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Polarization analysis of near-field probe for tip-enhanced Raman imaging

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1. Introduction

Polarization controlled illumination in Raman spectroscopy is of tremendous advantage as it allows one to study highly directional intrinsic properties of a sample [1]. Tip-enhanced Raman scattering (TERS) microscopy, which realizes Raman imaging at super spatial resolution beyond the diffraction limit owing to the resonant excitation of localized surface plasmon polaritons at the tip apex [2,3], has the potential to achieve such polarization imaging at nanoscale. However, neither evaluation nor control of the polarization properties of near-field light in TERS is as straightforward as in usual far-field illumination, because the polarization of near-field light is influenced by the random shape, size and orientation of the metallic nanostructure attached to the apex of the tip used in TERS.

2. Experimental

Analysis of Near-Field Polarization by Defocused Imaging

In this work, we demonstrate our idea to investigate the ambiguous polarization of the near-field probe from the scattering pattern produced by a metallic tip. Under dipole approximation, we measured the scattering pattern by defocused imaging, where, in order to keep the information about the direction of the dipole oscillation intact, the image of the dipole was formed at a plane away from the focal plane. Figure.1 indicates the experimental setup for defocused imaging and defocused patterns depending on the direction of the dipole oscillation. The direction of the dipole oscillation was determined from the asymmetry in the defocused pattern, and then the polarization of near-field light was evaluated from the oscillation direction by calculating the intensity distribution of near-field light through Green's function.

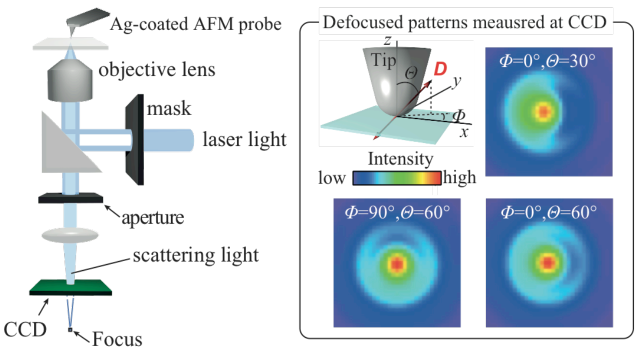

Figure.1 Overview of defocused imaging to investigate the dipole induced at the apex of a TERS tip.

TERS Imaging by Using Tips after Polarization Evaluation

The effectiveness of our method was verified by obtaining TERS images of single-walled carbon nanotubses (SWNTs), where intensity contrast and orientations of SWNTs tell us the state of the excitation polarization. After evaluating the dipole oscillation and hence the polarization of fabricated tips, we used those tips to measure TERS images from SWNTs. Figure.2 shows an example of the TERS images. SWNTs oriented in the direction of the dipole oscillation were strongly excited, and the contrast of the TERS image indeed depended on the oscillation direction of the dipole. Moreover we found the TERS image was in excellent agreement with the calculated TERS image, verifying that the polarization of near-field light was quantitatively estimated by our technique.

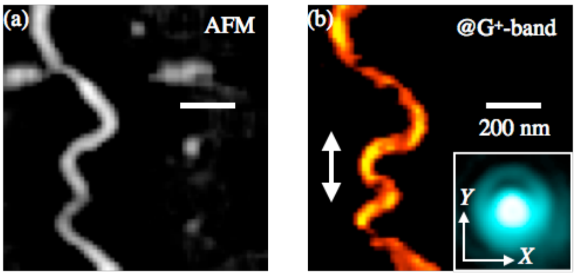

Figure.2 (a) AFM and (b) TERS images of SWNTs. The white arrow in (a) means the in-plane component of the dipole oscillation induced in the tip used in the measurement. Inset shows the defocused image of the dipole indicating the oscillation direction.

3. Conclusions

We have developed a method to successfully analyze the polarization of near-field light in TERS from the scattering pattern produced by the induced dipole in the metallic tip. The success of our technique was confirmed from TERS images, where the contrast agreed rather well with the calculation. Our technique would open the path for nanoscale imaging of some directional intrinsic properties, such as the molecular orientations. It would lead to better quantitative analysis in TERS imaging, giving a better understanding of the behavior of nanomaterials.

References

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