Supporting Information for Gyroid Optical Metamaterials: Calculating the Effective Permittivity of Multidomain Samples

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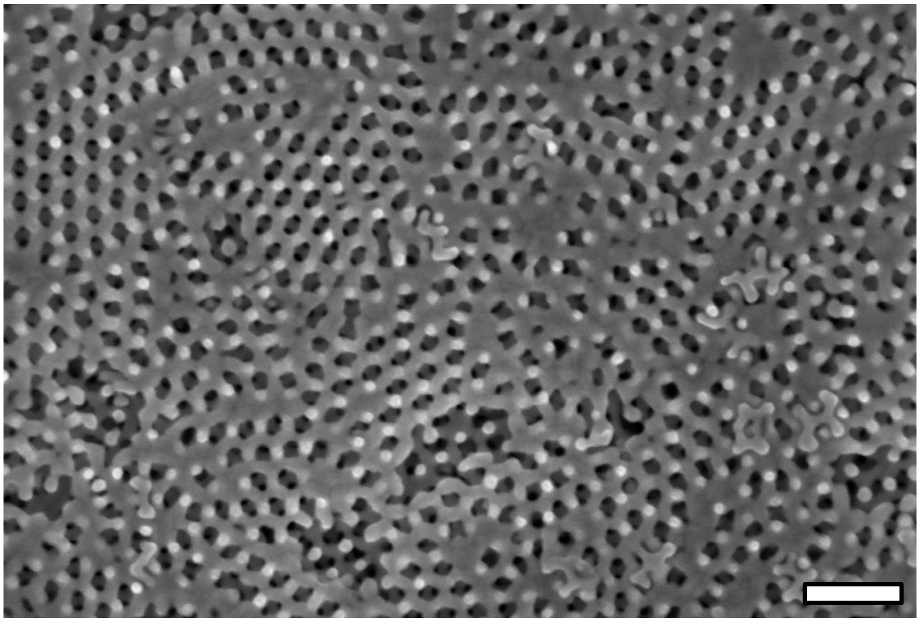

Figure S1: **Small gyroid metamaterial domains.** Electron micrograph of gold gyroid metamaterial sample (top view) highlighting the small size and random orientation of the domains (scale bar: 200 nm).

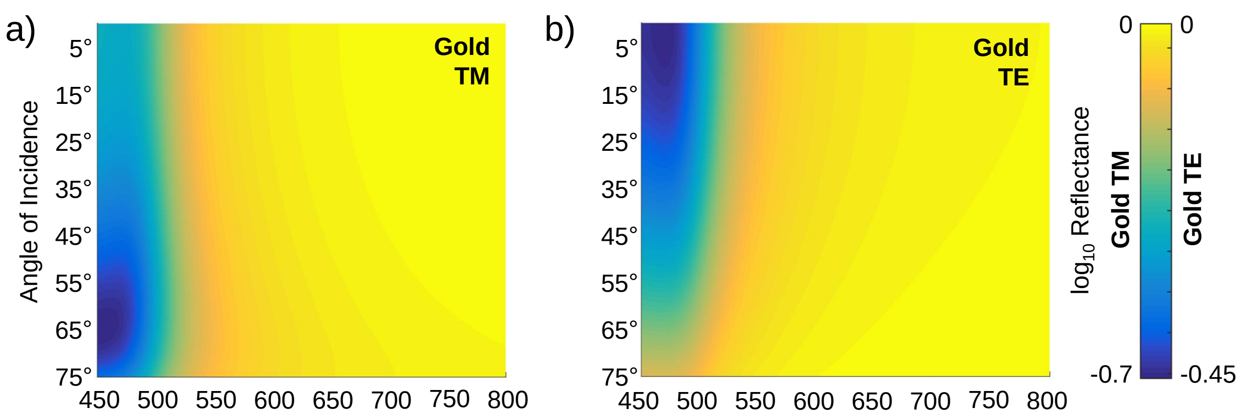

Figure S2: **Reflectance spectra of pure gold.** a) Reflectance spectra of pure gold as a function of angle of incidence calculated from $\varepsilon_{\mathrm{Au}}(\omega)$ for transverse magnetic (TM) polarized light (log colour scale extends from -0.7 to 0). Tabulated data for the permittivity of bulk gold is that found in Olmon et al[1]. b) The same for transverse electric (TE) polarized light (log colour scale extends from -0.4 to 0).

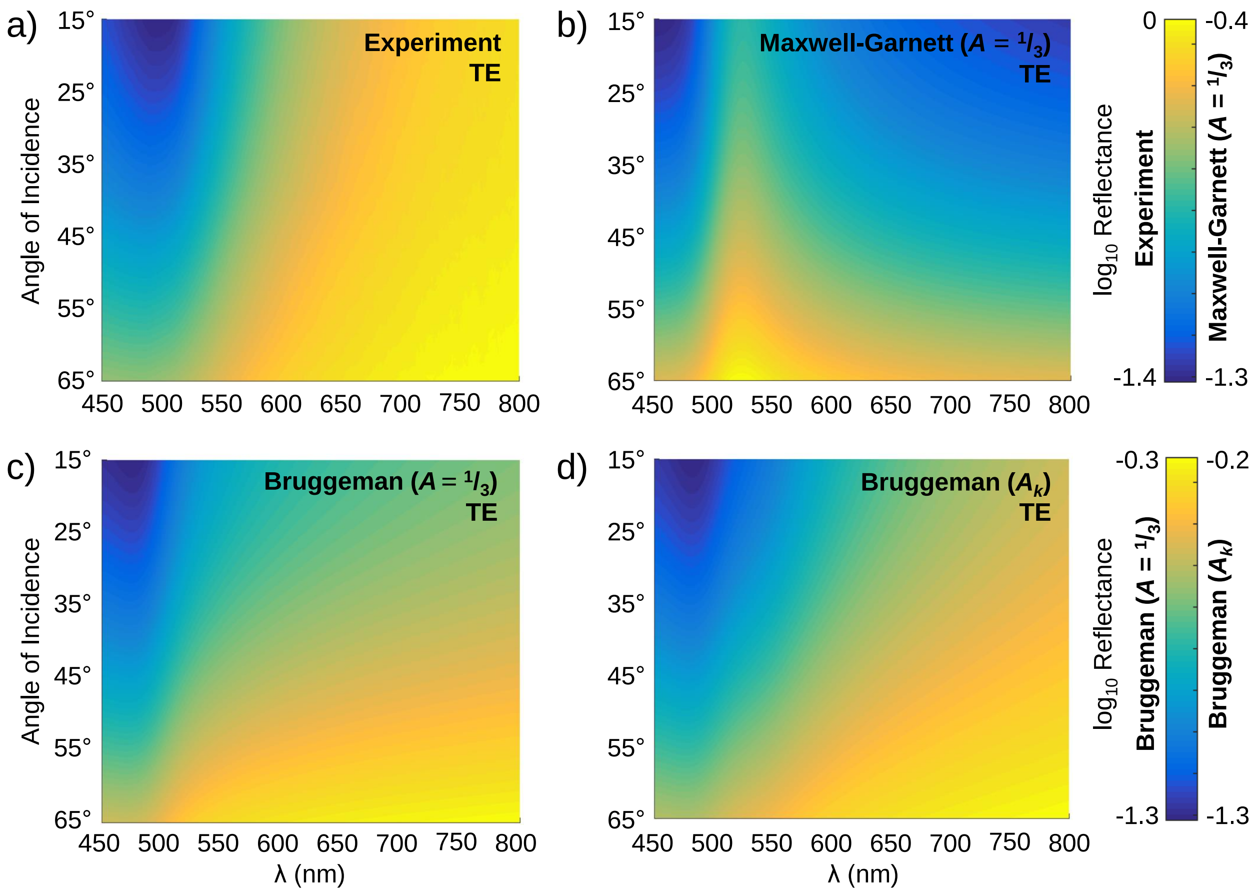

Figure S3: **Comparison of measured and modelled TE reflectance spectra as a function of angle of incidence using three effective medium theories.** a) The measured reflectance spectra for transverse electric (TE) polarized light (log colour scale extends from -1.4 to 0). b) The reflectance spectra generated using the Maxwell–Garnett theory (log colour scale extends from -1.3 to -0.45). c) The same generated using the Bruggeman theory and $A = 1/3$, i.e. spherical inclusions (log colour scale extends from -1.3 to -0.3). d) The same generated using the Bruggeman theory and $A_1 = 0.66$, $A_2 = 0.34$ and $A_3 = 0$. i.e. ellipsoidal inclusions (log colour scale extends from -1.3 to -0.2).

Bruggeman Effective Medium Theory

We here present a brief derivation of Equation 3 in the main manuscript. Consider the effective permittivity $\bar{\varepsilon} : \mathcal{V} \to \mathrm{GL}(2, \mathbb{C})$ of a homogenised metamaterial within a domain $\mathcal{V} \in \mathbb{R}^3$ with volume V defined by the equation

$$\langle \vec{D} \rangle = \bar{\varepsilon} \langle \vec{E} \rangle \ , \tag{1}$$

where $\langle f(\vec{r}) \rangle = \int_{\mathcal{V}} d^3 r \, f(\vec{r})/V$ is the volume average, and the electric field $\vec{E}$ and the displacement field $\vec{D} := \varepsilon \vec{E}$ are considered functions $\mathcal{V} \to \mathbb{C}^3$. Further assume that the local permittivity $\varepsilon(\vec{r})$ is piecewise constant, that is it takes the value ε_n within a subdomain $\mathcal{V}_n \subset V$ ($\mathcal{V} = \sum_n \mathcal{V}_n$, $i = 1, 2, \ldots, N$).

A short calculation reveals that

$$\langle D_i \rangle = \sum_n \varepsilon_n f_n \langle E_i \rangle_n \ ,$$

where we take some vector component D_i of $\vec{D}$, $\langle \cdot \rangle_n$ denotes the average over $\mathcal{V}_n$, and $f_n = V_n/V$ is the fill fraction of the respective subregion. Substitution into Equation 1 thus yield a general expression for the effective permittivity:

$$\bar{\varepsilon}_{ij} = \sum_n \varepsilon_n f_n \frac{\langle E_i \rangle_n}{\langle E_j \rangle} = \sum_n (\varepsilon_n - \varepsilon_m) f_n \frac{\langle E_i \rangle_n}{\langle E_j \rangle} + \varepsilon_m \delta_{ij} \ . \tag{2}$$

For a two-component system, this is evidently equivalent to

$$(\bar{\varepsilon} - \varepsilon_1) \, f_1 \frac{\langle E_i \rangle_1}{\langle E_j \rangle} + (\bar{\varepsilon} - \varepsilon_2) \, f_2 \frac{\langle E_i \rangle_2}{\langle E_j \rangle} = 0 \ . \tag{3}$$

This equation is exact. However, the contribution of the average field in domain n to the total average is unknown. In the following, three approximations are made:

- that all single connected regions within each subdomain are small compared to the effective wavelength, i.e. each diameter d satisfies $\lambda_0 \gg \bar{\varepsilon} d$, where λ_0 is the vacuum wavelength of the light,

- that the domains are small compared to the system size, i.e. V is large compared to d in any direction of space,
- and that all subdomains can be replaced by a monodisperse arrangement of ellipsoids of arbitrary direction.

We note that these assumptions are both in contradiction to the gyroid metamaterial geometry which is comprised of two interpenetrating domains of infinite extend, and can never truly be realised any physically realistic geometry. The resulting effective medium equation is nevertheless widely applied and provides the necessary degrees of freedom to provide a reasonable fit to our experimental data.

Consider a single closed subdomain of region 1 of ellipsoidal shape, embedded within the metamaterial of permittivity $\bar{\varepsilon}$. Further assume that the average field $\langle E_i \rangle$ is given and due to the dimension mismatch can be used as a boundary condition at infinity for $V \to \mathbb{R}^3$. The corresponding electric field in the inclusion can be found analytically using the static Maxwell equations[2]:

$$\frac{E_k}{\langle E_k \rangle} = \frac{1}{1 + (\varepsilon_1/\bar{\varepsilon} - 1)\, A_k} \ .$$

Here, k labels the principal axes of the ellipsoid and A_k is the depolarisation factor defined in Equation 2. Converting this equation to an arbitrarily oriented ellipsoid (labelled s) with respect to the global coordinate system yields

$$\frac{E_{i,s}}{\langle E_j \rangle} = \sum_{k_s} \frac{\cos(\phi_{ik_s}) \cos(\phi_{k_s j})}{1 + (\varepsilon_1/\bar{\varepsilon} - 1)\, A_{k_s}} \ , \tag{4}$$

where angle ϕ_{ij} is the angle between the two axes i and j. For randomly oriented ellipsoids (i.e. assuming a uniform distribution of angles) we can now calculate the average field in domain 1 as

$$\frac{\langle E_i \rangle_1}{\langle E_j \rangle} = \frac{1}{N_s} \sum_s \sum_{k_s} \frac{\cos\left(\phi_{ik_s}\right)\cos(\phi_{k_s j})}{1 + \left(\varepsilon_1/\bar{\varepsilon} - 1\right) A_k} \approx \frac{\delta_{ij}}{2} \sum_k \frac{1}{1 + \left(\varepsilon_1/\bar{\varepsilon} - 1\right) A_k}. \tag{5}$$

In the last step, we approximated the sum over all inclusions by an integral. This is possible because the number of single inclusions N_s is large; a consequence of the second approximation above. Following the same procedure for domain 2 and substituting Equation 5 into Equation 3 yields Equation 3 in the main manuscript where $\bar{\varepsilon}$ is denoted ε_{eff}.

References

(1) Olmon, R. L.; Slovick, B.; Johnson, T. W.; Shelton, D.; Oh, S.-H.; Boreman, G. D.; Raschke, M. B. Optical dielectric function of gold. *Phys. Rev. B* **2012**, *86*, 235147.

(2) Stratton, J. A. *Electromagnetic Theory*; John Wiley & Sons, 2007.