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► **To cite this version:**

Ruben Esteban, Marine Laroche, Jean-Jacques Greffet. Optical antennas for enhanced efficiency. 23rd European Photovoltaic Solar Energy Conference & Exhibition, Jun 2008, Stanford, United States. hal-00576086

HAL Id: hal-00576086

<https://hal-iogs.archives-ouvertes.fr/hal-00576086>

Submitted on 24 Aug 2022

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Optical Antennas for Enhanced Efficiency

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Abstract : High efficiency solutions for photovoltaics requires taking advantage of IR photons energy. Possible solutions are based on up-conversion processes which have a low cross section. We will discuss different solutions to enhance the absorption.

OCIS codes: (350.6050) (350.4238)(240.6680) (250.5403)

1. Introduction

Advanced solutions for enhanced efficiency include the use of up-conversion processes to recover part of the IR energy of the solar spectrum [1,2]. As the up-conversion process has a low cross section, it is necessary to find solutions to increase the absorption. There are other ideas that aim at using the energy of the UV photons. These ideas are based on the generation of an out of equilibrium hot-electrons gas and the subsequent fast extraction of energy [3]. Such a system is possible if a strong absorption by a very thin system is possible. It is clear that these ideas require the development of optical resonators that allow to produce highly absorbing devices based on low absorbing materials. Part of the challenge for enhanced efficiency photovoltaics is thus to design good absorbers using low absorbing materials and/or thin systems.

It is simple to analyse the different possibilities to address this issue. Absorption by a device is proportional to the integral over the device volume of $\text{Im}(\epsilon)|E|^2$, namely the imaginary part of the dielectric constant and the square modulus of the electric field in the absorbing device. The standard approach to solve this question is to increase the thickness of the solar cell in order to increase the absorption volume. Yet, this is not desirable when dealing with electron-hole pairs that can recombine. An alternative route is to modify the material in order to increase its intrinsic absorption given by $\text{Im}(\epsilon)$. In the case of silicon, the intrinsic absorption is low for energies close to the gap because there is an indirect gap. A lot of work in the field of material science has been devoted to address this issue [4]. Finally, one can use optical resonators to enhance the local fields E . It follows that complete absorption can be obtained with a given material and a small volume provided that the enhancement of the local field is large enough. In what follows, we use the name « optical antennas » for devices that play this role. Simple microcavities can be used in principle. Many applications in the field of resonant detectors have been reported along these lines [5]. Other structures can be envisioned. In particular, it is known that Raman processes can be dramatically enhanced using metallic nanoparticles. More recently, there have been reports on fluorescence enhancement by metallic nanoparticles [6-9]. The purpose of this work is to analyse the potential of metallic nanoparticles for enhancing up-conversion and its potential application to solar energy.

In particular, we consider here two up-conversion mechanisms which are often predominant in systems consisting of rare earths such as Erbium or Erbium-Ytterbium[4]: excited state absorption (ESA) and non-radiative energy transfer (NRET). ESA is a mechanism where a given molecule absorbs two photons at ω and reemits a photon at 2ω . NRET is a mechanism where two rare earths (Erbium and Ytterbium) are first excited to an intermediate level by photons at ω . A non-radiative energy transfer between them is then followed by a radiative desexcitation at 2ω . The up-converted signal far from saturation is proportional to the square (or a higher power) of the light intensity, which reminds to the typical fourth power relationship of the Raman signal with the local electric field; one could thus hope to find similar advantages of localized plasmonic resonances for up-conversion than already proved for SERS. Both NRET and ESA are, however, incoherent

absorption-emission processes, and in this sense resemble fluorescence more closely than Raman, so that large enhancements are far from guaranteed.

In this paper, we derive simple analytical equations which serve to study the influence of the environment on up-conversion processes. They allow to discuss the similarities but also the significant differences with both the Raman and fluorescence case. The desire is not to reproduce all the multiple phenomena on a real system, but to exemplify in general terms how the modification on decay rates and local fields derived from the Maxwell equations influence the emission. Accordingly, relatively simple energy level schemes enter our models, and we ignore other possible effects such as a modification in the crystal field due to microscopic changes in the atomic structure. For numerical illustration, we consider metallic particles of different diameters that exhibit localized Surface Plasmon Polariton (SPP) resonances.

2. Modelling and numerical solution

In this section, we will analyse the influence of the metallic nanoparticle on the Raman, fluorescence and up-conversion processes. We will emphasize differences and derive simple formulas that give the ratio of the emitted power by a molecule with and without metallic nanoparticles. It is particularly important to stress the difference between the Raman scattering process and the up-conversion process. Whereas enhancement of several orders of magnitude can be obtained for the Raman cross section, both fluorescence and up-conversion processes can hardly be enhanced by more than an order of magnitude using metallic nanoparticles as we show below.

The interaction between the molecule and the nanoparticle is modelled by solving numerically Maxwell equations using the multiple multipole technique [9]. Our numerical simulations will focus on the ESA mechanism as the NRET mechanism is not found to be improved by the presence of the metallic nanoparticles. We shall explain the underlying physical mechanism. We find that the ESA process can be enhanced by almost an order of magnitude with silver nanoparticles for some distances as seen in Figure 1. We consider the up-conversion process for a model atom with absorption levels separated by hc/λ . Depending on the relative position of the atom absorption frequency and the resonance of the metallic particle, the effects change significantly. It is seen that for distances lower than 5 nm, the up-conversion is quenched. The energy is not radiated but transferred to the particle and converted into heat. The maximum up-conversion enhancement is obtained for relatively large (~100 nm radius) spheres, which present a local field enhancement and radiative decay rate dipolar maxima considerably red-shifted with relationship to the non-radiative decay rate maximum[6]. The above results are based on a model that uses a simplified description of the energy levels of the quantum emitter. We shall discuss how differences on the level scheme of the up-conversion quantum emitters influence the results, and allow larger signal increases than shown in Fig.1.

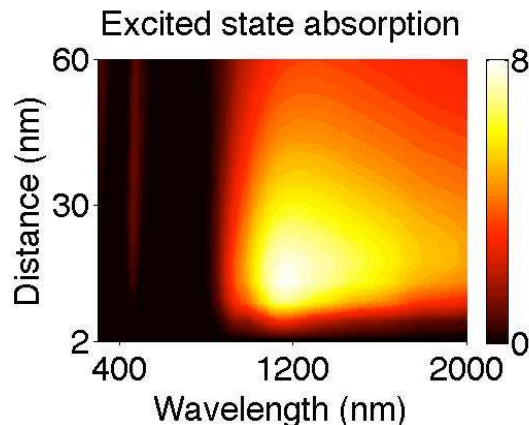


Figure 1 : Power emitted by an atom located at a distance d from silver nanoparticle of 100nm radius placed in a glass of dielectric constant 2.25, normalized by the power emitted by the same molecule without nanoparticle versus the absorption energy of the atom hc/λ . The maximum up-converted signal enhancement is near the frequency of the dipolar resonance.

This enhancement for the ESA process is interesting if the primary goal is to enhance up-conversion. For solar energy applications, the key question is the overall efficiency. As the up-conversion cross section is very small, an enhancement by a factor 8 is certainly not enough to make it comparable with the absorption cross section of the metallic nanoparticle. It follows that introducing metallic nanoparticles essentially increases the conversion of solar energy into heat.

3. Conclusions

In summary, we have developed a model that accounts for the up-conversion process in the presence of a metallic nanoparticle. We account for the local enhancement of the field, the enhanced emission rate and the non-radiative transfer to the particle. Our numerical simulations show that the ESA process can be enhanced by a factor of 8 when using silver nanoparticles with a radius of 100 nm. This is a significant enhancement but is considerably smaller than what is obtained for Raman processes. With smaller radii, the enhancement is significantly decreased. While we predict an up-conversion efficiency enhancement of almost an order of magnitude, this is far too modest to be useful for solar applications. Indeed, although the absorption cross section of the rare earth has been increased due to the presence of the nanoparticles, the absorption cross section of the nanoparticles is many orders of magnitude larger so that most of the solar energy goes into heating the metallic nanoparticles. Other solutions based on resonators made with dielectric materials will be explored in the near future. Preliminary results appear to be very promising.

4. References

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