Comment on "Negative refractive index in artificial metamaterials"

Alexander V. Kildishev,[1,*] Vladimir P. Drachev,[1] Uday K. Chettiar,[1] Vladimir M. Shalaev,[1] Douglas H. Werner,[2] and Do-Hoon Kwon[2]

[1]*Birck Nanotechnology Center, Purdue University, West Lafayette, Indiana 47907, USA*
[2]*Department of Electrical Engineering, Pennsylvania State University, University Park, Pennsylvania 16802, USA*
[*]*Corresponding author: Kildishev@purdue.edu*

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We dispute Grigorenko's statement [Opt. Lett. **31**, 2483 (2006)] that measuring only the reflection intensity spectrum is sufficient for determining the effective refractive index. In addition, our simulations do not confirm his conclusions regarding the negative refractive index and the negative permeability of the nanopillar sample in the visible range.

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A simpler way to obtain the effective μ and ε, and thence n, of a thin layer of a metamaterial, has been reported recently by Grigorenko [1,2]. The Grigorenko method (GM) is built on a best-fit procedure of a given reflection spectrum ($R(\lambda)$) with *a priori* prescribed dispersion relations for $\varepsilon(\lambda)$ and $\mu(\lambda)$ [2]:

$$\epsilon(\lambda) = 1 + F_e\lambda^2/(\lambda^2 - \lambda_e^2 - \imath\lambda\Delta\lambda_e),$$
$$\mu(\lambda) = 1 + F_m\lambda_m^2/(\lambda^2 - \lambda_m^2 - \imath\lambda\Delta\lambda_m), \qquad (1)$$

The method uses six fitting constants (F_m, λ_m, $\Delta\lambda_m$, F_e, λ_e, and $\Delta\lambda_e$) and an "effective optical thickness," which is smaller than the actual, physical thickness of the sample. In the other method [3] the optical characterization is done by spectral analysis of the complex transmission and reflection coefficients, t and r [3], and then by retrieval of the effective refractive index, $n = n' + \imath n''$, and impedance, $\eta = \eta' + \imath\eta''$, by using the actual thickness of the sample. This technique gives unambiguous results for n and η and the effective values of permeability, $\mu = n\eta$, and permittivity, $\varepsilon = n/\eta$.

Grigorenko claims [2] that a sample with Au nanopillars covered with a thin glycerine layer shows $n' = -0.7$ at green light and has a quality ratio of $-n'/n'' = 0.4$. But in contrast with these controversial statements, only $n' > 0$ was obtained in our simulations for the same geometry. Additionally, neither $\mu < 0$ nor $\epsilon < 0$ at green light was obtained from three different simulation tools: finite-element software, a combination of periodic finite-element method–boundary integral (PFEBI) solver, and a parallel 3D finite-difference time-domain solver. A unit cell is shown in Fig. 1(a).

References [1,2] are focused on the characterization of a single layer of coupled Au "nanopillars" arranged in a biperiodic array. The works [1,2] refer to a simulation using a commercial FE solver, where a Drude model is used with the parameters for Au from Ref. [1], p. 337. We note that this model is incapable of adequately describing the behavior of Au in green light. Figure 1(a) compares the Drude model with the parameters from [3] versus the experimental data [4]. The area near the green light differs greatly, and we thus comment that the results of FE simulations shown in [1] are of little assistance to the problem.

Now we consider the GM first shown in [1] and then discussed in some detail in [2]. The method is a four-step recipe: (i) Determine $R(\lambda)$ of a given sample. (ii) Prescribe fixed dispersive relationships for the material using Eq. (1). (iii) Match $R(\lambda)$ using the fitting constants and an "effective optical thickness." Note that the effective thickness in GM is defined separately either using ellipsometry or by taking it as a "mass thickness," i.e., it is much smaller than the actual physical thickness of the layer of nanopillars [the physical thickness is denoted as h in Fig. 1(a)]. Finally (iv) calculate n using the fitting parameters obtained in Step (iii) with Eq. (2) of [2].

Below we compare GM with our simulations. To verify that we understood GM [1,2] correctly, we compared the approximated $R(\lambda)$ calculated using the original fitting parameters (listed in the caption of Fig. 2(d) of [2]) with the same spectrum obtained from measurements also shown in Fig. 2(d) of [2]. Indeed, the fitting parameters selected by the author in [2] provide an adequate match with the experimental curve as shown in Fig. 2(a). Figures 2(a)–2(d) also show the results of the formal calculations using Eq. (1) and Eq. (2) of [2]. However, the results of the GM

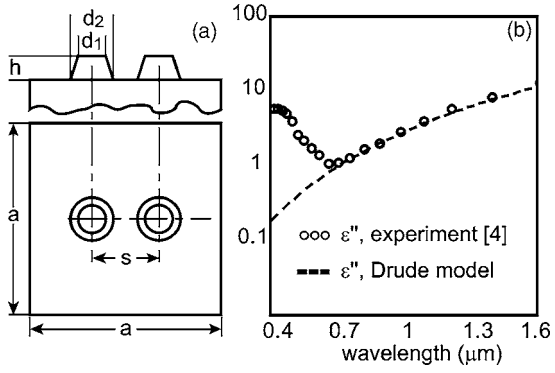

Fig. 1. (a) Unit cell with $a = 400$ nm, $h = 90$ nm, $s = 140$ nm, and $d = (d_2 + d_1)/2 = 110$ nm taken from [1,2]. (b) ϵ'' plotted using the Drude model [3] and the data of [4].

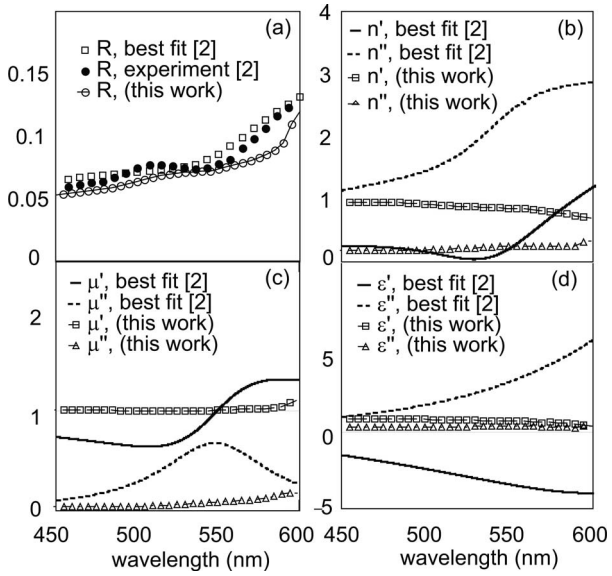

Fig. 2. (a) $R(\lambda)$ obtained for the sample of Fig. 1 using a best-fit technique [1] versus the experiment also shown in [2]. (b)–(d) Effective parameters $(n', n'', \mu', \mu'', \epsilon', \epsilon'')$ obtained from the best fit [2] versus the same parameters obtained from our numerical simulations.

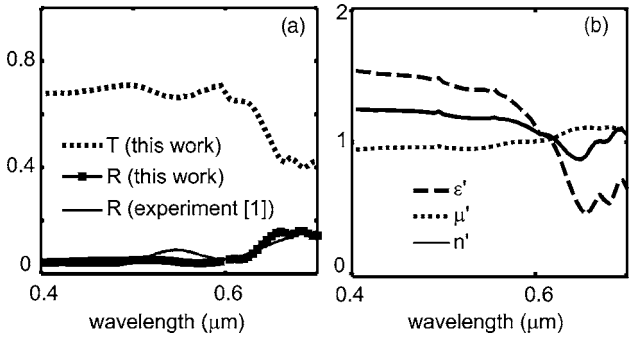

Fig. 3. (a) $T(\lambda)$ and $R(\lambda)$, calculated for the glycerine-covered sample using PFEBI. The simulated $R(\lambda)$ is compared with the experiment of [1]. (b) ϵ', μ', and n' calculated from t and r for the same sample.

contain an internal inconsistency and differ strongly from our electromagnetic simulations.

To validate the results of the GM, the PFEBI solver has been utilized to obtain the spectra of t and r; then, a standard method [2] has been used first for obtaining n and η, and then μ and ϵ. The PFEBI results are confirmed by two other solvers. Since $\mu' < 0$ and even $n' < 0$ are claimed by Grigorenko in green light, the spectra shown in Fig. 2 are centered. In our simulation model shown in Fig. 1(a), the dimensions, $a = 400$ nm, $s = 140$ nm, $d = d_2 - d_1 = 110$ nm, and $h = 90$ nm, are taken directly from [1,2], while $d_1 = 93$ nm and $d_2 = 127$ nm are obtained using the averaging of selected measurements of zoomed micrographs and our estimates of possible slopes in e-beam lithography. The simulation takes into account the substrate; experimental optical constants of bulk Au are used for the nanopillars [4]. While $R(\lambda)$ for the measured, best fit, and simulated results agree relatively well [Fig. 2(a)], further comparison indicates a strong difference between the values of the effective parameters [Figs. 2(b)–2(d)].

The major ambiguity is that neither the phase changes of the reflected light nor the transmission spectra (nor phase changes in transmission) are considered in the GM [1,2]. Thus, $R(\lambda)$ alone is believed to carry sufficient information for obtaining the complex μ and ϵ. Further, an "effective optical thickness" is used throughout both papers instead of the physical thickness normally taken in any homogenization. Since the effective optical thickness is much smaller than the actual, physical thickness (e.g., 12 nm versus 90 nm; see Supplementary Information, p. 3 in Ref. [1]), any resonant effects due to this effective thickness are much stronger and leave any sample almost no chance of deviating from the behavior most sought after by the author(s) in [1,2].

Possible artifacts of Eq. (1) are not discussed anywhere in [1,2]. Specifically, the knowledge of the complex μ and ϵ obtained from $R(\lambda)$ suggests that an unknown spectrum of $T(\lambda)$ also could be restored. We also compare the "restored" $T(\lambda)$ calculated using the six original fitting parameters and the "effective thickness" with the result obtained for the same sample using our simulations and the standard restoration technique. The restored $T(\lambda)$ exhibits unusual behavior; any magnetic resonance due to localized plasmonic effects is expected to accompany relatively sharp enhanced absorbance at the resonance. Regrettably, this is not the case for the restored $T(\lambda)$. Unfortunately, our simulations also show that the retrieval of the effective permeability (μ') and permittivity (ϵ') for the glycerine-covered sample [1] is also inconclusive, and the claim for a negative refractive index in the visible [2] is not well grounded. The disagreement is clearly shown in Figs. 3(a) and 3(b).

To further illustrate the misleading "reflectance best-fit concept," we have made an experiment using a standard absorbing filter with a reflectance spectrum similar to the nanopillars sample as shown in Ref. [5]. Amazingly, according to GM the standard glass filter could also show $n' < 0$ and $\mu' < 0$.

In summary, we demonstrate that it is doubtful that a sample with the geometry and materials proposed in [1,2] is capable of exhibiting either $\mu' < 0$ or $n' < 0$. It also seems that even $\epsilon' < 0$ at 520–570 nm is questionable for this sample either with or without a layer of glycerine.

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