

***Final Draft***  
of the original manuscript:

Hedayati, M.K.; Faupel, F.; Elbahri, M.:  
**Tunable broadband plasmonic perfect absorber  
at visible frequency**  
In: Applied Physics A (2012) Springer

DOI: 10.1007/s00339-012-7344-1

# Tunable Broadband Plasmonic Perfect Absorber at Visible Frequencies

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## Abstract

Metamaterials and plasmonics as a new pioneering field in photonics joins the features of photonics and electronics by coupling photons to conduction electrons of a metal as surface plasmons (SP). This concept has been implemented for variety of application including negative index of refraction, magnetism at visible frequencies, cloaking devices amongst others. In the present work, we used plasmonic hybrid material in order to design and fabricate a broad-band perfect plasmonic metamaterial absorber in a stack of metal and Copper-PTFE (Polytetrafluoroethylene) nanocomposite showing average absorbance of 97.5% in whole visible frequencies. Our experimental results showed that the absorption peak of the stacks can be tuned upon varying the thickness and type of the spacer layer due to the sensitivity of plasmon resonance to its environment. To the best of our knowledge this is the first report of plasmonic metamaterial absorber based on copper with absorption around 100% in entire visible and NIR.

## 1. Introduction

The general attention in solar use fast raised in the mid-70's due to the renewed interest in substitute energy resources which resulted from the "oil crisis" [1]. Graded structure made of metal-dielectric composite along with anti-reflectors were the basic of the developed structure for solar absorption which were fabricated by electroplating or vacuum deposition methods (Details can be found in [1]). Nowadays, due to the fast growing field of nanotechnology and great demand for nanoscale system, the trends to realize a system showing black absorption for solar and sensoric purposes with sub-wavelength dimension increased. However, so far most of the applied techniques (e.g. perforated metallic films[2], grating structured systems [3] and metamaterials [4-6] ) are either costly or has low

fabrication tolerance and their absorption resonance is narrow-band which limits their utility for energy harvesting. Recently, we show experimentally for the first time fabrication of a broadband perfect plasmonic absorber in a stack of gold and gold-SiO<sub>2</sub> nanocomposite showing nearly 100% absorbance at visible frequencies [7-8]. In this report, we implemented the same idea but in a polymeric nanocomposite and replacing Gold with a Copper to reduce significantly the material cost. In copper based absorber, the bandwidth is broader and since the Copper is cheaper than Gold, the new developed absorber is more cost effective for practical application. In addition, the average value of absorption in visible spectrum is above 97% which is the best reported broad band plasmonic perfect absorber so far.

## 2. Experimental procedure

A cylindrical vacuum chamber was used for sputtering of the metal and polymeric film. By installing two magnetron with an angle of 50°, co-sputtering was done in order to make a nanocomposite out of a Copper and PTFE targets. Simultaneous sputtering from different sources allows deposition of nanocomposites with different filling factors (ff) and thicknesses. For acquiring a uniform thickness for the film and homogeneous metal distribution for the composite, all depositions were done on the samples attaching to a rotatable sample holder. For dielectric deposition (PTFE in this case) a RF power and for conductive targets (Copper) a DC power supply were used. The details of co-sputtering methods can be found in our former reports [9]. Thickness measurements was performed using Dectak 8000 profilometer. Optical analysis carried out by a UV-Vis-NIR spectrometer (Lambda900, Perkin Elmer). Although Aluminum was used as a mirror for reflection measurement, the data was normalized to a perfect reflector in order to achieve absolute value of reflection. Since all the base film thickness was 100nm which is far greater than the skin depth of copper, it was assumed that the transmission is zero. Therefore, all calculation of absorbance is based on:  $A+R=100\%$ , where A stands for absorption and R is reflection.

### 3. Results and discussion

Schematic of the structure which was used in this work is shown in Figure 1 (a). It is composed of an optically thick copper film as base layer (100nm) and a nearly percolated Copper-PTFE nanocomposite as top layer which are separated by a dielectric inter-layer (PTFE). TEM of the nanocomposite used in this work presented in Figure 1 (b). One can see that the distance between the particles are very small and the composite is near-percolation.



Figure 1(a): Schematic draw of the perfect absorber structure

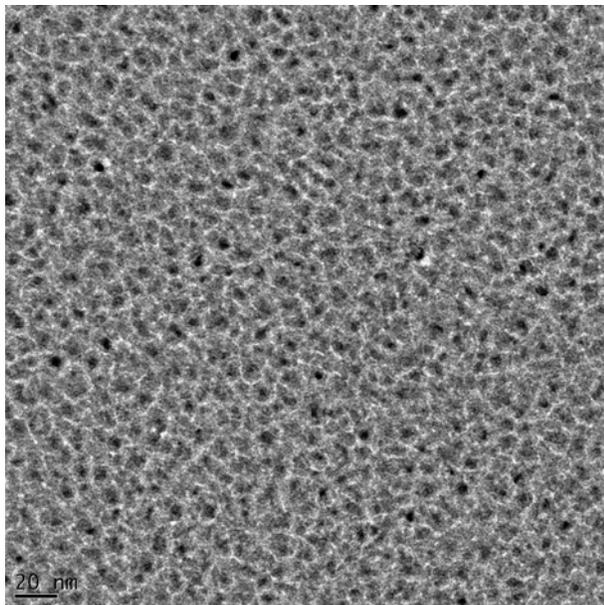


Figure 1(b): Top-view TEM image of the near percolated copper-PTFE nanocomposite.



Figure 1(c): Photo of the perfect black absorber in comparison with bare copper film.

Figure 1 (c) show the photograph of the optimized sample compare to the bare copper film where one can see the black appearance of the highly absorber system compare to the shiny surface of metallic film. Optical measurements showed that 20nm spacer layer of PTFE gives the best performance of the device and thickening or thinning the thickness of interlayer reduced (increases) the absorption (the reflection) of system. Figure 2a shows the absorption spectra of 20nm Cu-PTFE composite on 100nm copper base layer separated with different thickness of interlayer. It is obvious that changing the spacer layer thickness from the optimum value of 20nm will alter the efficiency of the device and the absorption drop. We attributed the drop to the lack of coupling of the top nanoparticles and the base layer. Indeed, in such a system, excitation of anti-parallel currents between nanoparticles (embedded within the nanocomposite) and base layer (known as magnetic resonance) induced by dipole-image interaction will trap the light within the inter-layer. However, it seems that upon increment or decrease of spacer-layer thickness, dipole-image interaction becomes less efficient and results in a drop in absorption [6-7]. Deposition of the composite on a bare base layer further support the mentioned idea in a sense that when there is no distance between the film and nanoparticles, no light can be confined and therefore the reflection enhances (Figure 3 (green curve)).

The optical response of such a stack is not only dependent on the spacer layer thickness rather than the filling factor of the composite plays a crucial role in the efficiency of film. UV-vis measurement of the samples prepared with different filling factor showed that the best performance achieved when the ratio of sputtering rate of copper to PTFE is 2.33. On the other hand, changing the sputtering ratio to 2.0 or 2.66 reduce the overall absorption of the device. Figure 2 (a) shows the absorption spectra of the stacks with different sputtering ratio of Copper and PTFE. It is obvious the highest absorption is happened when the ratio is 2.33. When the ff raises far above the percolation threshold the reflectivity of the upper layer goes up and consequently the optical response of the stacks become similar to thick copper film. In other word, as soon as the

composite starts turning to a continuous metal film, the light can not pass through the top layer anymore and it acts as a metal reflector. Therefore the absorption of the structure dramatically drops. Comparing the current results with our previously report on gold base perfect absorber, one can see that the broadness of the peak is more than that of gold one. In other words, the average value of absorption in the visible range (400-750nm) is around 97% which shows the higher efficiency of Copper-absorber compared to Gold one which shows the higher efficiency of former one. In fact, the filling factor of optimized condition in copper system (70%) is far above that of gold (40%) and therefore it is expected to be highly reflective. However, a broader resonance and perfect absorption was observed which we attributed to the plasmonic damping of copper which occurs due to interband processes via electron photon as well as electron-electron scattering [10].

The role of magnetic resonance in the high absorption of the structure was shown in our last work [7] however one can not rule out the significance of interference in the low reflectivity of such a structure. The influence of interference in perfect absorber has been recently studied by Chen [11]. He showed in a typical metamaterial absorber there is minor near-field interaction or magnetic response among the neighboring metal structures. In addition, the surface currents with anti-parallel directions originated from the interference and superposition, rather than excited by the magnetic component of the incident electromagnetic fields [11]. We believe that Chen's theory might hold for grating on a film or resonator designed for MIR range of frequencies but it can not be applied in our structure. Because in our device, the inter-particles distance is less than 5nm (Figure 1 (b)) and hence one can not neglect the huge light confinement between the particles beside the light trapping in the interface [12]. In addition, the resonance band of our system is few hundred nanometers (i.e. very broad low reflection) which can not be explained only by interference phenomena.

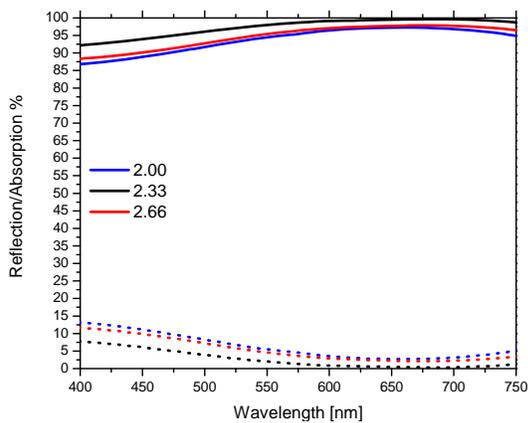


Figure 2(a): Absorption (solid lines) and reflection (dot lines) spectra of the near percolation nanocomposite with three different sputtering ratio of polymer and metal on a 100 nm gold film coated with 20nm PTFE layer as spacer

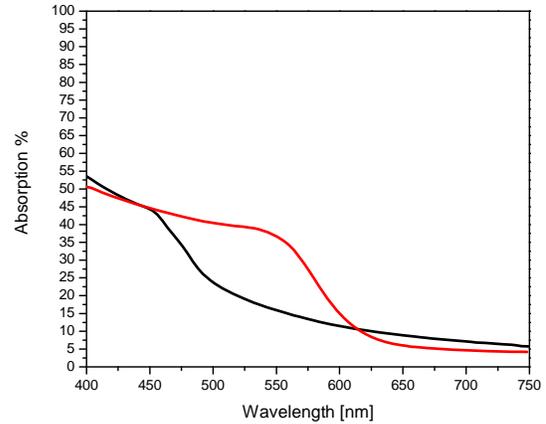


Figure 2(b): Absorption spectra of 20nm Cu-PTFE composite (black) with sputtering ratio of 2.0 on 20nm PTFE on glass in comparison of 100nm copper film (red).

Measuring the optical response of the metal alone as well as the composite film with the same condition deposited on glass substrate showed that the high absorption of the system is not originated neither from the composite alone nor from the metal. Figure 2 (b) shows the absorption spectra of composite and bare copper film on glass. It is clear that the average absorption value in both cases is less than 30% which is far below the 97% of copper-perfect absorber. This results further support the idea of plasmon coupling in the highly absorber structure and shows that the Ohmic losses of the device within the metallic particles due to the localized particles plasmon resonance of copper nanoparticles is not the dominating process of the overall absorption.

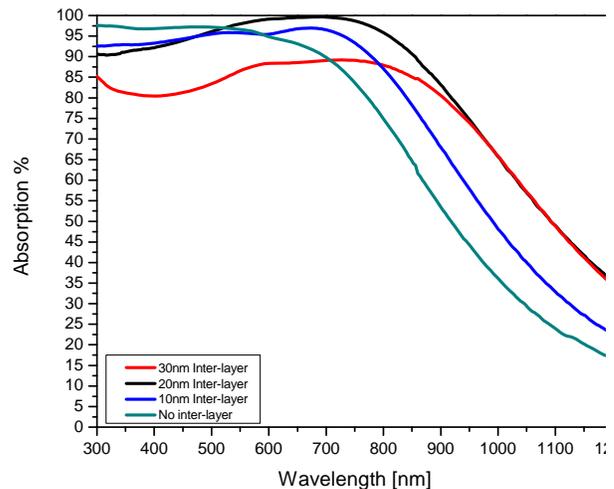


Figure 3: Absorption spectra of 20nm Cu-PTFE composite on PTFE spacer layer with different thickness on 100nm Cu. Green curve show the composite on a base layer without any inter-layer for comparison.

In addition, the optical study of the system with different spacer layer thickness further support the significant role of dipole-image interaction in perfect absorption of our

developed system (Figure 3). As it was mentioned above, increasing the distance between the metal film and the nanocomposite by adding a thick spacer layer, disturbs the resonant condition and results in a weaker coupling. Different to our previous report[7] a critical spacer layer of 20nm was observed where the absorption width and intensity is maximized. The significant drop in the absorption observed when the thickness of spacer layer exceeds 50nm which we attributed to the lack of efficient dipole-dipole interaction. Indeed, the difference of spacer layer in Gold and Copper system can be attributed to two effects. Firstly, the refractive index of PTFE is less than that of SiO<sub>2</sub>. Secondly, the plasmon resonance of Gold and Copper response differently to the certain dielectric [13]. Beside the two mentioned parameters, the ratio of gold to SiO<sub>2</sub> matrix in gold-perfect absorber was less than the ratio of Copper in PTFE in copper-one. Indeed, all of the difference originated from the different plasmon resonance of Gold and Copper.

#### 4. Conclusions

In summary, we have developed and studied a new plasmonic metamaterial with almost perfect absorption of light in visible frequencies. Structure out of polymer matrix of composite and spacer layer and Copper as a metallic component showed that the perfect absorption can be achieved with other system rather than Gold. However the thickness of the inter-layer and the filling fraction of metal should be varied depends on its dielectric function. We concluded that interference theory can not explain the broadband perfect absorption of our developed system since both the particle size and inter-particles distance is too small that one can not neglect the strong electrical filed confinement within the structure. Due to the simple fabrication technique that we employed, the production cost is very low compare to the competitive methods such as e-beam lithography. In addition, the higher fabrication tolerance of our highly absorber structure makes it an outstanding candidate for future application in photovoltaic and sensor.

#### Acknowledgements

Funding by the German Research Foundation (DFG) through the projects EL 554/1-1 and SFB 677 (C1,C9) are acknowledged. M.E. would like to thank the Initiative and Networking Fund of the Helmholtz Association's (grant No. VH-NG-523) for providing the financial base for the start-up of his research group.

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