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SHAPE OPTIMIZATION OF NANOPARTICLES FOR OPTICAL METAMATERIALS

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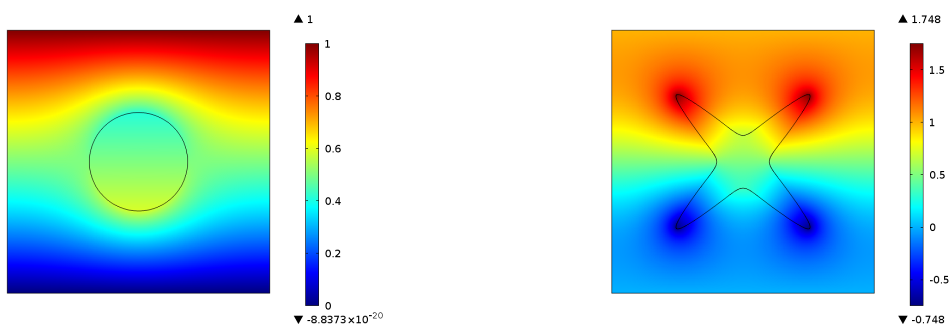
Abstract. A composite comprised of small particles distributed in a host matrix can be prescribed effective electromagnetic properties (ϵ_{eff}, n_{eff}, etc.) in the long wavelength limit. The effective properties are functions of the constituent material properties, volume fraction of the particles and also the shape of the particles. By optimizing the particle shape and tapping into the plasmonic resonance at the particle/matrix interface, we design composites with near-zero permittivity and very low loss at optical frequencies.



(a) $\epsilon_{\text{eff}} = 3.13 + 0.02i$

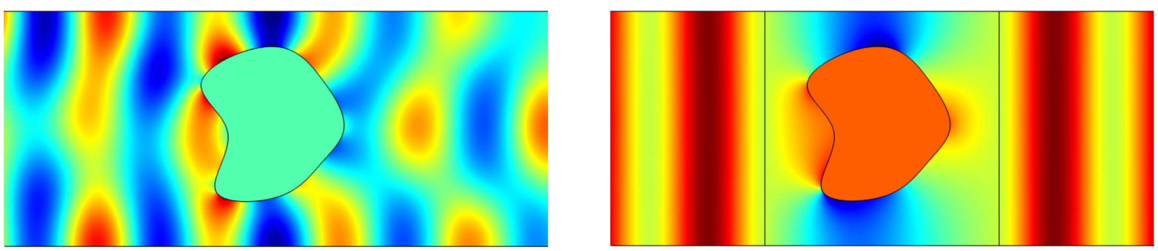
(b) $\epsilon_{\text{eff}} = -2.05 + 1.11i$

Figure 1: Small silver particles in a dielectric background, illuminated with light at 500 THz. The local electric potential field, as well as the effective permittivity ϵ_{eff} of the composite, is greatly influenced by particle shape.

1 INTRODUCTION

In recent years, there has been great interest in the design and manufacture of composites which possess specific electromagnetic properties. This is largely due to the experimental demonstration of a composite possessing negative refractive index in 2000 [1, 2]. This seminal work confirmed that judicious choice of the topology within a composite could result in that composite having effective material properties much different to those of its constituents. These so-called *metamaterials* have since been proposed for such extraordinary devices as the invisibility cloak [3] and artificial black hole [4].

The electromagnetic properties we are typically interested in controlling are the electric permittivity ϵ and magnetic permeability μ. Note that both are typically complex-valued and are functions of the frequency of light with which the material is interacting. A special class of materials, called Epsilon-Near-Zero (ENZ) materials, have been shown to be capable of producing a number of interesting phenomena, such as waveguides which maintain efficiency even in the presence of sharp corners [5] antennas with extremely high directivity [6], electromagnetic tunnelling [7] and even total reflection/transmission (a type of perfect cloaking) under some conditions [8, 9]. As the name suggests, ENZ materials are those with electric permittivity $\epsilon \approx 0$.



(a) Metal inclusion in vacuum (b) Metal inclusion in ENZ material

Figure 2: Light incident on an arbitrarily-shaped metal object produces a large degree of scattering (left). When the same object is submerged in a matched ENZ material, the incoming light waves are transmitted and remain planar; the object is invisible (right).

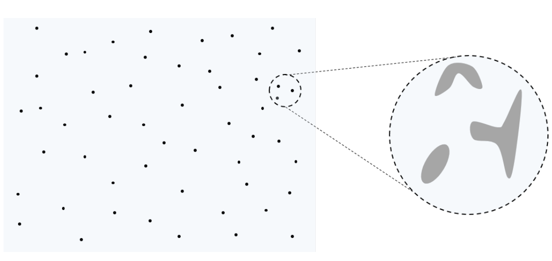
It is therefore desirable for us to attain materials which have ENZ behaviour. It is known that some single-phase (i.e. non-composite) materials, such as LiF and KCl, exhibit ENZ over part of the frequency spectrum [10, 11]. These *polaritonic* materials suffer two drawbacks: the frequency at which ENZ behaviour occurs cannot be changed, and the energy losses associated with using a specific material cannot be reduced. Thus, using a single-phase material to produce ENZ behaviour is possible in some sense, but in order to achieve prescribed/tunable ENZ behaviour (at a specific frequency and with a certain degree of loss), it seems we must explore other options.

The metamaterials research field has demonstrated one way of producing tunable ENZ behaviour- that is to induce electric resonance within a composite typically comprised of a dielectric matrix and inclusions with dimensions comparable to the incident wavelength of light. The electric resonance stems from either large induced currents (in the case of conductor/wire inclusions) [12] or from Mie scattering effects (in the case of spheroid inclusions) [13]. This method of producing ENZ behaviour has some problems, namely the bandwidth over which ENZ behaviour is attained may be too narrow to admit practical application, and the losses associated with these types of resonance may be very high. The interested reader is directed to the studies [14, 15, 16], which are optimization studies related to this type induced current/Mie resonance composite.

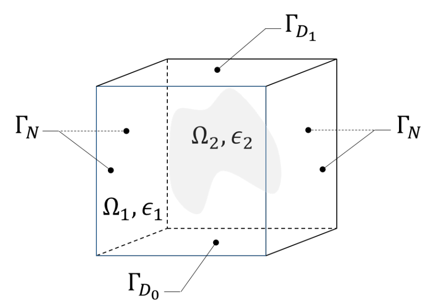
An alternate route to ENZ behaviour is via very small particles distributed in a host matrix (Figure 3a). If the particle and host materials have opposite signs of permittivity (such as silver and glass), local field enhancements refered to as *plasmonic resonance* can develop at the material interfaces. This type of resonance remains present in the long wavelength limit, and so exotic behaviour can be achieved for any wavelength which is sufficiently larger than the inclusions.

There has been extensive work devoted to the forward-problem of calculating the effective permittivity of these particle/matrix composites, given the constituent materials and particle topology. For simple sphere and ellipsoid particles, we have the classical Maxwell-Garnett and Bruggeman theories [17, 18]. More recently, a numerical technique for predicting the effective permittivity for arbitrary shapes was proposed [19] and used to analyse a number of complicated particle topologies including n-gons and Sierpinski shapes [20]. To the authors' knowledge, though, optimization of particle shape in order to obtain ENZ material behaviour has not yet been explored.

In the remainder of this paper, we will show that ENZ behaviour can indeed be induced in the long wavelength limit by optimizing the shape of the particle inclusions. We begin by detailing the procedure required to determine the effective properties of a particle/matrix composite. We continue by describing our chosen optimization algorithm. We conclude by applying the technique to silver particles distributed in a dielectric matrix, and also vice-versa.



(a) Composite comprised of small particles embedded in a host matrix



(b) Model for the determination of ϵ_{eff}

Figure 3: The behaviour of a particle/matrix composite as a whole can be determined from analysis of a single particle.

2 EFFECTIVE PROPERTIES FOR ARBITRARILY-SHAPED PARTICLES

In the long wavelength limit, the effective permittivity ϵ_{eff} of a matrix/particle composite can be retrieved via electrostatic analysis of a single particle, using the following [19]:

$$\epsilon_{\text{eff}} = \frac{1}{a\Delta u \Delta u^*} \int_{\Omega_1 \cup \Omega_2} \epsilon \nabla u \cdot \nabla u^* \ .dV \tag{1a}$$

so that

$$\nabla \cdot (\epsilon_i \nabla u) = 0 \qquad \text{in } \Omega_i \tag{1b}$$

$$u = u_1 \qquad \text{on } \Gamma_{D_1} \tag{1c}$$

$$u = u_0 \qquad \text{on } \Gamma_{D_0} \tag{1d}$$

$$\nabla u \cdot n = 0 \qquad \text{on } \Gamma_N \tag{1e}$$

$$(\epsilon \nabla u_1 - \epsilon_2 \nabla u_2) n_2 = 0 \qquad \text{on } \partial\Omega_1 \cap \partial\Omega_2 \tag{1f}$$

In the above, u is the potential field (voltage) and the geometry is as shown in figure 3b. $\Delta u = u_1 - u_0$ and a is the side length of the analysis cube.

Obviously, if the particle topology is non-symmetric, the effective properties may have to be evaluated for each of the x, y, z directions, and ϵ_{eff} becomes a tensor. In this work, we will analyse two-dimensional particles and constrain the designs to be double-symmetric. Thus, the scalar form of ϵ_{eff} is valid.

For our constituent materials, we choose silver, for which the properties are well known in the optical spectrum [21], and a simple dielectric with $\epsilon_{\text{di}} = 2.3$, which is common in the literature. We arbitrarily nominate to undertake our optimization studies assuming an incident light frequency of 500 THz (mid-optical spectrum), where the silver permittivity is in the order of $\epsilon_{\text{Ag}} = -14.0 + 0.8i$. Since our assumptions require the composite inclusions to be much smaller than the incident wavelength, it is implied that inclusions our inclusions must have dimensions in the nanometre range.

3 SHAPE OPTIMIZATION

As discussed earlier, this research is targeting ENZ materials. Recalling that ϵ_{eff} is generally complex-valued, we want both the real and imaginary parts to be as close to zero as possible. Accordingly, the objective function $J : \Omega \to \Re$ we choose is:

$$J = (\epsilon_{\text{eff}}\ \epsilon^*_{\text{eff}})^{1/2} \tag{2}$$

We have now set the stage for our optimization problem. We aim to determine the particle shape such that we obtain the desired effective medium properties, which are evaluated via equations (1) and (2).

There are a number of topology optimization techniques available for the above class of problem. The SIMP method [22], popular in structural mechanics, was deemed less suited to this problem, since the electric field ∇u (along with any topological sensitivity information) tends to vanish inside metals (Ω_2). The level set method [23] was also considered, however the shape derivative on $\partial\Omega$ is awkward to evaluate for the discontinuous ∇u field at the interfaces. Also since the problem is resonant in nature, any gradient-based method can be unstable and/or tend to direct designs in the opposite direction to the optima (see, for example [15]).

For the above reasons, we decided to parametrize the geometry and use a non-gradient optimization method. The so-called *Superformula*, first proposed by Gielis [24], is an extension to the superellipse formulae and is capable of producing a variety of shapes using only a small number of tuning parameters, as demonstrated in figure 4. In two dimensions, the formula reads

$$r(\theta) = \left(\left| \frac{\cos(m\theta/4)}{a} \right|^{n_2} + \left| \frac{\sin(m\theta/4)}{b} \right|^{n_3} \right)^{-1/n_1} \tag{3}$$

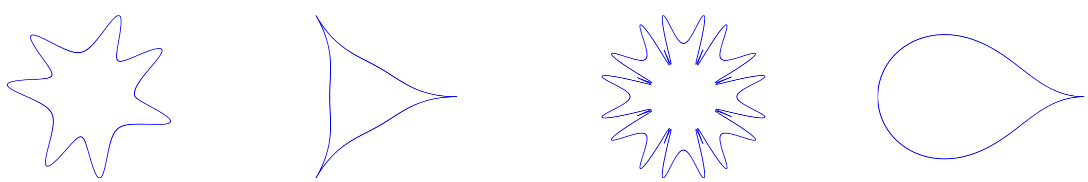


Figure 4: Some shapes generated with the Gielis Superformula

As stated earlier, in order to simplify things, we work with double-symmetric shapes and so we set $a = b$ and $n_2 = n_3$ in the above. Even on this simplification, (3) can still produce a diverse set of shapes.

In order to more easily constrain the size of the particles, we use a modified version of (3), where we set $a = b = 1$, and we introduce a zooming factor z_f which constrains the maximum particle dimension in the $x-$ and $y-$ directions. The design vector for this problem is thus reduced to $(m,\ n_1,\ n_2, z_f)^T$, and we developed a particle swarm (PSO) algorithm in order to solve it.

The PSO algorithm was based on the theory of [25], and included a regrouping feature as per [26] in order to prevent stagnation at local optima. For completeness, we state that for all the results reported in the sequel, the cognitive and social factors of the algorithm were set to 1.5, inertia weight was set to 0.6 and a swarm population of 30 was used.

4 RESULTS AND DISCUSSION

The results of the optimization procedure are illustrated in Figure 5. Note that some of the topologies shown comprise a background of dielectric with silver inclusions, whereas others comprise a silver background with dielectric inclusions.

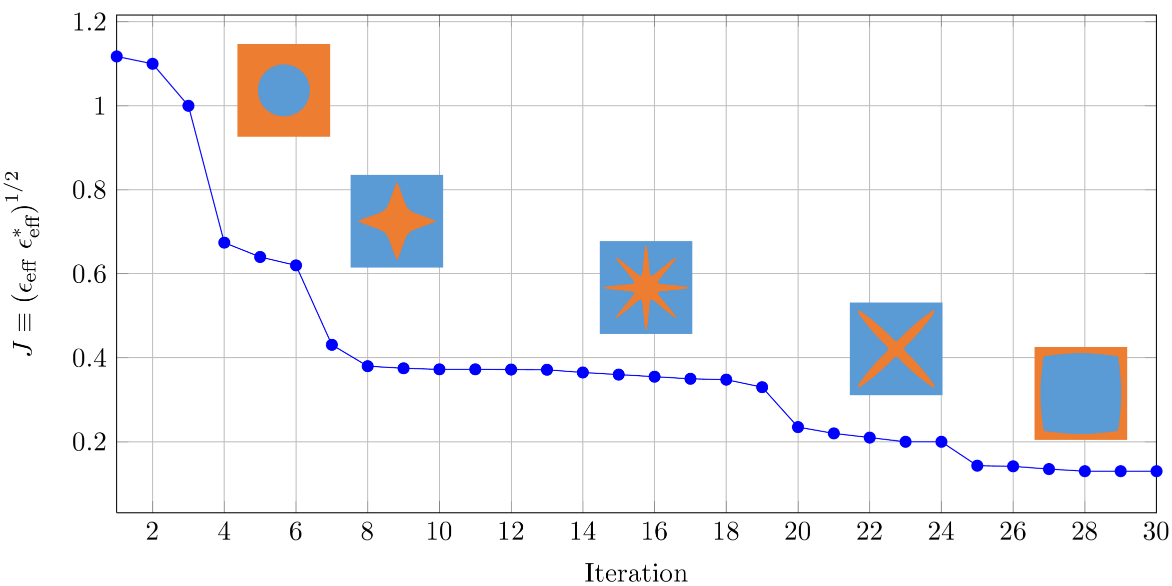


Figure 5: Convergence history of the optimization algorithm. Note that blue indicates dielectric; orange indicates silver.

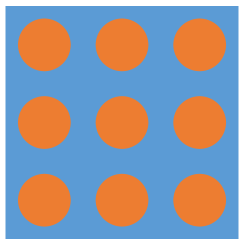
The frequency response of two of the optimized designs, as well as a simple design for comparison, is shown in Figure 6. The comparative design (Fig. 6a and 6b) possesses a resonant-type frequency response typical of metamaterials, and reminds us that simple-shaped metallic inclusions are capable of producing ENZ behaviour, though typically accompany large losses, indicated by high values of ϵ''_{eff}.

The optimized topologies shown in Fig. 6c - 6f also have ENZ behaviour, though with greatly reduced loss. Interestingly, the frequency response for these designs is not of the resonant-type; rather they appear akin to Drude curves. Such designs may prove very useful in applications where exotic behaviour is required over a broad range of frequencies.

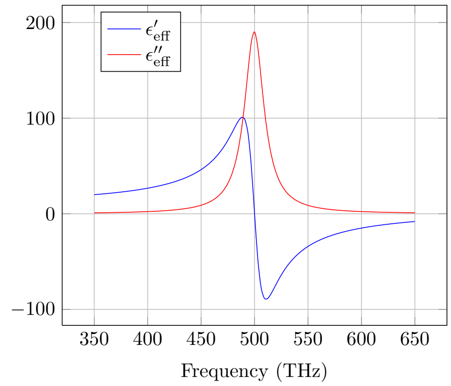
5 CONCLUSION AND OUTLOOK

We have shown that by applying shape optimization techniques to the shape of particles distributed in a matrix, the composite as a whole can possess an electric permittivity near zero. Compared with simple shapes, the optimized composites demonstrate greatly reduced energy loss, and have a much more stable frequency response. Accordingly, we believe such composites may find application in devices which require low-loss ENZ behaviour over a broad range of frequencies.

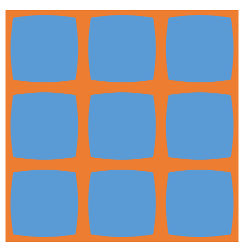
It remains to be explored how the internal topology of the particles could affect the composite performance. In addition, manufacturability constraints should be incorporated, in an attempt to realise a practical amount of the proposed materials. Such will be the focus of future studies.



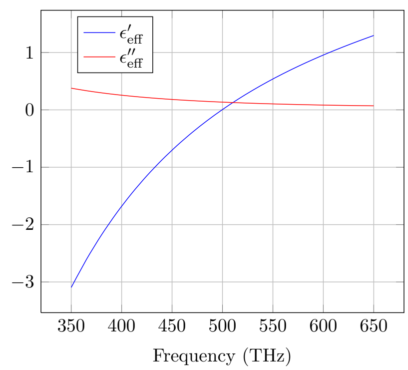
(a) $\epsilon_{\text{eff}} = 0.00 + 190i$ ($J = 190$)



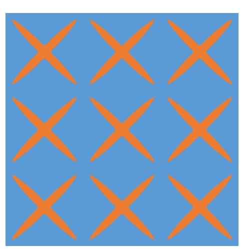
(b)



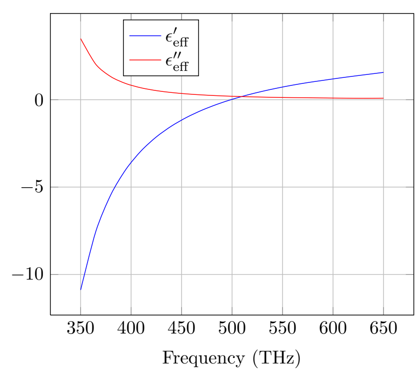
(c) $\epsilon_{\text{eff}} = 0.01 + 0.13i$ ($J = 0.13$)



(d)



(e) $\epsilon_{\text{eff}} = 0.02 + 0.20i$ ($J = 0.20$)



(f)

Figure 6: Frequency response of (a-b) Simple comparison shape (c-d) Optimized dielectric inclusions (e-f) Optimized silver inclusions. The optimized designs demonstrate ENZ behaviour with very low loss.

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