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Local Surface Plasmon Resonance of metallic nanoparticles embedded in amorphous silicon

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Abstract

This work reports a theoretical study aimed to identify the plasmonic resonance condition for a system formed by metallic nanoparticles embedded in an a-Si:H matrix. The study is based on a Tauc-Lorentz model for the electrical permittivity of a-Si:H and a Drude model for the metallic nanoparticles and the polarizability of an aluminium sphere-shaped particle with radius of 10–20 nm. We also performed FDTD simulations of light propagation inside this structure reporting about the effects caused by a single nanosphere of aluminium, silver and, as a comparison, an ideally perfectly conductor. The simulation results show that it is possible to obtain a plasmonic resonance in the red part of the spectrum (600–650 nm) when 10–20 nm radius aluminium spheres are embedded into a-Si:H.

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1. Introduction

The optical path length of an optoelectronic device like a solar cell or a photodiode can be defined as the distance that an (unabsorbed) photon can successfully travel within the device before escaping out. This feature is usually described in terms of device thickness.

Light entering a device engineered with good light trapping features may get to be reflected back and forth many times, presenting an optical path many times higher than its physical thickness. This effect may enhance light absorption and, generally speaking, device performance. An attractive approach to improve the light trapping involves the introduction of plasmonic nanostructures coupled to absorbing

semiconductors [1]. Amorphous silicon (a-Si:H) has an optical gap of about 1.75 eV and it presents good absorption properties under visible light irradiation. This characteristic, together with an attractive low cost of production, has gathered the attention of scientific community during the last two decades of the last century. Today, arrived to its maturity, a-Si:H is one of the most used semiconductors in the photovoltaic industry for production of thin film solar cells. However, the poor transport properties and the high defect density of this material limit the thickness of the absorbing layers to about 500 nm. Also the degradation induced by the Staebler-Wronsky effect can in some measure be minimized by the use of a thin intrinsic layer [1]. Such thickness limitation forces the absorption of wavelengths longer than 550 nm to low values and suggested the use of a-Si:H in tandem junction configuration [1].

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In recent years, much attention has been recently given to the use of metal nanoparticles (NPs) embedded in semiconductor materials due to the possibility of enhancing light absorption in the semiconductor through the plasmonic near-field enhancement. Such an enhancement is expected at photon energies in the vicinity of the localized surface plasmon resonance condition (LSPR) of the metal nanoparticle. The LSPR condition depends on the metal/interface properties, on the particle size and shape, and finally on the average distance in the particle distribution [1,2]. Light trapping for improving solar cell efficiency based on plasmonic effects is reported in literature mainly for silver nanoparticles embedded in silicon solar cells [1,2] and in thin film amorphous silicon solar cells [1-3]. Reports on aluminium nanoparticles embedded in a-Si:H solar cells have also been recently published [1-3]. In this work we report about a theoretical study aimed to identify the plasmonic resonance condition for a system formed by metallic nanoparticles embedded in an a-Si:H matrix. The study is based on a Tauc-Lorentz model for the electrical permittivity of a-Si:H, a Drude model for the metallic nanoparticles and the polarizability of an aluminium sphere-shaped particle with radius of 10-20 nm. a-SiC:H has also been considered in our simulations, as it is used as a standard material for deposition of p-layers in a-Si:H solar cells. We also performed FDTD simulations of light propagation inside this structure reporting a comparison among the effects caused by a single nanosphere of aluminium, silver and, as a comparison, an ideally perfectly conductor. We considered a perfect conductor as an idealized material with infinite conductivity acting as a perfect reflector at optical frequencies. Such a comparison in the simulations permits the outlining and understanding of the plasmonic effects in opposition to standard optical reflections.

2. Theory

Optical properties of metal nanoparticles present unique characteristics, which, depending on size and shape, could get to be very different from the characteristics of bulk material. Coupling of light with the NPs surface produces a localized surface plasmon resonance resulting in an enhanced optical scattering and an enhanced optical field around the particle and thus enhancing the fraction of total power absorbed in the embedding semiconductor. The coupling strength of the NP and the embedding medium can be

measured by the nanoparticles polarizability which, in its turn, defines the nanoparticles scattering cross section. If we assume the size of the nanoparticles to be much smaller than the wavelength of the incident radiation, the field intensity can be considered constant all over the nanoparticle region. This approximation will lead to a simplified expression for the particle (complex) polarizability (α) that, for a simple spherical geometry, is quantified by the following expression:

$$\alpha = 4\pi r^3 \frac{\epsilon_m - \epsilon_s}{\epsilon_m + 2\epsilon_s} \quad (1)$$

where r is the radius of the nanosphere, ϵ_m and ϵ_s the permittivity of the metal and of the embedding semiconductor, respectively.

Once known the value of the polarizability, the nanoparticles scattering cross section for a determined wavelength λ can be calculated as:

$$\sigma_{\text{scat}} = \frac{1}{6\pi} \left(\frac{2\pi}{\lambda} \right)^4 |\alpha|^2 \quad (2)$$

Both the permittivities ϵ_m and ϵ_s are function of the light wavelength (λ). It can be easily deduced by Eqs. (1) and (2) that the satisfaction of the condition $\epsilon_m \approx -2\epsilon_s$ leads to a maximization of the polarizability and the scattering cross section. Under this condition, a surface plasmon resonance can be then expected.

The analysis we present is based on the Urbach-Tauc-Lorentz model [1] for the electrical permittivity ϵ of a-Si:H and its Lorentz reduction for application in Finite Difference Time Domain (FDTD) simulation [1]. The permittivities of all the metals considered in this study are taken from experimental results published in literature [1,2]. The FDTD simulations were executed with the 32-bit version of the OptiFDTD software [1].

3. Discussion

Aiming to identify the LSPR condition for a system of metal nanoparticles embedded in a matrix of a-Si:H, the polarizability of a single metal nanosphere has been analysed. In Fig. 1 it is reported the absolute value of the polarizability of a metal nanosphere, with a 10 nm radius, embedded in a-Si:H and a-SiC:H. In this case, the analysis is performed on copper, gold and silver, which are the mostly utilized metals in plasmonic applications. This plots are obtained combining, through Eq. (1), the experimental results for the metal permittivity and the UTL model for

a-Si:H permittivity. In both cases and for all the three metals, the peak of the polarizability is located in the near infrared region, around 1.2 eV. Because of the very low absorption of a-Si:H in the infrared range, a plasmonic resonance located at these wavelength would not be of any help in the process of light trapping and absorption optimization. The experimental results reported in literature [8-10] confirm this consideration, reporting about only a small enhancement of the solar cell response produced by the introduction of silver plasmonic nanostructures. The strongest enhancement is observed at longer wavelengths, from 700 to 800 nm, which is in the wavelength range when the photon energy approaches the absorption limit defined by the a-Si bandgap.

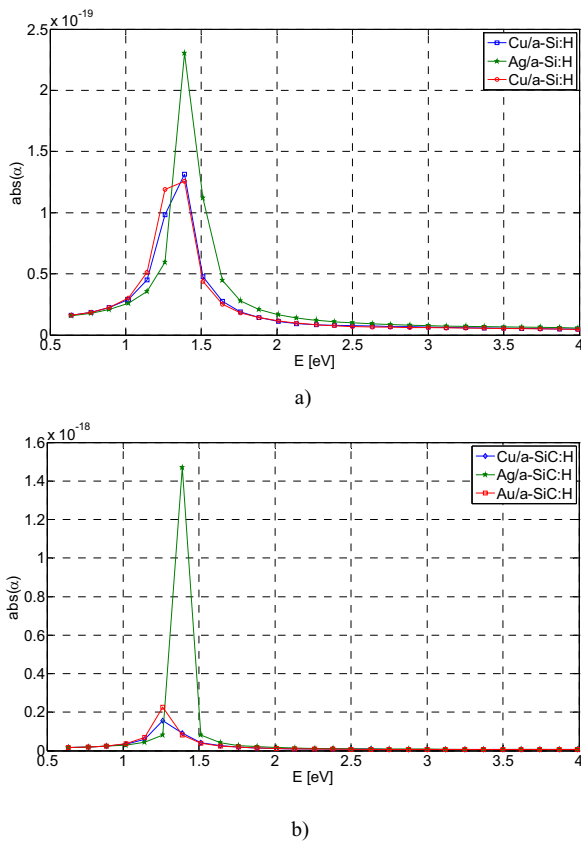


Fig. 1. Absolute value of the polarizability of a metal nanosphere, with a 10 nm radius, embedded in a-Si:H (a), and a-SiC:H (b), for three different metals: copper, gold and silver.

We should then look for a metal having the condition $\epsilon_m \approx -2\epsilon_s$ satisfied for wavelengths in the red part of the visible range. In Fig. 2 it is reported the analysis of this condition for an interface between a selection of metals and a-Si:H or a-SiC:H. It can be seen here that, for wavelengths of interest, the interface between a-Si:H and Cu, Au or Ag, the resonance condition is

not satisfied and no polarization peak should be expected. It should be anyway noted that a flattening of the NP would lead to a different expression for the polarizability and that a correct control of the NP shape could lead to a red shifting of the resonance condition.

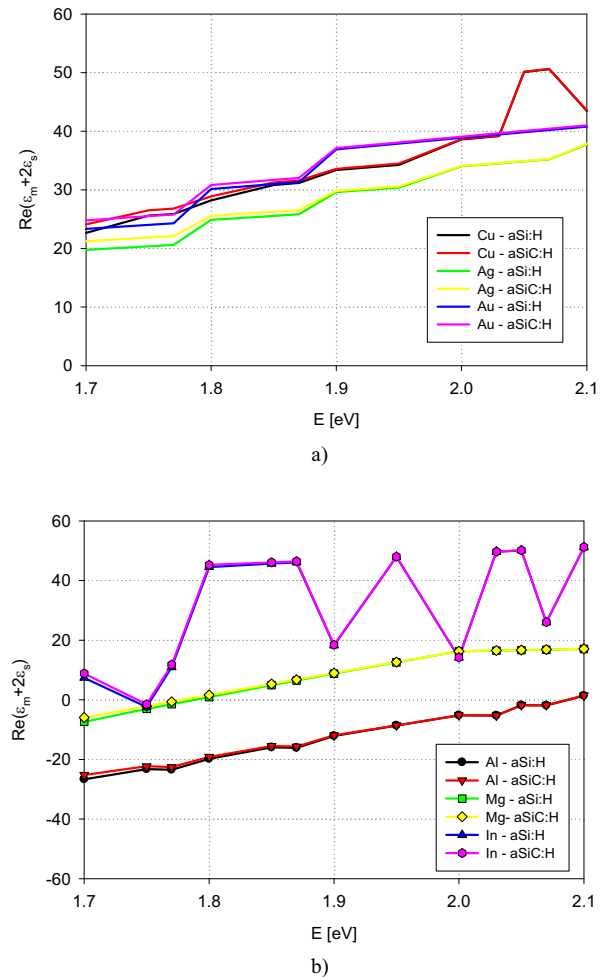


Fig. 2. Analysis of the condition $\epsilon_m \approx -2\epsilon_s$ for an interface between a selection of metals and a-Si:H and a-SiC:H: a) Cu, Ag and Au; b) Al, Mg and In.

Maintaining the spherical shape for the NPs, we have performed this analysis in a large set of metals, finding that the three metals that better satisfy the resonant condition in the red part of the visible range are aluminium, indium and magnesium. Fig. 3 reports the polarizability of a nanosphere made of one of these three metals, with a 10 nm radius, embedded in a-Si:H and a-SiC:H.

Our further analysis will be restricted to the LSPR of an aluminium nanosphere embedded in a a-Si:H matrix. In Fig. 4 it is reported the result of a FDTD

simulation of an Al nanosphere with 10 nm radius embedded in a-Si:H and illuminated with a light of 620 nm wavelength.

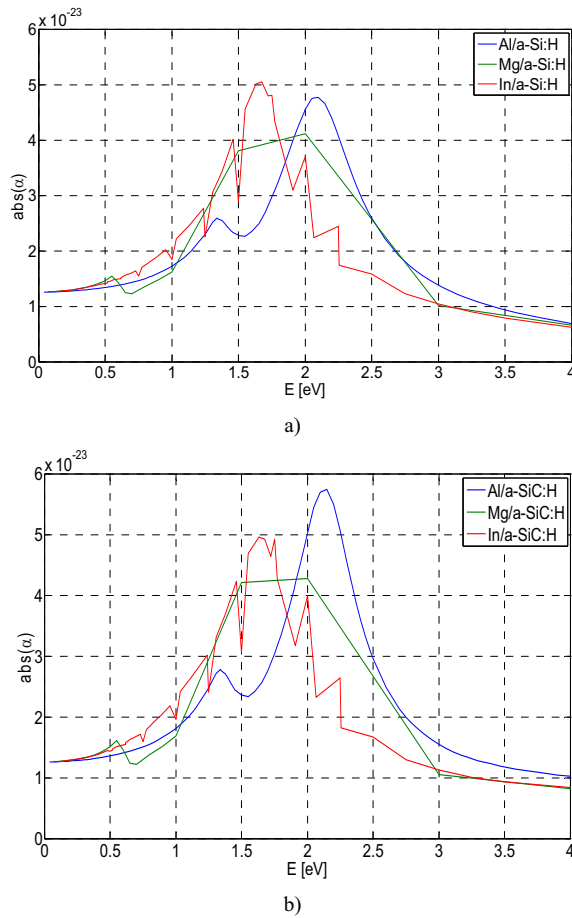


Fig. 3. Absolute value of the polarizability of a metal nanosphere, with a 10 nm radius, embedded in a-Si:H (a), and a-SiC:H (b), for three different metals: Al, Mg and In.

The analysis of the Poynting vector shows a scattering radius much larger than the physical dimension of the NP (see the large dark area in the NP surrounding). Comparing the Poynting vector obtained by simulating the light propagation in a situation with and without the presence of the aluminium NP (subtracting one from the other) it is also possible to observe a gain in the light intensity redistribution in the NP surroundings caused by a LSPR effect at the metal-semiconductor interface. The combined effect of the increased scattering radius and the LSPR in a “well” distributed array of nanoparticles can be exploited to enhance the light absorption in the red part of the spectrum. As a preliminary result, our FDTD simulations indicate a distribution of a linear array of nanospheres having an average distance of about

180 nm as a good starting point for the optimization of the NP distribution.

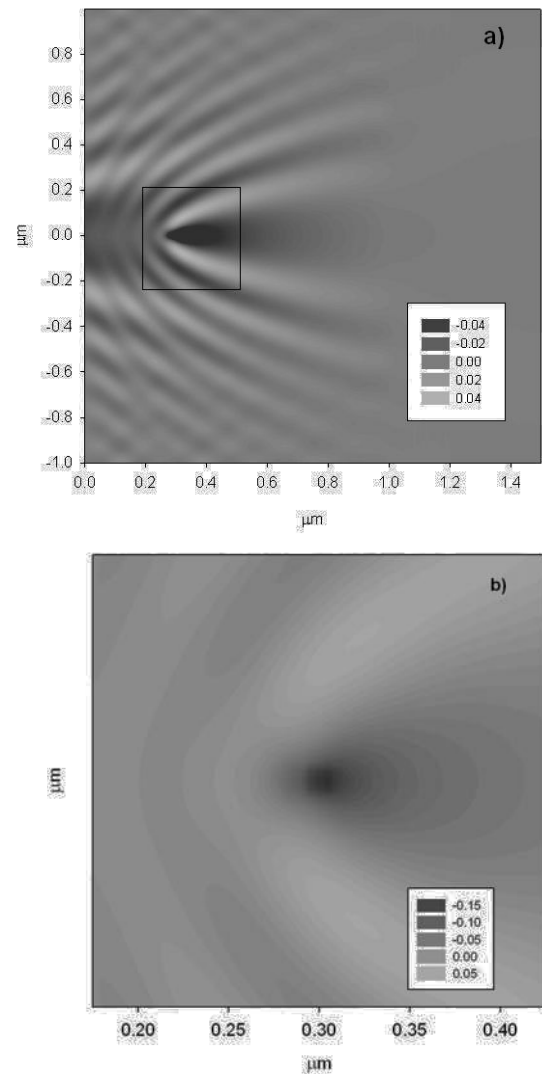


Fig. 4. FDTD Simulation of an Al nanosphere with 10 nm radius embedded in a-Si:H and illuminated with a light of 620 nm wavelength: a) Poynting vector gain caused by the NP, b) Detail of the NP surrounding; see square area depicted in a).

4. Conclusions

We have presented results, obtained by means of an analytic study and a numerical simulation, about the resonant condition necessary to produce a LSPR effect at the surface of metal nanosphere embedded in an amorphous silicon matrix. The study is based on a Lorentz dispersive model for a-Si:H permittivity and a Drude model for the metals. Pointing at the red part of the visible spectrum as the most critical for solar cells applications in terms of light absorption enhancement,

the best choice for the metal nanoparticles appears to be aluminium, indium or magnesium. No difference has been observed when considering a-SiC:H.

FDTD simulation of an Al nanosphere embedded into an amorphous silicon matrix shows an increased scattering radius and the presence of LSPR induced by the metal/semiconductor interaction under red light ($\lambda = 620$ nm) illumination. A further study should include the effect of the nanoparticles (nanoellipsoids) shape in controlling the wavelength suitable to produce LSPR. Also, a further study should be directed to determine an optimal spatial distribution of Al nanoparticles, with variable shapes, capable to enhance light absorption in the red part of the visible spectrum, exploiting light trapping and plasmonic effects.

Acknowledgements

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