Optimizing Scattering Coefficients of Disordered Metamaterials Using the Finite-Difference Time-Domain Method

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***Abstract*—A technique based on the finite-difference time-domain method is presented for calculating optical transmission and reflection of disordered nanoparticle stacks. The approach is used to optimize a nanoparticle stack exhibiting greater than 98% average absorption over the visible spectrum.**

I. Introduction

Most electromagnetic metamaterials consist of a repeating unit cell with sub-wavelength features designed to impart a specific scattering response in a particular frequency range [1]. Metamaterials can be designed to achieve permittivities and permeabilities not found in natural materials, and these exotic material parameters can then be used to design metalenses, cloaks, mirrors and absorbers. For the case of highly absorbing metamaterials, many applications require low reflection and transmission over a broad wavelength range and over a wide range of incident angles. However, ordered metamaterials typically provide high absorption over only a limited wavelength range and over a limited range of incident angles. Alternatively, the use of disordered metamateirals has become an interesting alternative for achieving high absorption over extremely large bandwidths and incident angles [2]. However, the lack of long range order makes their analysis and design more challenging. In this work we describe our progress on obtaining reflection and transmission coefficients in disordered metamaterials consisting of dense agglomerations of gold nanoparticles using the finite-difference time-domain method (FDTD) [3], [4].

II. Dense Nanoparticle Stacks

In addition to impressive absorption, disordered metamaterials are amenable to low-cost solution-based fabrication techniques. Our approach begins with synthesis of gold nanoparticles in solution using the Turkevitch method in which nanoparticle size is controllable. In our work diameters between 5 nm and 45 nm are used. A single drop of the nanoparticle solution is deposited in a glass well using a micropipette and allowed to dry. Then a second drop is applied and allowed to dry, and this process is repeated until an opaque layer is formed, which typically requires 15-20 drops. This process is schematically shown in Fig. 1 (a), and fabricated samples are shown in Fig. 1 (b). After each drop, the reflection and absorption is measured using the configuration shown in Fig. 1 (c).

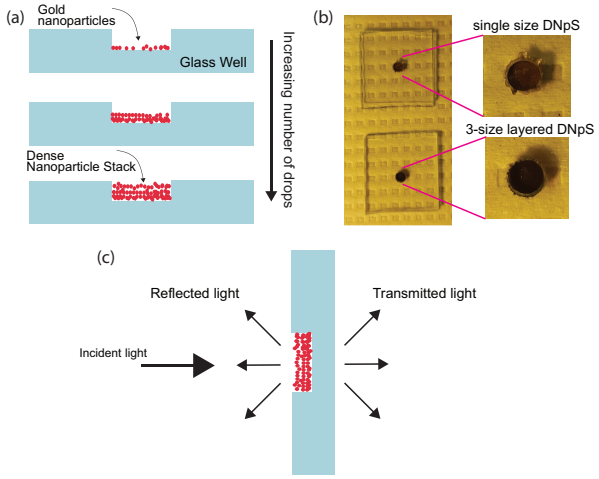

Fig. 1. (a) Experimental process for fabricating layer-by-layer dense nanoparticle stacks. The substrate is borosilicate glass, and the red spheres represent gold nanoparticles. (b) Photographs of fabricated samples. Single size DNpS indicates that nanoparticles of only a single size were used (diameter = 18 nm); whereas 3-size DNpS indicates that nanoparticles of three different sizes were used to construct the DNpS (diameters = 12, 18 and 48 nm). (c) Experimental arrangement for measuring reflection and transmission.

III. Numerical Approach

To gain further insight into the electromagnetics of our metamaterials, the reflection and transmission coefficients were determined numerically using FDTD. If the nanoparticles had been placed in an ordered arrangement, then a minimum-sized repeating unit cell can be employed to minimize computational demands. However, in our disordered materials, this approach cannot be applied directly. Therefore, we defined a unit cell that contained a large set of randomly distributed particles, and then applied periodic boundary conditions on this disordered supercell. The scheme is depicted in Figs. 2 (a)-(b).

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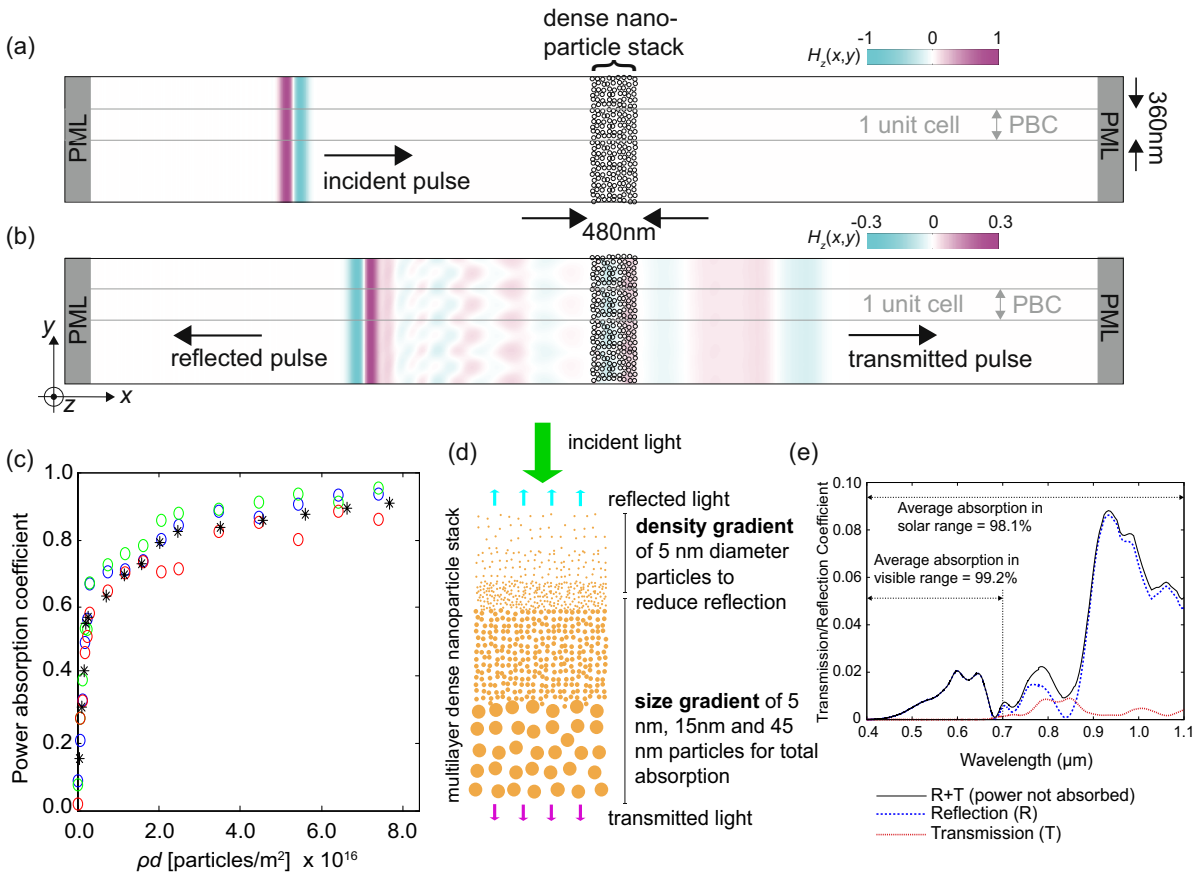

Fig. 2. (a) Computational domain depicting incident pulse on a dense nanoparticle stack. Periodic boundary conditions (PBC) are implemented along the y-direction. Four unit cells are depicted concatenated along y. The boundaries along the x-direction are terminated with perfectly matched layer (PML) absorbing boundary conditions. (b) Same as (a) but at a later time when the pulse has scattered from the dense nanoparticle stack. Reflected and transmitted field is visible. (c) Comparison between numerically calculated and experimentally measured absorption as a function of ρd where ρ is particle density and d is sample thickness. (d) Particle arrangement for structure optimized for greater than 98% absorption. (e) Numerically calculated scattering spectra for optimized structure. Coefficients in (c) and (e) are unitless power coefficients. (c)-(e) reproduced from [4], with the permission of AIP publishing.

A two-dimensional implementation of FDTD was used, so the gold nanoparticles are actually infinitely long cylinders. We deliberately used the TM$_z$ polarization, so that the electric field would be perpendicular to the axis of the cylinder. In this sense, one can consider a cylindrical scattering cross section σ_c per unit length of cylinder along z. Then in order to obtain the approximate scattering cross section for a cylinder of finite length l, multiply σ_c by l. In order to compare the 2D FDTD results to the three-dimensional experimental results, we assume $\sigma_s \approx \sigma_c 2r\delta$ where σ_s is the spherical cross section of the nanoparticles, r is their radius, and δ is an empirical fitting factor. Fig. 2 (c) shows a comparison between the absorption measured experimentally and calculated using our 2D approach with a fitting factor of $\delta = 0.29$. The agreement is very good.

The data shown in Fig. 2 (c) corresponds to a sample with engineered disorder. We intentionally deposited nanoparticles of three different sizes with the largest particles at the exit side of the sample, and the smallest particles on the incident side. Intuitively, the small nanoparticles scatter less, so small particles on the front of the sample will reduce reflection while larger particles at the back will reduce transmission. The photograph in Fig. 1 (b) shows that the 3-size layered DNpS is indeed darker to the naked eye than the single size DNpS.

This gradient approach was further explored using our numerical approach in which we incorporate both a density gradient and a size gradient as depicted in Fig. 2 (d). Our optimized structure exhibits better than 98% absorption averaged over the entire visible spectrum as shown in Fig. 2 (e).

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