Some Interesting Effects when Homogenizing Plasmonic Composites

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INTRODUCTION

The effective permittivity $\varepsilon_{\mathrm{eff}}$ of a composite is usually not the volume average of its constituents. The impact of the microstructure of the composite is important even for ordinary dielectric composites with modest contrasts between the constituents, and the effect can be dramatic for plasmonic composites with negative permittivity and small losses. In the idealized case, with lossless plasmonic inclusions ($\varepsilon_{\mathrm{i}} < 0$) in a dielectric environment ($\varepsilon_{\mathrm{e}} > 0$), we can theoretically get any real value for $\varepsilon_{\mathrm{eff}}$ due to plasmonic resonances. When realistic losses are taken into account, the plasmonic resonances are damped, but still the (real part of) $\varepsilon_{\mathrm{eff}}$ can be significantly larger than ε_{e} or smaller than ε_{i}. Moreover, if we consider the plane-wave transmission and scattering from a slab with such resonant inclusions, a straightforward retrieval of $\varepsilon_{\mathrm{eff}}$ yields reasonable results, but the simultaneously obtained effective permeability μ_{eff} seems unphysical.

In this presentation, we discuss these interesting effects using a geometrically very simple composite.

QUASISTATIC EFFECTIVE PERMITTIVITY USING MIXING FORMULAS

Consider a 2D composite with circular inclusions with permittivity ε_{i} occupying an area fraction p in an environment with permittivity ε_{e}. Many mixing formulas for estimating the effective permittivity $\varepsilon_{\mathrm{eff}}$ of such a composite are available [1], and two of the most popular and useful ones are the Maxwell Garnett formula

$$\varepsilon_{\mathrm{eff}} = \varepsilon_{\mathrm{e}} + \frac{2p\,\varepsilon_{\mathrm{e}}\,(\varepsilon_{\mathrm{i}} - \varepsilon_{\mathrm{e}})}{\varepsilon_{\mathrm{i}} + \varepsilon_{\mathrm{e}} - p\,(\varepsilon_{\mathrm{i}} - \varepsilon_{\mathrm{e}})}, \tag{1}$$

and the (symmetric) Bruggeman formula

$$\varepsilon_{\mathrm{eff}} = \frac{1}{2}\left(B \pm \sqrt{B^2 + 4\varepsilon_{\mathrm{i}}\varepsilon_{\mathrm{e}}}\right), \qquad B = \varepsilon_{\mathrm{e}} - \varepsilon_{\mathrm{i}} + 2p\,(\varepsilon_{\mathrm{i}} - \varepsilon_{\mathrm{e}})\,. \tag{2}$$

For ordinary dielectric mixtures, it is well known that the Maxwell Garnett formula is better suited for mixtures with well separated inclusions, while the Bruggeman formula is more accurate for random mixtures where clustering is allowed [1].

The difference between the estimates (1) and (2) can be significant for mixtures with $1 \le \varepsilon_{\mathrm{e}} \ll \varepsilon_{\mathrm{i}}$ but both estimates are within the interval $\varepsilon_{\mathrm{e}} \le \varepsilon_{\mathrm{eff}} \le \varepsilon_{\mathrm{i}}$ for all $0 < p < 1$. However, if one constituent has negative permittivity, say $\varepsilon_{\mathrm{i}} < 0$ and $\varepsilon_{\mathrm{e}} > 0$, the situation is qualitatively different. Then, the Maxwell Garnett formula predicts one plasmonic resonance at a certain p, while the Bruggeman formula gives a region with complex (lossy) effective permittivity although both ε_{i} and ε_{e} are real. One example is shown in Figure 1, and more details on the applicability of mixing formulas for plasmonic composites can be found in [2].

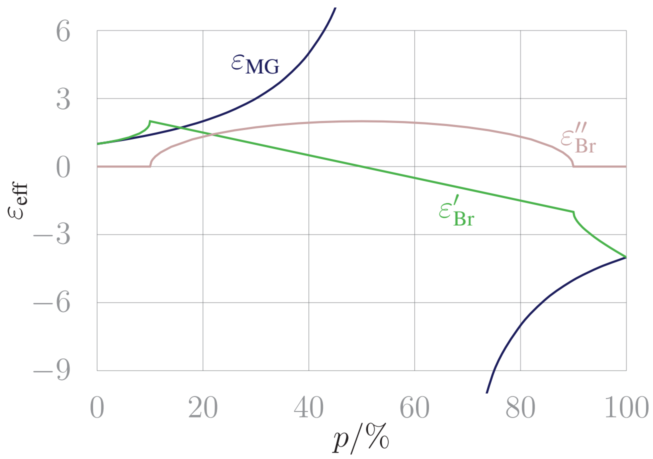

Figure 1: Quasistatic effective permittivity using the Maxwell Garnett (ε_{MG}) and Bruggeman ($\varepsilon_{\text{Br}} = \varepsilon'_{\text{Br}} - j\varepsilon''_{\text{Br}}$) formulas for a composite with cylindrical inclusions with $\varepsilon_{\text{i}} = -4$ occupying an area fraction p in air ($\varepsilon_{\text{e}} = 1$). The Maxwell Garnett formula predicts a plasmonic resonance at $p = 60\%$, while the Bruggeman formula predicts (quite strange) losses for $10\% < p < 90\%$.

The complex frequency dependent permittivity of many metals [3] (approximately) obey a Drude dispersion of the form

$$\varepsilon(\omega) = \varepsilon' - j\varepsilon'' = 1 - \frac{\omega_{\text{p}}^2}{\omega^2 - j\omega\nu}, \qquad \begin{cases} \omega_{\text{p}} = \text{plasma frequency}, \\ \nu = \text{damping frequency} \ll \omega_{\text{p}}, \end{cases} \tag{3}$$

where the real part ε' is negative below ω_{p}, while the imaginary part ε'' can be relatively small. Using a mixture with ε_{i} given by this Drude model, we get the Lorentz-dispersion

$$\varepsilon_{\text{eff}}(\omega) = 1 + \frac{\omega_{\text{p,eff}}^2}{\omega_{0,\text{eff}}^2 - \omega^2 + j\omega\nu_{\text{eff}}}, \qquad \begin{cases} \omega_{\text{p,eff}} = \omega_{\text{p}}\sqrt{p}, \\ \omega_{0,\text{eff}} = \omega_{\text{p}}\sqrt{(1-p)/2}, \\ \nu_{\text{eff}} = \nu, \end{cases} \tag{4}$$

for the effective permittivity using the Maxwell Garnett fomula (1). Choosing $\nu = \omega_{\text{p}}/100$ and the fairly small area fraction $p = 10\%$, we get the result shown in Figure 2. If the circular inclusions are arranged in a square lattice, this estimate is extremely accurate in the quasistatic limit compared with the series solution in [4].

S-PARAMETER RETRIEVAL

The probably most common way to determine the effective material parameters $\varepsilon_{\text{eff}}, \mu_{\text{eff}}$ for metamaterials is to simulate or measure the S-parameters for a slab, i.e., the reflection and transmission of a normally incident plane wave, and compute $\varepsilon_{\text{eff}}, \mu_{\text{eff}}$ assuming that the composite slab behaves as a homogeneous isotropic one [5, 6]. However, the validity (or accuracy) of this simple model has also been criticized [7, 8]

In this case, we take a five layer slab with the same composite as in Figure 2 and simulate the S-parameters using a 2D finite-element solver (COMSOL Multiphysics 3.5a). Assuming a square lattice of circular inclusions and scaling the unit cell side-length a differently compared with the plasma wavelength λ_{p} of the Drude model (3), we get the results in Figure 3. The retrieved ε_{eff} seems very reasonable and it converges, as expected, towards the quasistatic prediction with smaller unit cells compared with the wavelength.

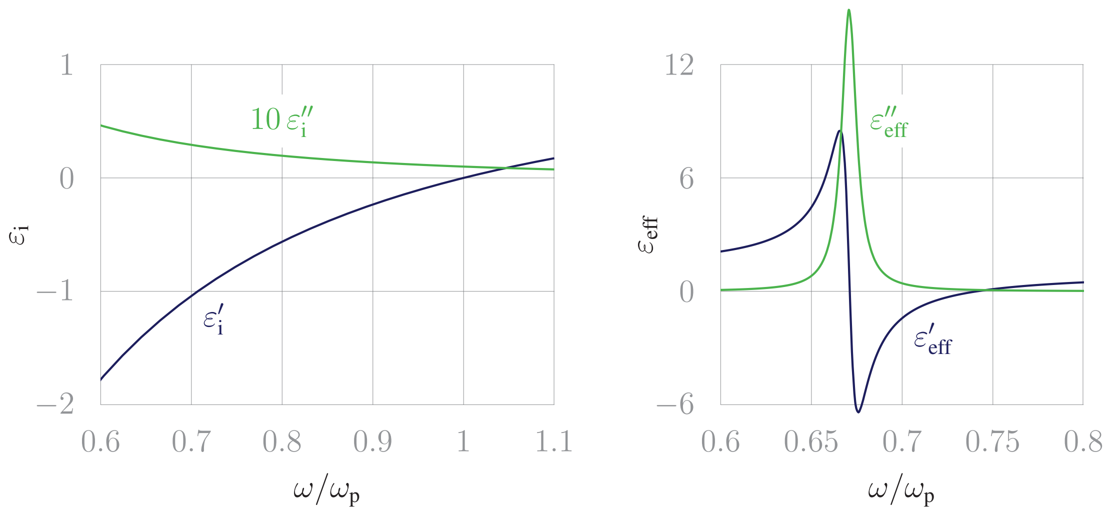

Figure 2: A composite with $p = 10\%$ cylindrical inclusions with the Drude dispersion on the left gives the effective permittivity with the Lorentz dispersion on the right using the Maxwell Garnett formula.

However, the retrieved μ_eff shows a similar kind of "anti-resonance" as many metamaterials with more complicated resonant inclusions. This is clearly not reasonable, and it suggests that the five-layer slab is not actually effectively homogeneous enough near the (physically perfectly reasonable) plasmonic resonance. Indeed, comparing the oblique reflection and transmission for the composite slab with the homogenized one reveals that the retrieved ε_eff and μ_eff are not very accurate.

CONCLUSIONS

A composite with plasmonic inclusions with realistic losses can have an effective quasistatic permittivity with a strong resonance at some frequency. However, a slab of the composite does not necessarily behave as a homogeneous one. The problems in the homogenization of the above presented composite slab are actually very similar to the ones in many contemporary metamaterial-realizations, and so this geometrically very simple composite can be useful as a benchmark problem.

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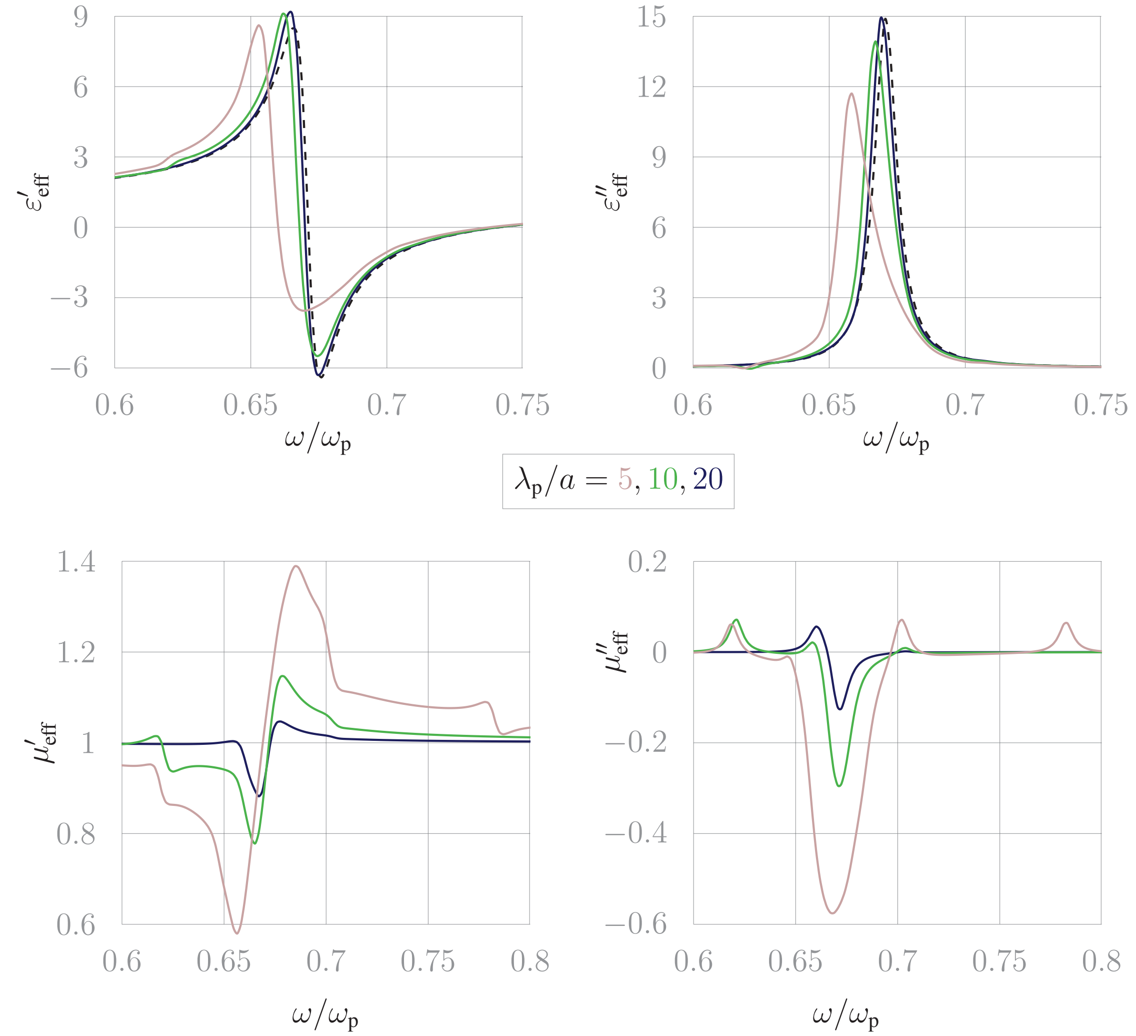

Figure 3: Effective permittivity $\varepsilon_{\text{eff}} = \varepsilon'_{\text{eff}} - j\varepsilon''_{\text{eff}}$ (top) and permeability $\mu_{\text{eff}} = \mu'_{\text{eff}} - j\mu''_{\text{eff}}$ (bottom), retrieved from simulated S-parameters. The permittivity seems reasonable and converges towards the quasistatic prediction (dashed line), but the permeability shows an unphysical "anti-resonance" in addition to weak Fabry–Pérot resonances.

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