vdots \\ K(\omega_1, \omega_N) & \cdots & 0 \end{pmatrix} \right]^{-1} \begin{pmatrix} g(\omega_1) \\ \vdots \\ g(\omega_N) \end{pmatrix}, \tag{13}$$

with the discretized frequencies ω_i and the identity matrix $\mathbf{I}$ of dimension $N \times N$. By solving the matrix equation, Eq. (13), we can uniquely determine the branch integer m for the real part of the refractive index n_{eff}. Note that diagonal elements of the first matrix in Eq. (13) are zero because the principal values are defined to avoid the singularity of the integrand in Eq. (10).

The remaining ambiguity in determining the homogenized parameters is the effective thickness of the homogenized slab, d_{eff}. Metamaterials are usually composed of artificial nanostructures with uneven surfaces, and thus the boundary of the homogenized slab is often ambiguous. To determine the boundary of the homogenized slab, we assert that the effective thickness d_{eff} can be determined by minimizing the spectral averaged branch error

$$\Delta m \equiv \langle m(\omega) - \mathrm{round}\{m(\omega)\} \rangle_{\omega}. \tag{14}$$

The bracket in Eq. (14) represents the spectral average over the domain of interest. The branch m should be integer in the ideal case, but homogenization artifacts such as the finite size of meta-atoms can cause non-integer solutions of m in the integral equation Eq. (10). Therefore, the averaged deviation of m from the integer in the spectral range of interest can be a measure of the homogenization artifacts. However, the following subsection will demonstrate that the choice of the effective thickness d_{eff} can minimize the deviation of m from an integer.

2.2 Homogenization of a gold nanoparticle array

To demonstrate the causal homogenization of metamaterials, we test the case of a gold nanoparticle array.

Figure 2A shows a monolayer of gold nanoparticles of radius $R=10$ nm and periodicity $P=26$ nm, homogenized by the effective parameters. We vary the effective thickness d_{eff} to find the optimum value for minimizing the spectral averaged branch error Δm in Figure 2B. As the effective thickness d_{eff} increases, the branch m starts to deviate from the integer. The deviation is small at small frequencies but can be significant in the high-frequency region. Figure 2C shows that the spectral averaged branch error Δm is minimized at $d_{\text{eff}}=2R=20$ nm. Numerical simulation was carried out using the home-built finite-difference time-domain (FDTD) software. The grid size for the FDTD simulation is 1 nm. Two outer surfaces of the simulation domain normal to the wave propagation direction were perfectly matched layers (PMLs) of 10 grids, and four outer surfaces parallel to the propagation direction were determined by the periodic boundary condition (PBC). Material property of gold was taken from tabulated data [26]. The transmission coefficient t (the reflection coefficient r) is evaluated by the ratio between the incident fields and the transmitted (reflected) fields, which is spatially averaged at the plane normal to the incident wave vector in order to obtain the zeroth-order transmission (reflection). The wavelength range spans from 300 to 900 nm with the step size 1 nm ($N=600$), which corresponds to the frequency step size $\Delta\omega'/2\pi=1.11$ THz.

Having determined the effective thickness, we can obtain the remaining effective parameters ε_{eff}, μ_{eff}, n_{eff}, and z_{eff}. Figure 3 shows the effective parameters of the homogenized gold nanoparticles. Near the frequency of 600 THz,

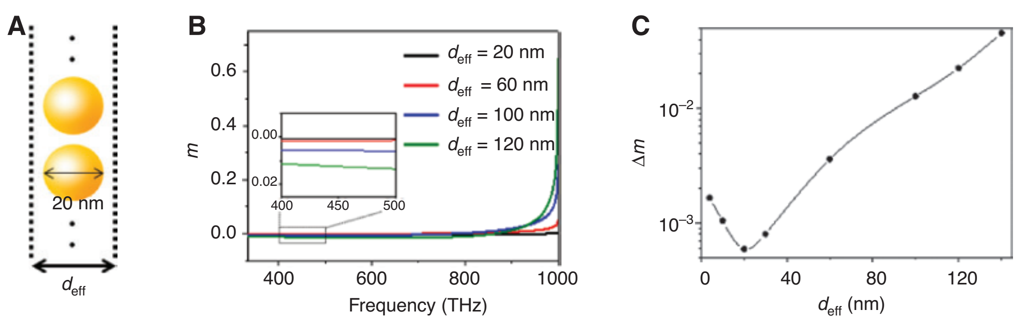

Figure 2: Error minimizing choices of the effective thickness d_{eff} and the branch m.
(A) Schematic of a gold nanoparticle monolayer. (B) The branch m for different choices of effective thickness d_{eff}. (C) The spectral averaged branch error Δm against effective thickness d_{eff}.

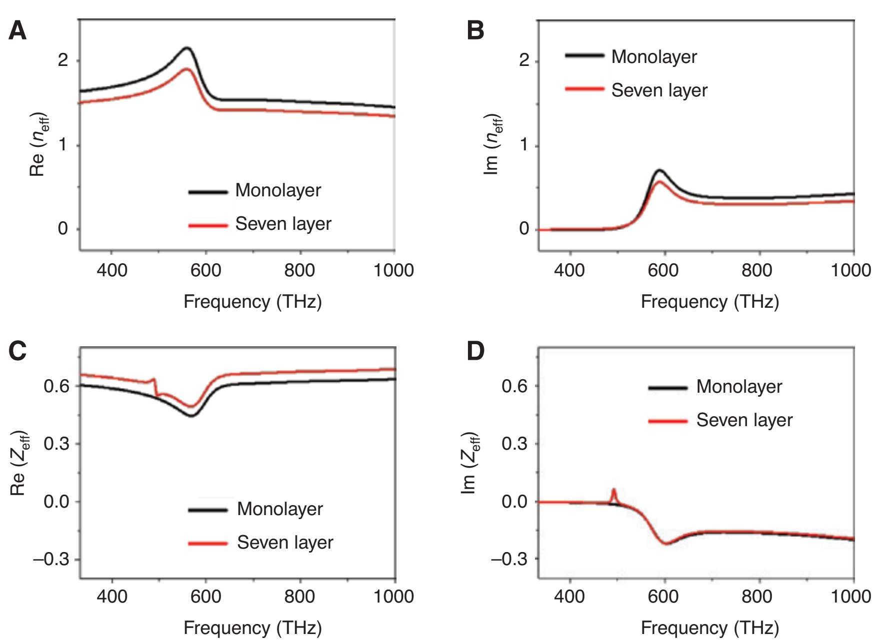

Figure 3: Effective parameters for the homogenized gold nanoparticle monolayer.
(A) The real and (B) the imaginary part of the effective refractive index n_{eff} and (C) the real and (D) the imaginary part of the effective impedance z_{eff} of the monolayer (black lines) and seven layers (red lines) of gold nanoparticles.

we can find the surface plasmon resonance of the gold nanoparticles, which gives resonant peaks in the refractive index n_{eff} in Figure 3B. The permittivity and permeability can be simply obtained by the relations $\varepsilon_{eff}=n_{eff}/z_{eff}$ and $\mu_{eff}=n_{eff}z_{eff}$, respectively. Note that the homogenized parameters presented in Figure 3 agree well with the theoretical predictions [27]. In Figure 3, our homogenization method for a monolayer yields $m=0$ because the monolayer of gold nanoparticles is too thin to have a nonzero branch m.

To confirm the ability of our method to determine the nonzero branch m for bulky metamaterials, we homogenized seven layers of gold nanoparticles of periodicity $P=26$ nm in Figure 4. The homogenized slab is thick compared to the wavelength range of light. By minimizing the spectral averaged branch error Δm, we find that the effective thickness d_{eff} is 182 nm. Note that $7P=182$ nm. As shown in Figure 4A,B, the effective impedance z_{eff} and the imaginary part of the refractive index $\mathrm{Im}(n_{eff})$ are obtained uniquely because the homogenization procedure lacks ambiguity, as shown in Eqs. (1) and (3). The ambiguity occurs in the real part of the refractive index $\mathrm{Re}(n_{eff})$. Figure 4C shows multiple branches of the real part of the refractive index $\mathrm{Re}(n_{eff})$. Without considering the causality in the homogenization procedure, the branch $m(\omega)$ can be any integer that makes the refractive index continuous in the spectral window of interest. For example, the zeroth-order branch ($m=0$) is chosen for frequencies below 493 THz and the first-order branch ($m=1$) is chosen for higher frequencies to avoid abrupt, noncontinuous changes in the refractive index. Other choices are also possible with the integer difference $\Delta m=1$ at 493 THz. For example, we can choose $m=1$ below 493 THz and $m=2$ above 493 THz when the causality condition is not imposed. Another noncausal choice of m depicted in Figure 4C is $m=-1$ below 493 THz and $m=0$ above 493 THz. This degree of freedom when choosing the branch cannot be reduced without the causality condition. However, the causality condition can determine the set of branches $m(\omega)$ in the spectrum uniquely. Figure 4D shows the branch m solution for the integral equation Eq. (10) for seven layers of gold nanoparticles. The solution automatically guarantees the continuity of the real part of the refractive index $\mathrm{Re}(n_{eff})$. We also confirm that the retrieved refractive indices of the seven layers are consistent with that of monolayer, as shown in Figure 3.

3 Conclusion

We would like to emphasize once again that the refractive index n does not obey the KK relations [Eqs. (4) and (5)] because the refractive index n may have singularities in the upper half-plane of the complex ω space [24]. The refractive index can be related to the KK relation only when the permittivity and the permeability are sufficiently small, i.e. $n=\sqrt{\varepsilon\mu/\varepsilon_0\mu_0}\approx(1+\chi_e/2)(1+\chi_m/2)$, where χ_e and χ_m are the electric and magnetic

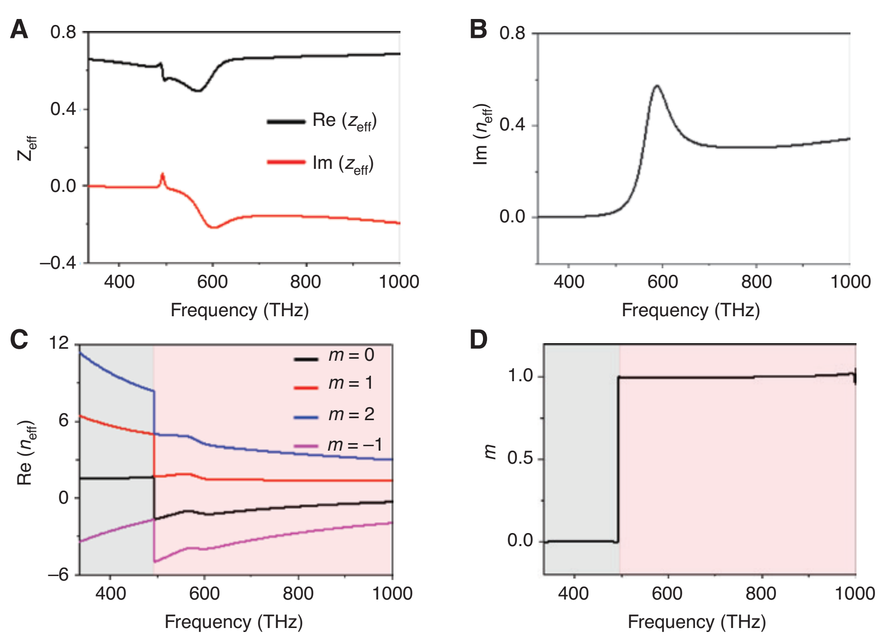

Figure 4: Effective parameters for the homogenized seven layers of gold nanoparticles.
(A) The effective impedance z_{eff} and (B) the imaginary part of the refractive index n_{eff} of the seven layers of gold nanoparticles.
(C) Examples of multiple branches of the real parts of the refractive index n_{eff} corresponding to the branch m without considering the causality condition. (D) The branch m determined by our homogenization method [Eq. (10)].

susceptibility, respectively. This is obviously not the case for metamaterials because they usually have strong electric and magnetic responses. Imposing the KK relation to the refractive index n may result in non-integer values of the branch m, implying noncausal parameter-retrieval results [8].

The KK relation is composed of the four Hilbert transformations between the permeability and the permittivity, as shown in Eqs. (4)–(7). Therefore, it provides the four equivalent forms of the causal branch m. (We introduce one of them in the main text. See Supplementary Material for the full equations.) The four equivalent forms of branch m give the same result for the causal metamaterial homogenization, but the results may be differently influenced by the truncation errors introduced during the numerical integration. Although the integral in the KK relation includes integration over the whole frequency range, the numerical implementation requires truncation of the integration range. Therefore, it is recommended that the permeability relations [Eqs. (4) and (5)] are used for electrically resonant metamaterials because electric responses have a limited effect on permeability and thus doing so can minimize the truncation errors in the numerical integration. However, for magnetically resonant metamaterials, the permittivity relations (6) and (7) should be used.

We demonstrated the homogenization scheme for linear dielectric metamaterials, but note that our method is also applicable to other media, such as anisotropic [28], chiral [29], bi-isotropic [4, 5], and gyrotropic media [30, 31]. The optical parameters of these media also obey causality and follow the KK relation [32, 33]. Thus, extending our method based on the KK relation to wide-ranging media types is straightforward.

Acknowledgments: This work was supported by the Center for Advanced Meta-Materials (CAMM) funded by the Ministry of Science and ICT as a Global Frontier Project (CAMM-2014M3A6B3063710). SeokJae Yoo and Jong-Ho Choe were supported the National Research Foundation of Korea (NRF) funded by the Ministry of Education (2017R1A6A3A11034238 and 2017R1A6A3A11034292).

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Supplementary Material: The online version of this article offers supplementary material (https://doi.org/10.1515/nanoph-2019-0069).