Non-linear Plasmonic Cavities made of Metallic Nanowires: Theoretical Analysis and Applications

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Abstract: This paper presents a numerical analysis of the physical mechanism involved in the formation of non-linear plasmonic cavitiy modes in cavities made of metallic nanowires. We employ a method by which both the surface and bulk contributions of the non-linear polarization to the second harmonic generation are taken into account. Our results show how the system geometry influences the properties of the plasmonic cavity modes, thus allowing for the tailoring of their optical response to specific applications. Such potential applications of these nano-structures to sub-wavelength nano-lasers and nano-sensors are also discussed.

1. Introduction

The field of metamaterials has in recent years provided a revolutionary approach to the study of the electromagnetic properties of materials. New advances in nanotechnology have allowed one to employ artificial building blocks in a bottom-up approach for designing and fabrication of new types of optical media with remarkable properties. This new approach has proved extremely successful since these primary building blocks, with size smaller than the operating wavelength, act as “photonic atoms” whose optical response can be tailored so as to provide a broad range of functionalities. Recent findings, for example, have lead to the demonstration of artificial media with magnetic response at THz frequencies [1] or negative-index of refraction [2]. In this article we will present our findings pertaining to the optical properties of non-linear plasmonic cavities made of metallic nanowires. We employ an advanced numerical method based on multiple scattering method (MSM) to describe the spatial distribution of both the fundamental and non-linear fields in these structures, both in the frequency and the time domain. We will also show how these devices can be employed to the design of efficient nano-lasers and plasmonic sensors.

The paper is organized as follows. Sec. 2 gives a brief description of the algorithm employed. In Sec. 3 we present typical results pertaining to the formation of cavity modes in the structures we investigate. Sec. 4 and Sec. 5 contain a discussion of the temporal behaviour of non-linear cavity modes and the influence of the background refractive index on the formation of plasmonic cavity modes. The main conclusions are presented in Sec. 7.

2. Description of the Numerical Approach

Our numerical approach is based on the multiple MSM, which has been successfully employed for the study of second-harmonic generation (SHG) from single [3] and multiple [4] cylindrical metallic scatterers. In what follows, we briefly outline the steps involved in solving the non-linear scattering problem. In the first instance, an incoming plane wave at the fundamental frequency (FF) is scattered by the nanowire structure. In mathematical terms, the Fourier-Bessel coefficients of the incoming wave are linked *via* a system-dependent scattering matrix, which uniquely describes the interaction of an electromagnetic wave with the scatterers, to the coefficients of the scattered wave. By solving the resulting system of linear equations, the field distribution at the FF can be fully determined.

The next step consists of calculating the sources of the field at the second harmonic (SH). Because we are working with metallic, and thus centrosymmetric, scatterers embedded in a dielectric, the non-linear polarization has two components. A dipole-allowed, localized, surface contribution given by:

$$\vec{P}^{(2\omega)}_{surf} = \epsilon_0 \chi_s \vdots \vec{E}^{(\omega)} \vec{E}^{(\omega)} \delta(\vec{r} - \vec{r_s}) , \qquad (1)$$

respectively, a nonlocal bulk contribution which, for an isotropic medium can be written as:

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

Once the non-linear source field is known, a multiple scattering approach similar to the one employed for the field at the FF can be used to determine the full field distribution at the SH.

In order to determine the temporal evolution of the field in a given structure we employ a three step algorithm. First, an incoming pulse with a Gaussian envelope is Fourier transformed into a series of monochromatic plane waves. The scattering problem is then solved for each of these components using the MSM formalism described above. Finally, the full optical field in the time domain can be retrieved by performing an inverse Fourier transform [5].

3. Non-Linear Plasmonic Cavity Modes

We begin the discussion of our results by describing the geometries we have employed in our simulations. As we have mentioned, we considered cavities formed from infinitely long, cylindrical, metallic nanowires embedded in a dielectric medium. In particular, we use cylinders made of Ag embedded in vacuum. As the excitation of resonant plasmonic modes leads to a strong field confinement inside the cavity, such a mode has a distinct signature (resonance peak) in the absorption spectra at the SH. Figure 1 summarizes our main results pertaining to the spectral properties of plasmonic cavity modes.

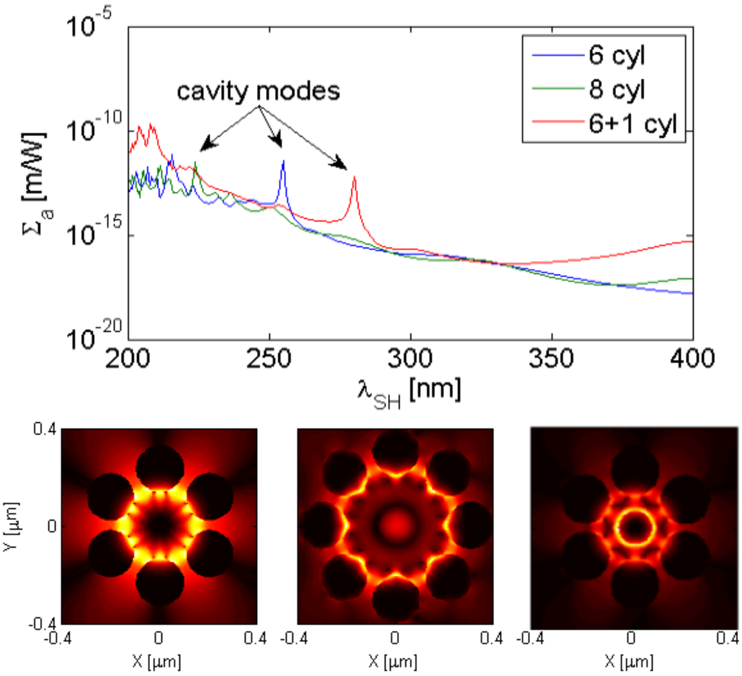

Figure 1: Top panel shows the absorption cross section for a six- and eight-cylinder cavity with $d = 30\ nm$ and a six-cylinder cavity containing a cylindrical inclusion with $r = 70\ nm$. Bottom panels show the corresponding field profiles at the SH. The geometries consist of Ag cylinders with $R = 100\ nm$.

The expected absorption peaks are clearly visible in the top panel of Fig. 1. These resonances correspond to the three main types of cavities we have investigated, namely, a six-cylinder and eight-cylinder "empty" cavity and a six-cylinder cavity that contains a cylindrical inclusion. The field profiles at the SH confirm the fact that these are non-linear cavity modes. The main conclusion we can draw is that the size and shape of the cavity has a very strong influence on both the spectral position and the shape of the resonant mode. The following sections will show how this idea can be used to fine tune the structures for various applications.

4. Temporal Dynamics of Cavity Modes

The main physical quantity describing the time domain behaviour of a plasmonic cavity mode is the mode's Q factor. The Q factor is defined as the product between the lifetime of the cavity mode and the resonance frequency of the mode. Consequently, the Q factor quantifies the decay lifetime of a plasmon resonance. As such, it is the main factor that must be taken into account when choosing an optical cavity for lasing applications [6]. Figure 2 summarizes our results regarding the temporal dynamics of non-linear plasmonic cavities modes.

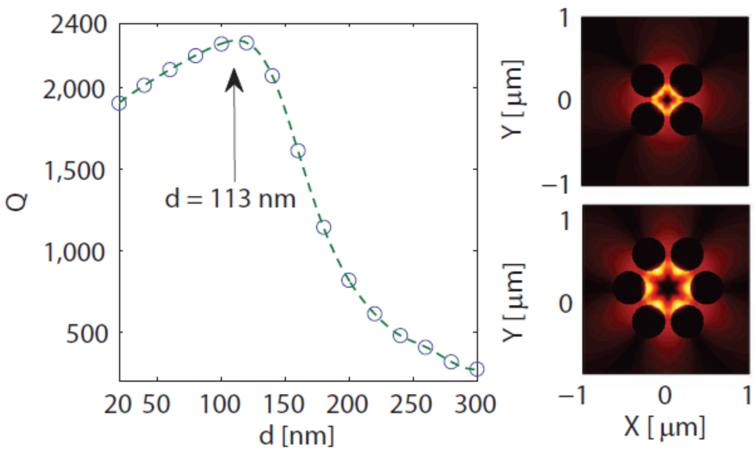

Figure 2: Left panel shows the dependence of the Q factor of a cavity as a function of the separation distance d. Left panels show electric field profiles for a four- and six-cylinder cavity with $d = 70\ nm$. Geometries are Ag cylinders with $R = 200\ nm$ whereas the incoming pulse has duration $T_0 = 238\ fs$.

Our results show that the Q factor increases with the separation distance up to a certain point, beyond which it decreases. This somewhat surprising finding requires further discussion. Thus, one would expect the Q factor to decrease monotonously with d since in this case the radiative losses increase. To explain this apparent inconsistency we write the Q factor of the cavity in the following form:

$$^{1}/_{Q} = {}^{1}/_{Q_{abs}} + {}^{1}/_{Q_{rad}}, \quad (3)$$

where, Q_{abs} and Q_{rad} are given by the losses due to absorption in the metal and the radiative losses, respectively. We can see now that, as d increases, the field confinement decreases and the absorption decreases leading to an increase in the Q factor. We can also conclude that the radiative component bellow separation distance of $d = 120\ nm$ is dominant, the radiative losses being almost zero, a characteristic of dark-plasmon modes. As expected, with further increases to the separation distance, the radiative losses become dominant and the cavity no longer confines the field. This result shows that, owing to the intricate interplay between the various geometrical properties of a cavity and that cavity's time domain behaviour, we can tailor these structures to achieve the best possible design for use in applications where high Q factors are crucial, such as sub-wavelength nano-lasers.

5. Plasmonic Cavities as Sensors

Plasmonic structures can also be employed as nanoscale sensors [7], for example, for chemical and biological applications. By employing non-linear plasmonic cavities, we can obtain a low-power device which can be tailored for specific spectral domains. The main physical property used to determine the efficiency of such a device is the ratio between the change in the refractive index of the sample and the location of the resonance peak corresponding to that index of refraction. In effect, this ratio can be considered the figure of merit of the device. We have used a six-cylinder cavity with a cylindrical inclusion to determine the shift in the wavelength of the plasmonic cavity mode as a function of the index of refraction of the background dielectric. Figure 2 summarizes our results.

Our data shows that the slope of the resonance line associated with the cavity mode is strongly influenced by the size of the cylindrical inclusion. In fact, the figure of merit of the cavity varies from $d\lambda/dn = 267.25\ nm$ to $d\lambda/dn = 306.53\ nm$ as the radius of the inclusion is increased from $r = 50\ nm$ to $r = 90\ nm$. This result shows that by changing only one variable in our design, the properties of the device as can be modified. In effect, such devices could be easily tailored to suite various applications with no need to change the base design of the structure.

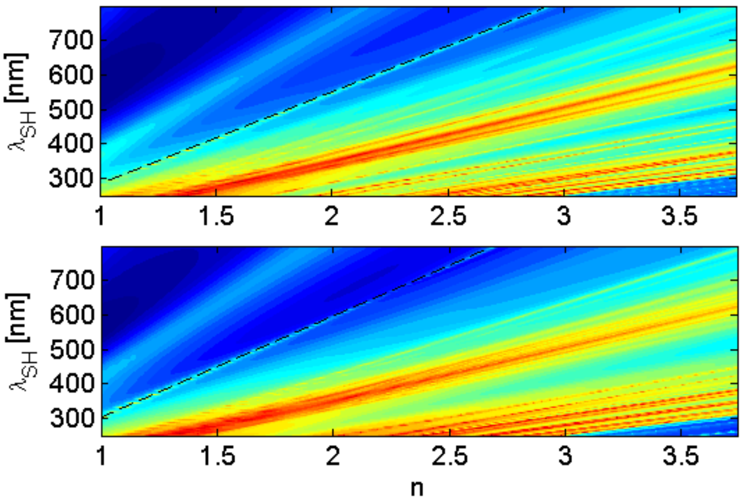

Figure 3: Absorption cross section as a function of wavelength and background refractive index. Geometries are Ag cylinders with $R = 100\ nm$ and $d = 50\ nm$ (top) and $d = 90\ nm$ (bottom). Dashed line highlights the location of the main cavity mode.

7. Conclusions

In conclusion, we have used a numerical method based on multiple scattering theory to investigate the formation of non-linear cavity modes in cavities made of metallic nanowires. We have demonstrated that the shape and size of a cavity has a strong effect on the properties of these modes. In particular, we have shown how, by tailoring the main geometrical parameters of such cavities, we can obtain cavities with high Q factor or cavities supporting plasmonic modes whose resonance wavelength is strongly dependent on the geometry of the cavity. Finally, we have discussed how these structures can be employed in the fabrication of nano-devices such as sub-wavelength lasers and nano-sensors.

Acknowledgment

The authors acknowledge the use of the UCL Legion High Performance Computing Facility, and associated support services, in the completion of this work. This work was supported by the UK Engineering and Physical Sciences Research Council (ESPRC), under Grant No. EP/G030502/1.

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