Experimental mapping of plasmonic patterns based on second harmonic generation microscopy

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Abstract

We demonstrate that second harmonic generation (SHG) microscopy constitutes a unique, important, user-friendly and widely-applicable method for visualizing plasmonic hotspots in metamaterials. Most importantly, though, SHG imaging has an impact, *literally*. Indeed, we prove that SHG imaging of samples can imprint the plasmonic patterns on the surface of the structures for subsequent imaging with structural characterization techniques such as SEM or AFM. Consequently, the plasmonic patterns can be mapped with the resolution of scanning probe techniques and thus the limitations imposed by the diffraction of light can be removed altogether.

1. Introduction

Many of the optical properties of metal-based metamaterials are determined by the excitation of surface plasmon resonances, which are collective oscillations of surface charges. These electron surface waves have the same frequency as the excitation electromagnetic field, but much shorter wavelengths, which allows their manipulation at the nanoscale. In other words, with the help of plasmons, light can be captured, modified and even stored in nanostructures. This emerging nanotechnology could find applications to new cancer treatment methods,[1,2] biochemical sensing,[3,4] solar cells,[5] optical computing,[6,7,8] negative refractive index materials,[9] and invisibility cloacks.[10] The imaging of surface plasmons provides a direct approach to map and understand the local electric fields that are responsible for the unusual electromagnetic properties of metamaterials; the far-field imaging of surface plasmons, however, is limited by the diffraction limit of light.

Currently, several experimental methods are available to achieve imaging resolution on plasmons beyond that imposed by the diffraction limit. For example, sub-diffraction-limit imaging of plasmons was reported, whereby a silver superlens was used – a device that relies on refraction in a negative refractive index material for focusing.[11,12] Another example is found in sub-wavelength optical imaging through a metallic nanorod array; the near-field components of dipoles in the source plane are plasmonically transferred through the rod array to reproduce the source distribution in the image plane.[13] However, the most widely used method for sub-wavelength imaging so far is scanning

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near-field optical microscopy (SNOM).[14] Nevertheless, SNOM requires a very long scanning time for large sample areas and high resolution imaging. Generally speaking, while there are experimental methods to achieve resolution beyond the diffraction limit of light, they come at a considerable increase in both cost and complexity.

2. Second Harmonic Generation Microscopy for mapping plasmonic patterns

In Fig. 1, we compare a SHG microscopy image with the results of a computational analysis used to determine the spatial distribution of the local field enhancement at the fundamental frequency. In our study, we use MAGMAS' and RSoft's DiffractMOD software tools, which provide the surface currents and electromagnetic field distribution, respectively, generated upon the interaction between light and chiral metallic nanostructures. In both cases, the numerical maps of the local field enhancement at the fundamental frequency match the experimental mapping of SHG sources. Consequently, the origin of the SHG can unambiguously be attributed to maxima of the surface charge density or local field enhancement, which in turn depend on the geometry of the structures and their material parameters. Our results suggest that SHG microscopy can be used efficiently for mapping the local field enhancement in nanostructured metamaterials.[15-18]

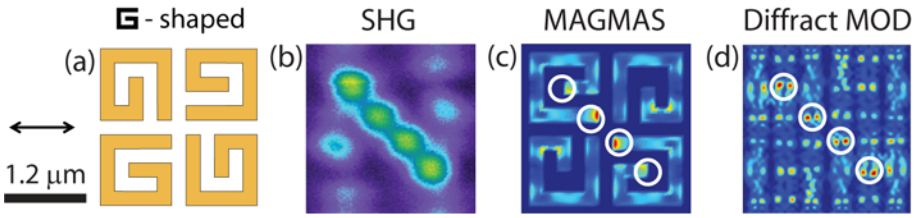

Fig. 1: In gold G-shaped nanostructures, mapping of the SHG sources matches the mapping of surface field enhancements at the fundamental frequency. In (a), the geometry of the sample configuration. Experiments and calculations were performed for linearly polarized light along the direction of the arrow. In (b), the SHG microscopy results. In (c), numerical simulations of the electric currents at the surface of the nanostructures calculated with the MAGMAS software. In (d), electric field at the surface of the nanostructures obtained with the Diffract MOD software. The color-coded intensities increase from dark blue, through green, then yellow, to red. For better comparison with the SHG results, the electric currents and field that are shown in (c) and (d), respectively, have been squared.

3. Hotspot Decoration (HD) Mapping of plasmonic patterns

In Fig. 2, we have demonstrated that, in measuring plasmonic patterns in nanostructures, the diffraction limit of light can be completely overcome. Indeed, upon illuminating nanostructures made of nickel or palladium, the resulting surface plasmon pattern is imprinted on the structures themselves. This imprinting is done through displacing material from the nanostructure themselves to the regions where the plasmon enhancements are the largest. In this manner, the hotspots are effectively decorated, allowing for subsequent imaging with scanning electron microscopy (SEM) or atomic force microscopy (AFM). The imprinting method is quite unique, combining aspects of both imaging and writing[15] techniques. The combination offers a resolution on local field enhancements that can, in principle, be brought down to that of the AFM.

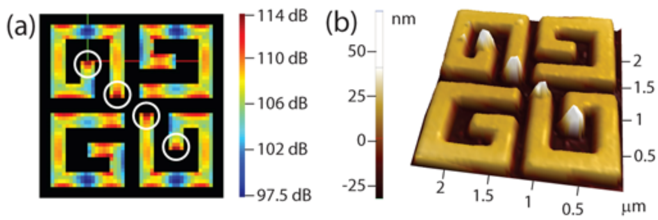

Fig. 2: In nickel G-shaped nanostructures, the plasmonic patterns are imprinted on the surface. In (a), MAGMAS numerical simulations of the electric currents (not squared) at the surface of the nanostructures. In (b), atomic force microscopy reveals that the hotspots have been decorated.

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4. Conclusion

Although SHG microscopy and HD Mapping of the plasmonic fields have first been demonstrated in G-shaped nanostructures, these methods are not restricted to this particular geometry. Indeed, in our presentation, it will be shown that either one of these methods (or both) can be used to map plasmonic hotpots in I-shaped, O-shaped, U-shaped and L-shaped nanostructures, on materials, such as Au, Ni, Pd and Y-Ba-Cu-O ceramic. We will present evidence that, by trapping the plasmon within the structures, SHG microscopy can achieve a resolution of λ/4, while a resolution of λ/8 was observed for HD Mapping. Both methods are very user-friendly and can be implemented by adapting a standard confocal for second harmonic imaging. Because such instruments are whidely available within research facilities, we belive that SHG microscopy and HD Mapping can quickly be adopted by other laboratories.

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