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RAPID COMMUNICATION

Upper bounds for the solar energy harvesting efficiency of nano-antennas

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Abstract
The radiation efficiency of nano-antennas is a key parameter in the emerging field of IR and optical energy harvesting. This parameter is the first factor in the total efficiency product by which nano-antennas are able to convert incident light into useful energy. This efficiency is investigated in terms of the metal used as conductor and the dimensions of the nano-antenna. The results set upper bounds for any possible process transforming light into electrical energy. These upper bounds are the equivalent of the theoretical upper bounds for the efficiency of conventional solar cells. Silver shows the highest efficiencies, both in free space and on top of a glass (SiO_2) substrate, with radiation efficiencies near or slightly above 90%, and a total solar power harvesting efficiency of about 60–70%. This is considerably higher than conventional solar cells. It is found that fine-tuning of the dipole dimensions is crucial to optimize the efficiency.

Introduction

Solar energy is expected to deliver a considerable contribution to the solution of human kind's energy problem. At this moment, 90% of the solar cells in the market are based on crystalline silicon wafers. The disadvantage of this technology is the lower efficiency by which the transformation of energy from optical frequencies to low frequencies is performed. Typical efficiencies are in the order of 20–30%. With these efficiencies, if human kind's energy need would be fully satisfied by present day solar cells, the required area would be about 400,000 km^2. Assuming that only 10% of the energy need would be provided by solar energy harvesting, it is easily seen that doubling the efficiency of solar panels corresponds to an area of 20,000 km^2. This is more than half the area of a country like Belgium. The efficiency of solar energy harvesting is a matter of high interest.

In recent years, the idea of using nano-rectennas (nano-antenna or nantenna + rectifier) to harvest solar energy has been suggested. It is claimed that the efficiency of this type of topology may be much larger. The figures mentioned go from a staggering 90% [1], to a more "down-to-earth" 30–40% [2]. It is suggested that the circuits themselves can be made of a number of different conducting metals, and the nano-antennas can be printed on thin, flexible materials like polyethylene, a very cheap and common plastic.

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In this paper, to the knowledge of the authors for the first time, realistic numbers are presented for the maximum efficiencies that can be reached with nano-antenna technology. These numbers are based on a detailed study of a single antenna topology, the basic dipole, for a range of different metals and different sizes.

The total efficiency of nano-rectennas consists of two parts. The first part is the efficiency by which the light is "captured" by the nano-antenna and brought to its terminals. Due to reciprocity, this efficiency is the same as the efficiency by which the antenna is able to convert input power given at its terminals into radiation. This efficiency is thus the radiation efficiency η^{rad} of the antenna. Although this efficiency has been very well studied for traditional antennas, the in-depth characterization of this parameter has not yet been addressed in the nano-antenna research community. To start with, in by far most papers on nano-antennas known to the authors, only gold is considered as metal. Concerning topologies, some information can be found [3,4]. However, Gao [3] considers only two structures of the same length and a very rough Drude model is used in the FDTD solver used, fitting the experimental material parameter data. It can be proven that this affects the efficiencies considerably. Huang [4] uses only a single frequency.

The second part is the efficiency by which the captured light is transformed into low frequency electrical power by the rectifier. At lower frequencies, rectifying circuits are common, but at IR and optical frequencies and in combination with nano-antennas, efficient rectification is a real challenge. A very interesting new technique to realize this transformation has very recently been introduced. M.W. Knight [5] and colleagues have made an optical nano-antenna that also works as a photodetector capable of converting light into either current or voltage. This was done by growing rod-like arrays of gold nano-antennas directly onto a silicon surface—so creating a metal–semiconductor (or Schottky) barrier formed at the antenna–semiconductor interface. The efficiency of the two steps combined was 0.01%. This very low figure is in sharp contrast with the efficiencies mentioned by Kotter [1] and Service [2], and it illustrates the long way still to go before real practical use can be made of solar energy harvesting with nano-antennas.

This paper considers the first step only, the capturing of the IR and optical waves and the transport of the energy embedded in these waves to the terminals of the nano-antenna. It may be clear that the intrinsic radiation efficiency of nano-antennas is a crucial factor in the energy harvesting debate. A three-fold increase in net energy yield would give enormous advantages if applied at a large scale.

It is essential to point out that the interaction between light and nano-antennas in the frequency bands considered can still be analyzed with a high degree of accuracy using classical electromagnetic theory [3,7,8]. The fact that at this small scale, no quantum effects have to be taken into account is really a crucial observation. It means that the concept of an "antenna", a device able to transmit and receive electromagnetic waves rather than particles, still works. Basically, the coupling between an electromagnetic (light-) wave and a nano-antenna (a so-called nantenna) is thus the same as it is at microwave frequencies, and can be studied in the same way.

Although nano-photonics, and especially plasmonics, is a rapidly growing research field [6], the more in depth study of nano-antennas as such has emerged quite recently [9–16]. Following a quite different path, but also quite promising in the area of photovoltaics is the study of the use of so-called nano-wires and nano-tubes, as investigated for example by the group of Lieber [17–19]. A recent review article concerning nanostructures for efficient light absorption and photovoltaics is [20].

From incident wave to received power

The radiation efficiency of an antenna is defined as

$$\eta^{rad} = P^{rad}/P^{inject} = P^{rad}/(P^{rad} + P^{loss}) \quad (1)$$

where P^{rad}, P^{inject}, and P^{loss} are the radiated power, the power injected at the terminals, and the power dissipated in the material, respectively. Both the transmitting and receiving process can easily be described by a very simple equivalent circuit. In receive mode, see Fig. 1, V_{open} is the voltage generated by the receiving antenna at its open terminals. V_{rec} is the voltage seen at the terminals when a current is flowing to the rectifier. This current generates power in the resistors R_{ant} and R_{loss}. The power in the loss resistor P^{loss} is the power actually dissipated in the metal of the antenna. The useful power is the power going to the impedance of the rectifier Z_{rec}. This power is $P_{rec} = \frac{R_{rec}}{2}\frac{V_{open}^2}{|Z_{ant}+Z_{rec}|^2}$. Note that there is also power that is actually scattered, or in other words "received and re-radiated" by the antenna. The maximal power going to the rectifier for a given incident field is under matching conditions, i.e. when $Z_{rec} = Z_{ant}^*$. The power in the rectifier is then $P_{rec} = \frac{V_{open}^2}{8R_{ant}}$. It is easily checked that under matching conditions the ratio of the power given to the rectifier in case of losses ($R_{loss} \neq 0$) and the power given to it in case of no losses ($R_{loss} = 0$) is exactly the radiation efficiency. Since the purpose of this paper is to derive upper bounds for the efficiencies, these optimal matching conditions will be assumed at all frequencies considered. However, note that matching the antenna and the rectifier is a challenge in its own, see [4].

Most solar radiation is in the visible and the near-infrared wavelength region (Fig. 2). In order to form an alternative to state-of-the-art solar cells, nano-antennas have to be designed for this part of the spectrum. Since the material properties in these bands may vary a lot with frequency, studies in the lower frequency bands, as already discussed in literature [1], may be useful to build up necessary know-how, but do not necessarily offer the solutions for the concrete problems at hand in the IR and optical frequency range.

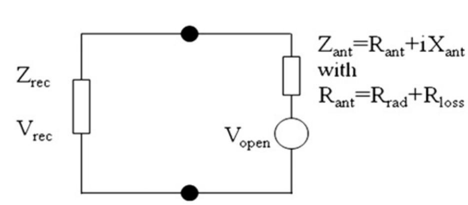

Figure 1 Equivalent circuit for a receiving nano-antenna.

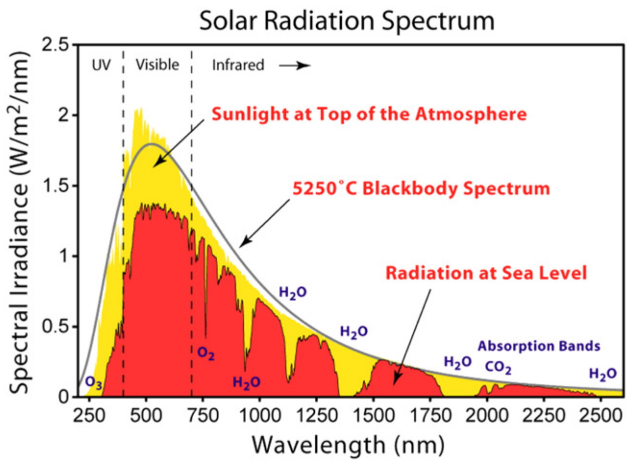

Figure 2 The solar spectrum. The major part of the energy is located in the visible and the near-infrared band. The contribution to the total energy of the part above 1500 nm is very small.

Having the specific application of solar energy harvesting in mind, a new parameter is proposed to characterize the antenna: the total harvesting efficiency. This parameter is defined as

$$\eta^{tot} = \frac{\int_0^\infty P(\lambda,T) \times \eta^{rad}(\lambda) d\lambda}{\int_0^\infty P(\lambda,T) d\lambda} \quad (2)$$

where λ is the wavelength, $\eta^{rad}(\lambda)$ is the radiation efficiency of the nano-antenna as a function of the wavelength, and P is Planck's law for black body radiation

$$P(\lambda,T) = \frac{2\pi hc^2}{\lambda^5} \times \frac{1}{e^{hc/\lambda kT} - 1} \quad (3)$$

where T is the absolute temperature of the black body (in K), h is Planck's constant (6.626×10^{-34} Js), c is the speed of light in vacuum (3×10^8 m/s), and k is the Boltzmann constant (1.38×10^{-23} J/K). In the case of solar energy harvesting, the temperature T is the temperature of the surface of the sun. The values calculated with (2) have to be considered upper bounds since practical solar radiation is filtered by the atmosphere. However, they provide an excellent figure of merit for the nano-antenna topologies investigated.

Although material properties at these frequencies are well-known, to date, as far as the authors know, no systematic analysis or measurement campaigns have been done in the area of nano-dipoles concerning the use of different materials. Also very scarce information is available on radiation efficiencies of these structures. This work reports on a systematic numerical study of the radiation efficiency of IR and optical nano-dipoles for five different metals. No Drude model is used but the experimental values of the material parameters are used directly. The solver used in this work is based on the solution of volumetric integral equations with the Method of Moments (MoM) [21,22]. Following the advice given by Vasylchenko [23], it was first benchmarked against another solver and against measurements. The second solver is well-known in the optical research community [24]. It uses a totally different modeling technique, i.e. the Finite Difference Time Domain technique (FDTD).

Benchmarking of the simulation tool against measurements was done as part of previous studies [8,25], and will not be repeated here. For a gold monomer topology with three different lengths, extinction cross sections of arrays of $50 \times 50\ \mu m^2$ with 2 μm pitch were measured using a Fourier transform infrared spectrometer equipped with a microscope. The agreement found between simulations and measurements in [25] is excellent. The main conclusion of the benchmarking is that both solvers yield very similar results, and correspond quite well with the measurements. There is only a slight frequency shift.

The dipole topology studied in this paper is depicted in Fig. 3. This topology is sensitive mainly to an electric field polarized in its longitudinal direction. Its reaction to this field is strongly depending on its size and the metal used. Any excitation of this dipole will excite two plasmons propagating in opposite directions and interfering with each other. This is clearly illustrated in Fig. 4, where the longitudinal current on an Aluminum dipole evaluated just below the top surface is depicted for different lengths of the dipole in free space. The dipole is considered in transmit mode and is excited in the middle. The wave effect is obvious. It is clearly seen that the excitation in the middle generates a plasmonic wave, which reflects at the end, causing interference. This explains the constant distance of the null (in dark blue) from the ends of the dipole. From the current pattern the wavelength of the plasmon inside the dipole is found to be around 160 nm. It is also seen that the length of the dipole determines the amplitude of the current. In Fig. 4 the largest currents are found for the dipoles of length 100 nm and 320 nm, where a clear resonance occurs.

For this dipole topology, a second benchmarking was done. In Fig. 5 efficiencies obtained with MoM and FDTD are compared for a 250 nm long gold dipole. It is seen that there is an excellent agreement.

The main target of this paper is to derive upper bounds for the efficiencies that can be reached for any possible process by which the IR and optical energy can be transformed.

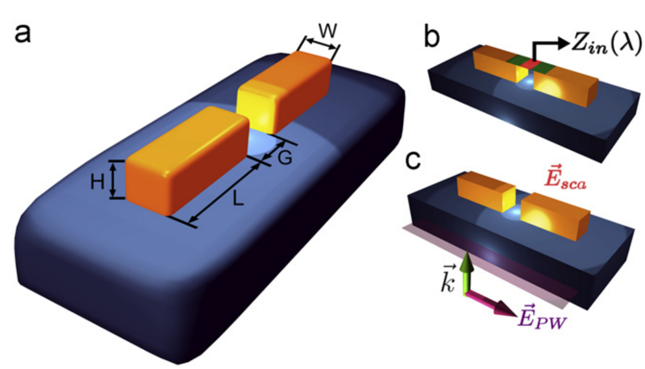

Figure 3 The dipole model studied. (a) W and H are set equal to 40 nm and the gap G is fixed at 10 nm, which is the same value as used in [3]. (b) The dipole as transmitting antenna with a model for the feeding structure located in the gap. (c) The dipole as receiving antenna excited by a plane wave, which is the case of interest.

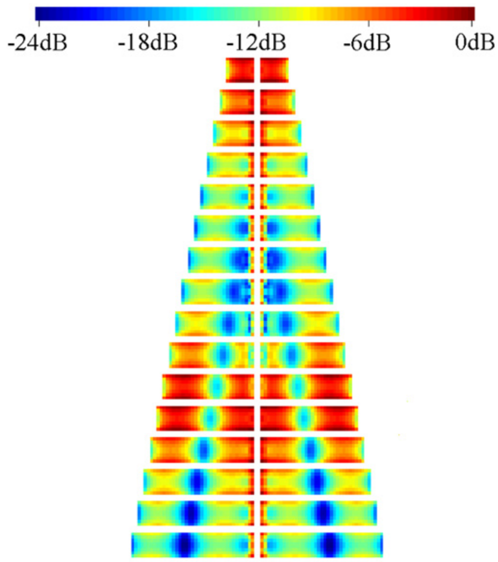

Figure 4 Current distribution on an Al dipole for lengths varying between 100 and 400 nm in steps of 20 nm. Width and height are 40 nm. The frequency is 1000 THz and the excitation is a constant current applied in the gap. The interference patterns and resonances of the plasmonic waves can be clearly seen.

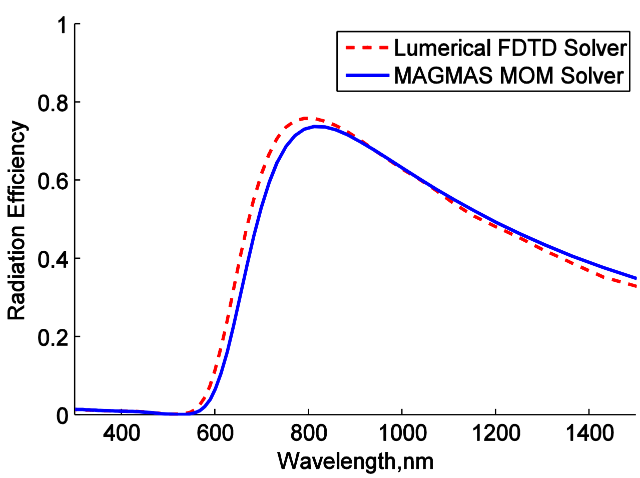

Figure 5 Comparison between FDTD and MoM for a gold dipole, L=250 nm.

These upper bounds are the equivalent of the theoretical efficiency upper bounds for conventional solar cells. This is done for five metals, including the most popular ones used in plasmonics. It is evident that our techniques can also be used for other metals.

Efficiencies in vacuum

The results of a first comprehensive study of the antenna are given in Fig. 6. There, efficiencies are given for silver, gold, aluminum, copper, and chromium, respectively, not taking into account the effect of any substrate layer. The permittivity of the materials used in the simulations is obtained through experimental ellipsometry. They are presented in Fig. 7. Both the radiation efficiencies as a function of wavelength, and the total harvesting efficiency are given. The results are revealing. It is clearly seen that chromium is not suited at all for energy harvesting. The maximum efficiency is in the order of 20%. Copper reaches efficiencies in the order of 60–70%, but the bandwidth is rather limited. This is reflected in the total efficiency which reaches a maximal value of about 30% for a dipole length of ca. 300 nm. The same is observed for gold, the material most used in this area. Its efficiencies are a bit higher than for copper, in the order of 70–80%, but the total efficiency reaches a maximum of about 35% for a dipole length also of 300 nm. Aluminum performs quite well and reaches efficiencies of 60–70% around 500–600 THz, which is in the middle of the visible range. Also, it shows reasonable efficiency values over the whole frequency band considered (200–1000 THz), in the order of 20–50%, yielding total efficiency values of about 50% for a whole range of dipole lengths. The highest values are obtained for silver. In the lower region of the band considered, silver is by far superior over all other metals. It reaches efficiencies over 80–90% in a remarkably wide band. Only at higher frequencies, aluminum outclasses silver. The maximal total harvesting efficiency reached by silver is 65.4% at a dipole length of 200 nm. Note also that clearly the frequency dependency of the efficiencies is totally different for the different metals, due to the specific frequency dependency of their complex permittivity. Also, since the efficiency is strongly depending on the imaginary part of the permittivity (corresponding to conductivity), the use of a proper value in the calculations is mandatory. Simple Drude models that may result in serious errors, up to about 100% [3], are incapable of providing a good prediction of the efficiency.

Efficiencies on a substrate layer

Since nano-dipoles have to be fabricated on a supporting layer, in a second study, the effect of a glass substrate is investigated. Also for the glass substrate the measured permittivities are used in the analysis. For the frequency range considered this permittivity is almost constant and about 2.1. The efficiencies as a function of frequency (or free space wavelength) for different thicknesses of the substrate are plotted in Fig. 8. It is clearly seen that the substrate does have a major effect. The efficiencies obtained are in most cases lower than in case a substrate is not present. Only for very large thicknesses (in principle going to infinity), both the efficiencies as a function of wavelength and the total efficiencies recover to reach about the same values as in the case without substrate. This observation does not pose a problem since the substrate layers used in practice are indeed very thick compared to the wavelength. The total efficiencies on a half space of substrate material are 61.6% for silver, 34.3% for gold, 50.3% for aluminum, 29.5% for copper, and 9.4% for chrome. This lowering effect is caused by the interference of the field waves reflected at the interfaces of the substrate. This is clearly illustrated in Fig. 9, where for three materials and for two selected frequencies, the efficiency is given as a function of the substrate thickness. The relation between

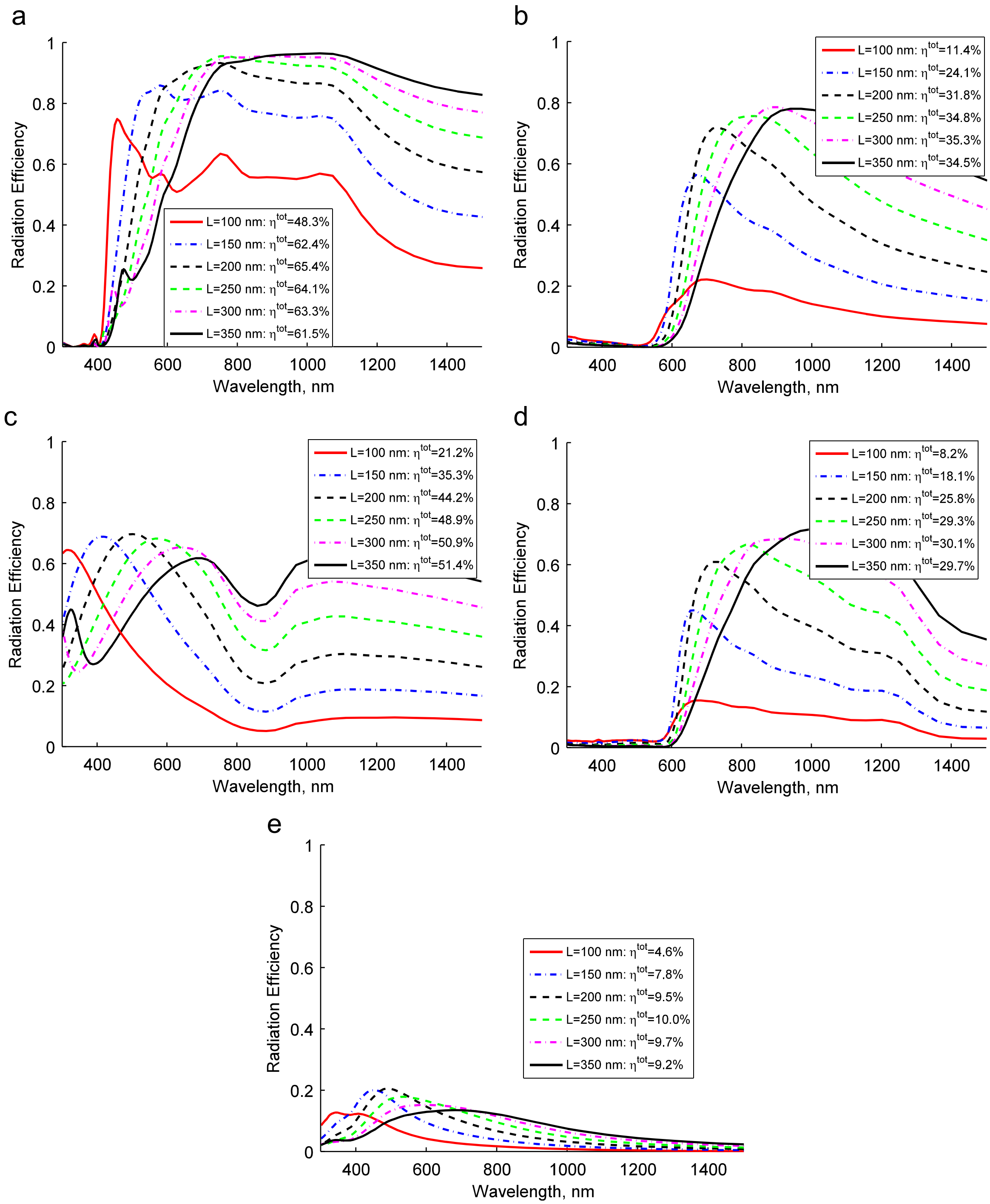

Figure 6 Radiation efficiency of nano-dipole as a function of free space wavelength and dipole length: (a) silver, (b) gold, (c) aluminum, (d) copper, and (e) chromium.

the radiation efficiency and the thickness clearly shows an oscillating behavior. Studying Fig. 9 in more detail it can be seen that the oscillation is governed only by the frequency and is independent of the material of the dipole. Also the period of the oscillation is about 400 nm in the first graph and about 350 nm in the second graph. This clearly proves that the effect is caused by the either constructive or destructive interference of the waves reflected at the two boundaries of the glass substrate. Also the specially shaped peaks can be explained. This behavior stems from the fact that the glass substrate is actually a dielectric slab waveguide. The introduction of a new propagating mode in this waveguide with increasing thickness of the substrate goes along with an extra power loss. This effect generates the discontinuity in the derivative of the efficiency function. For a certain thickness the surface wave is maximally

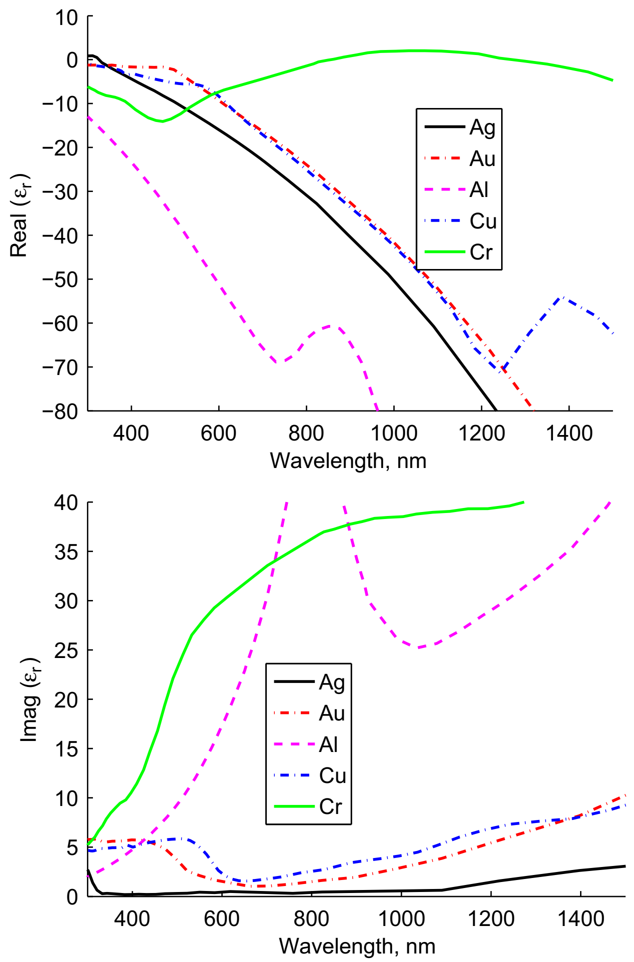

Figure 7 Real part (top) and minus the imaginary part (bottom) of permittivities of silver, gold, aluminum, copper, and chromium.

excited and the efficiency is minimal. The major conclusion of this second study is that (1) the presence of a substrate overall decreases the efficiencies, and (2) considering the same thickness of the glass, again silver is by far superior over the other materials.

Extraction of the effect of the material properties

It is worthwhile to explain these results from a physical point of view. Through (2), the total harvesting efficiency is completely determined by the radiation efficiency, which is a function of frequency/wavelength. The key issue is to try to reach the highest efficiencies around 500 nm, where the solar irradiance is the largest. This can be done by choosing the proper dipole length. The reason is that this length is one of the main factors that determines the response of the dipole. However, the properties of the material also have a strong effect on the response. That explains why a different optimal length is found for different materials. Also, efficiency is totally depending on losses, and losses are determined by the imaginary part of the permittivity.

It is possible to approximately assess any arbitrary material with respect to its harvesting capabilities. For this, we need to separate the effect of the plasmonic waves traveling along the dipole, and consequently the complex interferences, from the effect of the material. This is easily done for an elementary dipole antenna in free space, i.e. a dipole with very short length with respect to the wavelength. For such an antenna the radiated power is proportional to the square of the dipole moment

$$P^{rad} = \frac{\omega^2 \mu_0 \sqrt{\mu_0 \varepsilon_0}}{12\pi} \left(\int_V \mathbf{J} dV \right) \cdot \left(\int_V \mathbf{J}^* dV \right) \quad (4)$$

where V is the volume of the dipole, and $\mathbf{J}$ is the vector current distribution flowing in it. Using the relation between the total electric field and the (polarization and conduction) current in the material, i.e. $\mathbf{J} = j\omega(\varepsilon - \varepsilon_0)\mathbf{E}$, the losses can be expressed as

$$P^{loss} = \mathrm{Re}\left(\frac{1}{2} \int_V (\mathbf{E} \cdot \mathbf{J}^*) dV \right) = \mathrm{Re}\left(\frac{1}{2} \int_V \left(\frac{\mathbf{J}}{j\omega(\varepsilon - \varepsilon_0)} \cdot \mathbf{J}^* \right) dV \right)$$
$$= \frac{-\varepsilon_{im}}{2\omega((\varepsilon_{re} - \varepsilon_0)^2 + \varepsilon_{im}^2)} \int_V (\mathbf{J} \cdot \mathbf{J}^*) dV \quad (5)$$

so that the efficiency becomes

$$\eta^{rad} = \frac{1}{1 - (6\pi\varepsilon_{im}/\omega^3 \mu_0 \sqrt{\mu_0\varepsilon_0}((\varepsilon_{re} - \varepsilon_0)^2 + \varepsilon_{im}^2))(\int_V (\mathbf{J} \cdot \mathbf{J}^*) dV / (\int_V \mathbf{J} dV) \cdot (\int_V \mathbf{J}^* dV))} \quad (6)$$

For a standard short dipole with cross section A and length L carrying a standard linear "rooftop" type of current, the integrals over the current density can be analytically calculated, yielding

$$\eta^{rad} = \frac{1}{1 - (8\pi\varepsilon_{im}/\omega^3 \mu_0 \sqrt{\mu_0\varepsilon_0}((\varepsilon_{re} - \varepsilon_0)^2 + \varepsilon_{im}^2))(1/V)}$$
$$= \frac{1}{1 - (\varepsilon_{im,r}/((\varepsilon_{re,r} - 1)^2 + \varepsilon_{im,r}^2))(8\pi/k_0^3 V)} \quad (7)$$

Note that this expression confirms the fact that the imaginary part of the permittivity always has to be negative, as observed in Fig. 7. Keeping the electrical volume of the dipole constant and small, it is easily seen that the efficiency becomes dependent only on the permittivity. From a material perspective, the key issue is to get the ratio $\frac{-\varepsilon_{im,r}}{((\varepsilon_{re,r}-1)^2 + \varepsilon_{im,r}^2)}$ as low as possible. This is reached for a very low imaginary part with respect to the real part of the permittivity, or, when the real part approaches 1, for a very large imaginary part. Expression (7) is illustrated in Fig. 10 for the five metals considered while the electrical volume is kept constant at $k_0^3 V = 1$. This means that a different dipole size at each frequency is used in order to remove the effect of the topology itself. The strong dependency of the efficiency on the permittivity is clearly visible. The steep rise around 400 nm and 600 nm, for Ag on the one hand and Au and Cu on the other hand, is also clearly seen in the efficiency curves given in Figs. 6 and 8, but modulated there by the plasmonic waves and resonance. Fig. 10 reveals why silver outclasses the other materials, and why aluminum is a good alternative. They keep a high efficiency in the whole frequency band of importance. It is important to note that expression (7) can be easily used to preliminary assess the intrinsic harvesting capabilities of

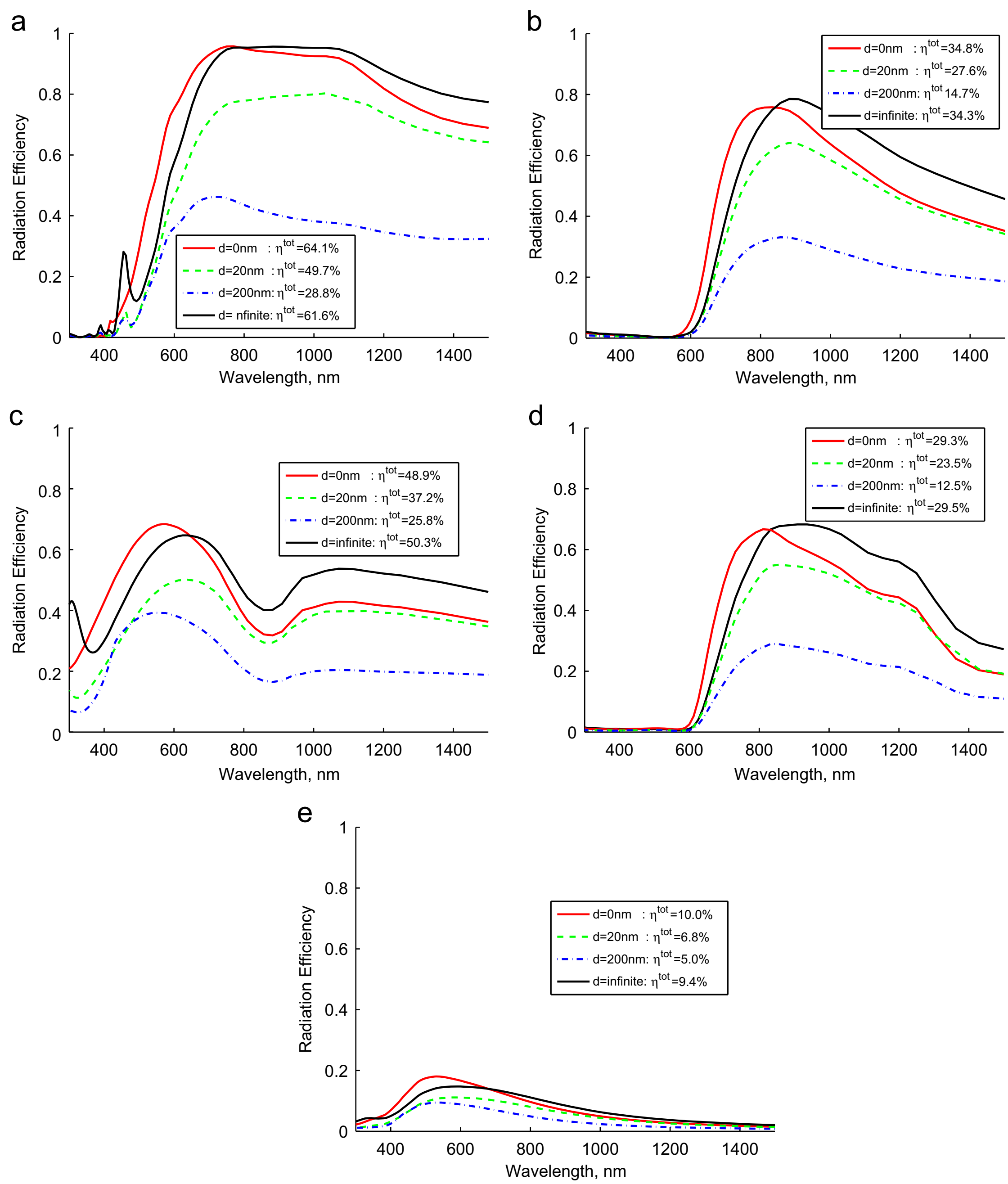

Figure 8 Efficiencies for different materials for a 250 nm dipole in terms of the thickness of the glass substrate, (a) silver, (b) gold, (c) aluminum, (d) copper, and (e) chromium.

arbitrary metals in a first approximate way. A high efficiency is needed in a band as wide as possible around 500 nm. This is only the case for silver and aluminum.

Conclusion

In this study upper bounds are derived for the efficiencies by which energy can be harvested from the sun using nano-antenna technology. To this goal, the parameter "total harvesting efficiency" is introduced. Both dipoles in free space and on a glass substrate are considered. For silver nano-dipoles, a maximum of about 60–70% is found. It is an open question whether it is possible to construct alloys with even lower losses at plasmonic frequencies, and thus higher efficiencies. A simple approximating formula is derived to assess the intrinsic harvesting capabilities of a material. Several challenges remain. Silver is more susceptible to oxidation, which can completely destroy its superiority, and which is one of the reasons why gold is so popular in this field. This can be solved by embedding it within the glass substrate, which requires to develop alternative fabrication

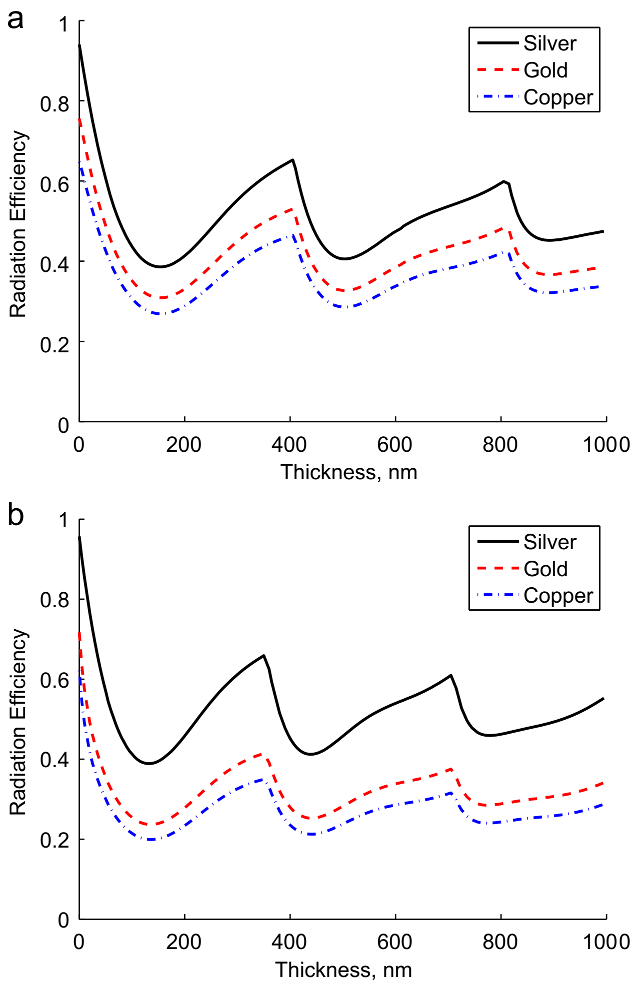

Figure 9 Efficiencies for different thicknesses of glass substrate, simulated, (a) 350 THz, (b) 400 THz. The resolution of the calculation is 5 nm.

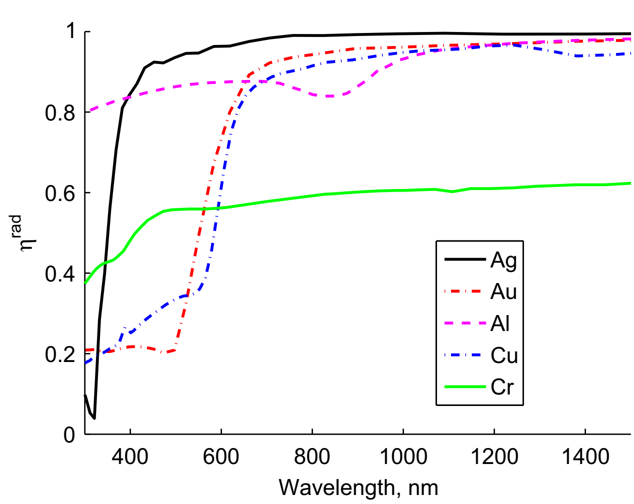

Figure 10 Small dipole efficiencies for different materials.

processes and technology. The problem can also be solved by using aluminum, which has a transparent oxide, but a bit lower total efficiency of around 50%.

Acknowledgments

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