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Grain structure for TERS microscopy

Atsushi Taguchi[1,2], Satoshi Kawata [1,2]

[1] Applied Physics, Osaka University, [2] RIKEN
E-mail: taguchi@ap.eng.osaka-u.ac.jp

Tip-enhancement has become an essential technique to achieve an extremely high spatial resolution beyond the diffraction limit of the light in probe scanning microscopy [1,2]. However, the reproducibility in obtaining an extraordinary large enhancement of the electromagnetic field is not high [3]. There have been a number of reported FDTD calculations for a variety of metallic structures with different shapes, sizes, alignments and materials of particles and tips. Although intense fields have been shown as hot spots in those calculations, such high values are not necessarily reproduced in real experiments. A plausible reason comes from the fundamental nature of the resonance modes of surface plasmons that the spectral band narrows as the field intensity increases. Unlike the lasing resonator pumped with a gain medium, plasmonic enhancement effect is a linear process, thus, a strong enhancement comes with a price of spectral bandwidth, which means Raman lines can be easily detuned from the bandwidth of the plasmonic system [4].

Another issue that has not been discussed much is the coupling/decoupling of near-field photons with far-field propagating photons and *vise versa*. Excitation of the near-field is usually made with far-field optics, and the detection of the near-field light-matter interaction is also done at the far-field. The coupling efficiency between near- and far-field photons is thus an important issue for the practical implementation of tip-enhancement Raman spectroscopy (TERS). However, the majority of FDTD calculations have concerned only the field intensity provided by very high Q systems, but not the radiation properties of plasmonic nano-systems having a rather moderate Q factor. This issue is again similar to the relation between laser amplification and reflectance of the cavity mirrors in readout throughput.

In this presentation, I will discuss efficient configurations of metallic nano-structures for TERS systems, respectively. The proposed metallic systems are composed of more than three particles. The field enhancements of such multi-particle systems have not been discussed before, except for the so called gap mode in particle dimers or bow-tie antennae. I found it is beneficial to have more than three particles in the vicinity of each other for nano-resolution TERS imaging where the molecule directly interacts only with the single particle at the apex.

I will show how the enhancement is increased when nanoparticles are aligned at a proper distance with neighboring ones. For TERS, rather than having an isolated single particle attached at the tip, arranging particles in a way that they surround the particle at the tip apex serves for a drastic increase of Raman scattering efficiency. Similar effects of multi-particle systems have been previously discussed for the nano-rod array aligned linearly with gaps in the context of spectral bandwidth [5]. The present study is an extensional work of this concept of multi-particle system for devising efficient TERS systems. The details will be discussed in the presentation [6].

References

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