

Designing, Simulating, and Optimising Nanoantennas that utilise Nonlinear Effects

A literature review

Project: EXSS-Sapienza-1

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Abstract

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1 Introduction

The goal of our project is to investigate, both experimentally and computationally, the applicability and efficiency of different nanoantenna designs for enhancing nonlinear optical effects at the nanoscale. Light manipulation using the nonlinear response of materials has existing and promising future applications in a number of areas, including frequency control for laser light, optical communications, spectroscopy, data storage, and sensing^[1]. Nonlinear effects often require large interaction volumes and high laser powers to occur, and nanoantennas are one of the proposed ways to alleviate this and bring nonlinear optics to small-scale applications.

2 Nonlinear optics

When considering the propagation of light in matter, it is often assumed that the response of the material is linear^[2], giving a simple expression for the polarisation:

$$\mathbf{P} = \epsilon_0 \chi \mathbf{E}, \quad (1)$$

with ϵ_0 being the permittivity of free space, and \mathbf{E} the electric field. As outlined in [3], this expression can be generalised to include terms that only gain significance at higher field magnitudes, and can be thought to correspond to potentials for the electrons in the material deviating from the simple harmonic approximation:

$$\mathbf{P} = \epsilon_0 (\chi^{(1)} \mathbf{E} + \chi^{(2)} \mathbf{E}^2 + \chi^{(3)} \mathbf{E}^3 + \dots) \quad (2)$$

$$= \mathbf{P}^{(1)} + \mathbf{P}^{(2)} + \mathbf{P}^{(3)} + \dots, \quad (3)$$

where $\chi^{(n)}$ is known as the n^{th} order nonlinear susceptibility, and is in general a tensor.

Now if we allow \mathbf{E} to vary in time we will also get time variation in \mathbf{P} , and that movement of charges drives a further electric field oscillation. One can then see how the nonlinear terms in (2) lead to additional frequency components in this new oscillation. For a simple example adapted from [3], we can take $\mathbf{E}(t) = \mathbf{E}_0 \cos(\omega t)$ and look at the second order polarisation:

$$\mathbf{P}^{(2)} = \frac{\epsilon_0 \chi^{(2)}}{2} \mathbf{E}_0^2 (1 + \cos(2\omega t)). \quad (4)$$

This then drives an electromagnetic wave (the *signal*) at angular frequency 2ω , compared to ω for the incoming (*pump*) wave. The process is known as second harmonic generation (SHG).

Other basic nonlinear processes of note include third harmonic generation (THG), sum and difference frequency generation ($\omega' = \omega_1 \pm \omega_2$), changes to the refractive index (a third order process), and others^[3].

The *phase matching* parameter, $\Delta k = k' - k$ is crucial in designing most nonlinear processes that involve a change in frequency^[3]. As both the pump (wavevector k) and signal (k') fields interact with the same electric dipoles, the best conditions for coupling between them is when they are in phase such that they don't interfere destructively. While achieving $\Delta k = 0$ is possible through anomalous dispersion, it is more feasible to do this by utilising birefringence^[4]. This is only possible for some materials, but if done correctly it can lead to quadratic dependence of the signal density on the interaction distance. On the other hand, if $\Delta k \neq 0$, choosing the wrong sample length can lead to a total loss of signal.

3 Material considerations

There is a number of factors to consider when choosing a material for nonlinear interactions. The nonlinear susceptibility is different for different materials^[5], and can also vary spectrally^[6]. Second order effects also demand an asymmetry of potential, which cannot be present in centrosymmetric materials, and as such no bulk second order effects will be present in them, only surface ones^[3].

While metals can be used as a medium for nonlinear effects, the currents appearing in them encounter resistance, which leads to losses. This is one of the reasons for a shift towards utilising dielectric materials, where we have displacement currents instead^[7,8], and can choose samples that have a large refractive index (needed for field confinement) and low absorption, preferably both at the pump and signal frequencies.

A useful property that some materials, like Indium Tin Oxide (ITO), exhibit is epsilon-near-zero (ENZ). In these materials the real part of the relative permittivity goes to zero for electromagnetic waves at a given wavelength (λ_{ENZ}). This has interesting effects on light propagation, but importantly for us it can lead to an enhancement of $\chi^{(3)}$, and greatly increased field confinement^[9]. This can give significant enhancement of nonlinear effects.

4 Nanoantennas

For some applications, especially in the field of optical devices, we may want to couple far-field radiation into the near-field by concentrating it in subwavelength nanostructures. This can be achieved using nanoantennas, which are small formations of metal or dielectric usually designed to have resonances that capture and enhance the field inside^[7].

Simple antenna shapes (for example cylinders) can be considered in terms of basic resonances similar to the ones described by the Lorenz-Mie theory for spheres^[10]. Fig. 1 shows a set of these resonances classified as electric, magnetic, and toroidal. Note that metallic antennas only produce significant resonances of the electric type, and mostly through surface effects^[11] – this is due to field suppression inside a metal. The ability to support both electric

and magnetic modes is thus another advantage of dielectrics.

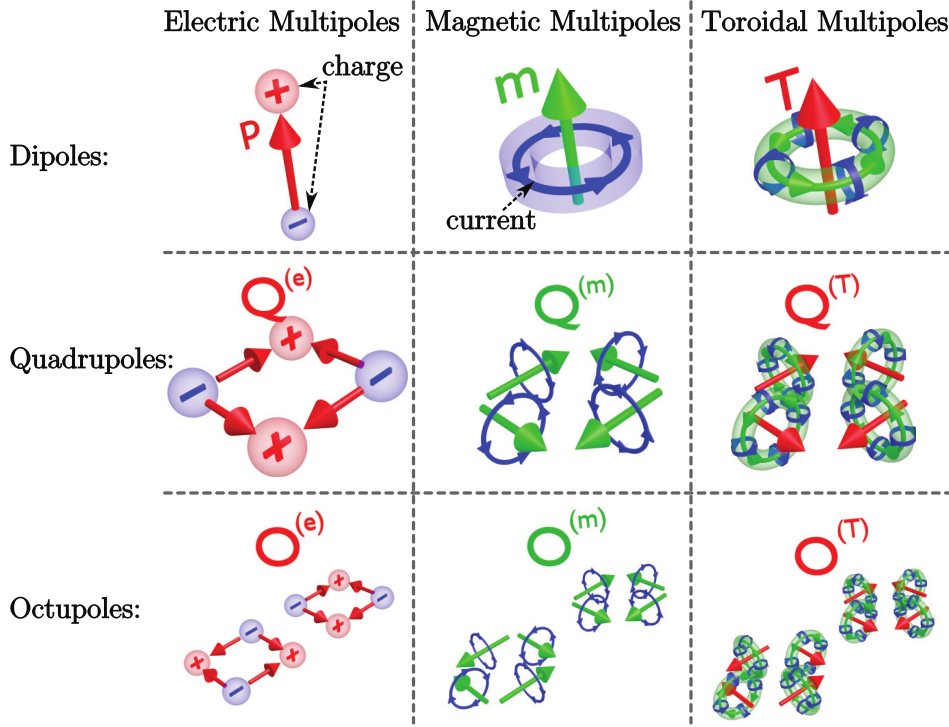


Figure 1: The three lowest orders of electric, magnetic, and toroidal modes. Superpositions of these can be excited in nanoantennas. Adapted from [12].

In practice these resonances can spectrally overlap and combine to form modes observable in the antenna’s scattering spectrum – their far-field response. A notable example of this is the anapole mode, for which the toroidal and electric dipoles destructively interfere in the far-field, creating an *dark* mode with low scattering^[13]. Mode superpositions can also be utilised to direct the antennas output through far-field interference.

While this discussion focused on simple antenna shapes, more complex arrangements can be used to achieve various output goals. For example, [7] demonstrates how a set of $\sim 1\ \mu\text{m}$ diameter silicon spheres laid out in analogy to the Yagi-Uda radio-frequency antenna design can result in output beam-widths as low as 40° .

5 Enhancing nonlinear effects with nanoantennas

An important advantage of nanoantennas as a nonlinear medium is that their subwavelength scale relaxes the phase matching condition^[10], making the design easier and allowing for simplified use of non-birefringent materials. The decreased size results in a disadvantage for nonlinear interactions – the smaller interaction volume. Beyond the intuitive diminishing of signal when we reduce the amount of material emitting it, there is also the significant dependence of the signal intensity on the interaction length, which can be quadratic in some cases (as seen in Section 2). To make up for this we rely on the field enhancing properties of nanoantennas.

Good field enhancement can be achieved by selecting the correct mode of the antenna. For example, [13] reports that the aforementioned anapole mode in a germanium nanodisk

results in a much better energy concentration in the antenna; spectral analysis shows that the less scattering is observed, the more energy is confined in the nanoantenna near-field, highlighting the significance of dark modes. The generated third harmonic intensity when the anapole is excited is predicted and measured to be an order of magnitude greater than for the spectrally close electric dipole resonances, and four orders of magnitude better than unstructured germanium film. The absolute THG efficiency is given as 0.0001%.

When choosing a mode to excite in a nanoantenna, it is important to consider where we want to be concentrating the field. The above experiment was concerned with THG, and so field enhancement inside of the antenna was desirable. [14] demonstrates a gallium phosphide (GaP) nanodisk for efficient SHG; as GaP is centrosymmetric the field needs to be enhanced at the disk's surface, where the symmetry is broken and second order nonlinear effects can occur. A resonant mode has been found for which the electric field magnitude is up to 30 times greater than the incident \mathbf{E}_0 (in contrast to the anapole mode, this one corresponds to a peak in scattering). Together with the fact that a GaP substrate with GaP cylinders on it has a larger surface area than the substrate itself, this gives a SHG enhancement by 3 orders of magnitude compared to just the substrate, for an absolute efficiency of 0.0002%.

The examples above demonstrate that it is important to excite a good mode to fit the desired outcome and get good enhancement of the nonlinear effect. Factors other than the antenna shape and the output amplitude can also be considered; for example [15] demonstrates how adjusting the thickness of an SiO_2 spacer between an AlGaAs nanodisk and a reflective ITO layer, as well as the pump beam shape and polarisation, can help excite a mode referred to as a quasi-BIC (bound state in continuum), leading to a 2-order-of-magnitude increase compared to other implementations of a similar antenna. The choices made in design are also reflected in the output field's shape, as illustrated in Fig. 2.

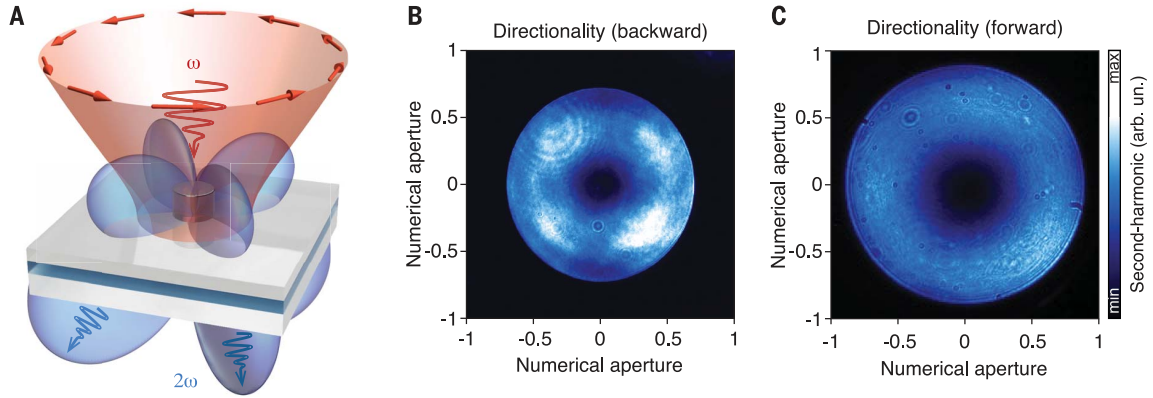


Figure 2: **(A)** The pump (red) excites a mode in the nanodisk, which in turn produces second harmonic radiation, shown in blue. The generated field also interacts with the antenna, producing the mode pattern seen on the picture. **(B)** and **(C)** show the far-field intensity of the generated radiation in different directions – the four lobes from (A) are well pronounced in the backwards-directed generation.

6 Predicting and optimising the nonlinear signal of an antenna

Experimental investigation of nonlinear effects is important to capture the full spectrum of their behaviours, as aspects like the material’s damage threshold and the way this limits laser pulse duration^[15]. Nevertheless, a thorough computational investigation is an important step in planning and preparing for experimental work, as it allows for exploring arbitrary antenna parameters without manufacturing samples, and for directly looking at field distributions inside the antenna.

6.1 Predicting the field inside of the antenna

6.2 Predicting the third harmonic signal

6.3 Methods of optimising the antenna signal

Small derivation from Lorenz reciprocity. Explain why this method is good. Finite-difference time domain stuff. Mention Lumerical. Mention the general tensor form of the nonlinear susceptibility.

Talk about the steps from [15]. Experimental methodology, computing. Combining materials. This section seems a bit useless...

7 Conclusion

You wish you were concluding^[16].

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