Numerical Investigations of Second Harmonic Generation in Metamaterials based on Metallic Nanowires

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Abstract: This paper presents a comprehensive theoretical and numerical study of the second harmonic generation (SHG) in a metamaterial consisting of, parallel, infinite, cylindrical nanowires made of centrosymmetric materials. We use a numerical algorithm based on the multiple scattering method to retrieve the field profiles at both the fundamental frequency (FF) and second harmonic (SH). The algorithm takes into account the contributions of both the surface and the bulk nonlinear polarizations. The mathematical formalism behind the theoretical approach and its numerical implementation arediscussed and typical results for various cylinder configurations are shown.

1 Introduction

The research field of metamaterials has seen a fast development in recent years, which has lead to a radical new approach to the study of the electromagnetic properties of materials. Due to advances in the nanotechnology, we can now safely say that artificial materials based on resonant structures can be created to encompass structures of size comparable to optical wavelengths in the visible spectrum, or smaller. It has been proven that these “artificial atoms” behave in ways similar to the real atoms inside a crystalline material. Indeed, research so far has shown the presence of magnetic moments [1], electrical polarizability [2] and also non-linear optical properties such as second-order non-linear optical response [3]. It is clear then, that a better understanding of these phenomena can lead to a new class of artificial materials that can be tailored to the exact needs of a given application. In order to achieve this, a solid theoretical body of knowledge is needed to both describe the optical effects observed so far in these structures as well as to facilitate the exploration of this new area of electromagnetics.

Due to the fact that most metals are centrosymmetric (*i.e.* they are invariant to the inversion symmetry transformation), it is important to develop a theory that describes the SH generation (SHG) by structures made of such materials. To this end, in a first step, the field at the FF must be rigorously calculated. While some progress has been made towards this end by examining the SHG from low-index contrast cylinders [4] or, for a more general case, a single metallic cylinder [5], these methods ignore the wave scattering at the FF or, respectively, cannot be applied to more than one scatterer. To overcome these drawbacks, we employ a method based on the multiple scattering matrix (MSM) algorithm which allows the treatment of an arbitrary array of cylinders with a high index contrast. Due to its algebraic nature, the method also permits ready field retrieval at any given point.

The work described in this paper focuses on a specific type of metamaterial that has shown to be of importance in the overall understanding of non-linear effects in metamaterials, specifically a distribution of metallic nanoparticles. In this configuration, the constituent structures have shown to support strong localized surface plasmon-polariton (SPP) modes [6]. When choosing an operating frequency corresponding to one of these resonant modes, the electromagnetic field inside the structure is significantly enhanced, which leads to a dramatic change in the linear and non-linear optical response of the material, including strong non-linear effects at a relatively small optical power.

2. Mathematical Formalism.

To solve the SHG problem, the total FF field must be determined first as it will serve as the source of the nonlinear polarization at SH; in turn, this polarization generates the field at the SH. The geometry of the problem consists of N parallel, infinitely long cylinders denoted by C_j and oriented along the z axis. The cylinders are characterized by their polar coordinates (r_j, φ_j), dielectric function $\epsilon_j(\omega)$, magnetic permeability μ_j and nonlinear surface and bulk constants described by the susceptibilities $\chi_{\varphi j}, \chi_{rj}$,.and the parameters α, β and γ, respectively (see [7]). The cylinders are embedded in a background medium of dielectric constant ϵ_b and magnetic permeability μ_b. For the sake of simplicity, it is assumed that $\mu_j = \mu_b = \mu_0$, though the formalism can easily accommodate the general case.

Assuming an incident wave of wave vector $\vec{k}$ characterized by its two conicity angles (ϕ_0 and θ_0) as well as its polarization angle δ_0, the longitudinal component of the FF, U_z (either E or H depending on the polarization of the incident wave) can be approximated by a Fourier-Bessel series at a given point $P(r_p, \phi_p)$, as the sum of the incoming field and the fields scattered by the cylinders [9]. Thus:

$$U_z^{total}(P) = \sum_m a_{mj} J_m\left(k\, r_j^p\right) e^{im\phi_j^p} + \sum_j \sum_m b_{mj} H_m^{(2)}\left(k\, r_j^p\right) e^{im\phi_j^p} \tag{1}$$

where r_j^p denotes the relative coordinate of the point P to the centre of cylinder C_j and J_m and H_m are Bessel functions and Hankel functions of the second kind, respectively. Using the same approach, the field surrounding a cylinder C_j can be expressed in a series as well:

$$U_{zj}^{total} = \sum_m c_{mj} J_m\left(k\, r_j^p\right) e^{im\phi_j^p} + \sum_m b_{mj} H_m^{(2)}\left(k\, r_j^p\right) e^{im\phi_j^p} \tag{2}$$

where the first term indicates the total "incoming" field on a cylinder (*i.e.*the sum of the incident field and the fields scattered by all the other cylinders, except C_j) and the second term is the decomposition of the field scattered solely by C_j.

By constructing the column vectors $\boldsymbol{C}_j = \{c_{mj}\}$ and $\boldsymbol{B}_j = \{b_{mj}\}$ the two terms can be linked in a matrix equation of the form:

$$\boldsymbol{B}_j = \boldsymbol{S}_j \boldsymbol{C}_j \tag{3}$$

in which $\boldsymbol{S}_j$ denotes the scattering matrix of cylinder C_j. It is worth noting that $\boldsymbol{S}_j$ is dependent only on the electromagnetic properties of C_j and is not influenced by the actual geometry or position of C_j.

Expanding this same idea to the entire system of N cylinders and denoting $\boldsymbol{B} = \{\boldsymbol{B}_j\}$ and $\boldsymbol{A} = \{\boldsymbol{A}_j\}$, column vectors, a system of equations linking the incident and scattered fields can be written for the structure as a whole:

$$\boldsymbol{B} = \boldsymbol{S}(\omega)\boldsymbol{A} \tag{4}$$

where $\boldsymbol{S}(\omega)$ is the global scattering matrix at the FF.

The vector $\boldsymbol{A}$ being known beforehand for a given incoming wave, the problem is reduced to solving (4) and determining $\boldsymbol{B}$. Then, the FF field at any point can be retrieved by using (1).

The components of $\boldsymbol{S}_j$ are directly derived from the continuity of the tangential components of the electromagnetic field at the boundary of C_j. $\boldsymbol{S}(\omega)$ for all N cylinders is then built from the matrices $\boldsymbol{S}_j$.

In the case of the SH, the field is generated by the nonlinear polarization, $\vec{P}_{nl}^{(2\omega)}$, which has a surface contribution,

$$\vec{P}_{surf}^{(2\omega)} = \epsilon_0 \chi_s : \vec{E}^{(\omega)} \vec{E}^{(\omega)} \delta(\vec{r} - \vec{r_s}) \tag{5}$$

as well as the bulk contribution:

$$\vec{P}_{bulk}^{(2\omega)} = \alpha[\vec{E}(\omega) \cdot \nabla]\vec{E}(\omega) + \beta\vec{E}(\omega)[\nabla \cdot \vec{E}(\omega)] + \gamma\nabla[\vec{E}(\omega) \cdot \vec{E}(\omega)] \tag{6}$$

Equation (4) must now take into account the non-linear boundary conditions for the cylinders, which must include the non-linear polarization [6].

This method has been successfully applied before in a photonic crystal made of a non-centrosymmetric material [8] and using a non-linear background with bulk polarization only. The formalism presented here is more general, as it allows for the inclusion of nonlinearity of the material from which the cylinders are made as well as the description of the non-linear surface polarization.

3. Results and Discussions.

This section presents the results of the algorithm applied to several configurations that present particular interest for testing the validity of the formalism, illustrating its versatility, as well as for the investigation of important practical applications. One such example is a two cylinder geometry, where the coupling of the fields scattered by the two cylinders, both at the FF and SH, is clearly illustrated by the plots in Fig. 1.

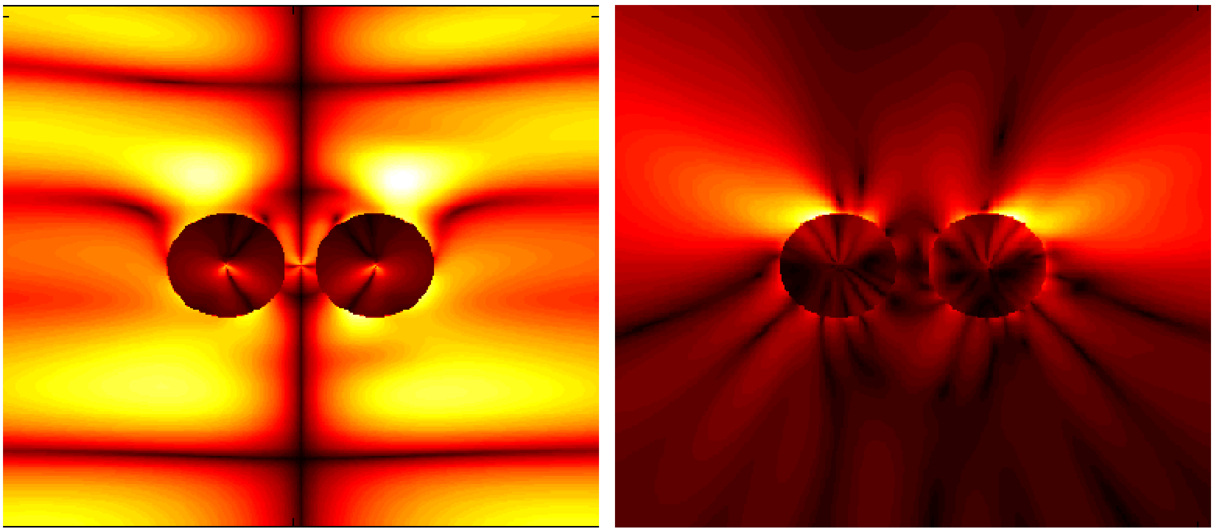

Fig. 1 Electric field profiles at the FF (left) and SH (right) frequencies for two cylinders of radius $R = 200nm$ and an incoming wave with $\lambda_0 = 567\ nm$

Fig. 2 shows the results of simulations for a larger structure with a rectangular geometry. It is worth noting that the formalism can accommodate for any type of distribution (both structured and random) of cylinders of variable radii and properties. It is also worthy to note the distinct difference between the FF field which is formed of incoming waves disturbed by the presence of the metallic cylinders and the SH field which shows both surface excitations not present in the former, as well as a profile consistent with a radiated field.

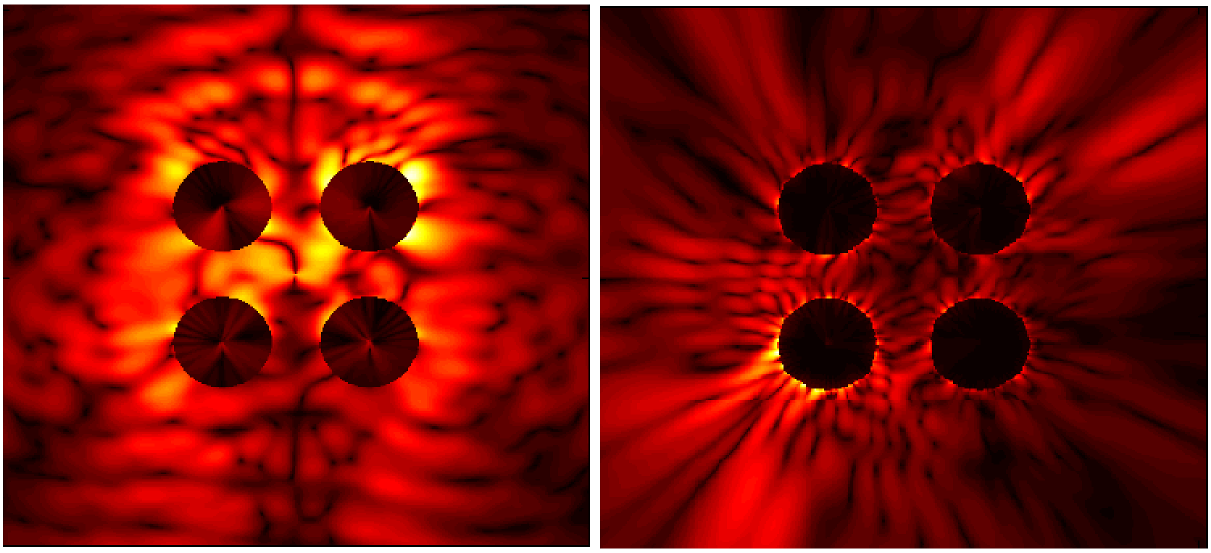

Fig. 2 Electric field profiles at the FF (left) and SH (right) for a rectangular geometry of cylinders of radius $R = 500\, nm$ for an incoming wave with $\lambda_0 = 567\, nm$

4. Conclusions.

In conclusion, we have presented a numerical method that can be used to determine the field distribution at the FF and SH, in nanostructures consisting of an arbitrary distribution of metallic nanowires. The modified MSM formalism presented here has demonstrated the excitation of SSPs at certain operating frequencies, which suggests that a strong SH field can be generated in the metamaterial, at a low optical input power. The algorithm has also proven to be adequate for a wide range of possible configuration both in structure size, distribution or electromagnetic properties.

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