Fabrication of Highly Sensitive TERS Probes using Focused Ion Beam

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Non-destructive and high-resolution structural characterization, including compositional analysis and impurities identification at the submicron and nanometer scale, is highly desired for exploring future functional materials and devices. Tip-enhanced Raman spectroscopy (TERS), which combines atomic force microscopy (AFM) and Raman spectroscopy, can simultaneously gather surface morphology and chemical information, such as bonding configurations and defects presence [1]. Although conventional gap-mode TERS requires the specimen to be placed on a gold substrate, there is a need to develop a sensitive TERS probe that can operate in non-gap mode without substrate selectivity. This would help eliminate sample thickness constraints and enable accurate measurement of two-dimensional materials without substrate effects [2]. In this study, a tip composed of Ag and Au was fabricated, and the tip apex was machined using a focused ion beam (FIB) to enhance amplification by reducing the tip's radius of curvature.

Graphene oxide (GO) monolayer films were prepared on a SiO_2 substrate by spin-coating and measured using an AFM-TERS instrument (AIST-NT + HORIBA XploRa Plus). For the probe preparation, a Si cantilever (OLYMPUS, OMCL-160TN-R3) was heated to form a thin SiO_2 layer, which was then sputter-deposited with silver and immersed in an $HAuCl_4$ solution. The tip apex was then shaved off using a focused ion beam (FIB) as shown in Fig. 1(a). Using this probe, Raman spectra of GO deposited on the SiO_2 substrate were obtained, with tip in-contact (I_{total}) and no tip contact (I_{FF}) as shown in Fig. 1(b). The TERS contrast ($C_{TERS}=(I_{total}-I_{FF})/I_{FF}$) was calculated at 6.67 [3].

Fig. 1(c) show topography and D-peak mapping image of the GO films. We have successfully performed non-gap mode TERS mapping of GO on SiO_2 substrates and found that the enhancement factor varied depending on the geometrical shape of the probe.

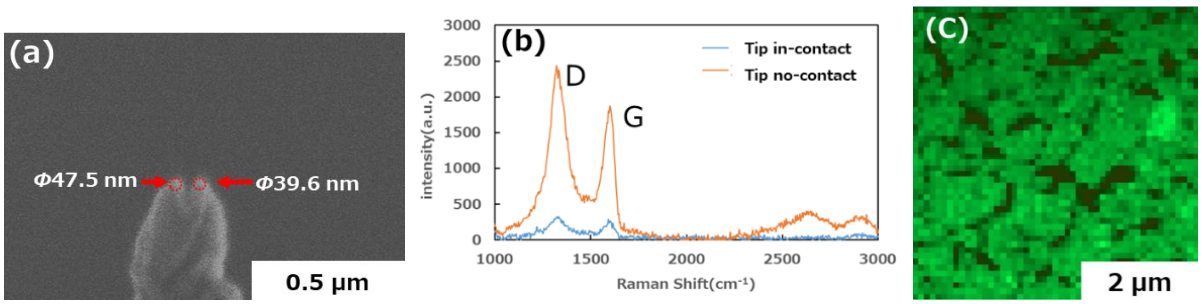

Figure 1. (a) Scanning ion microscope image of TERS probe after FIB fabrication, (b) TERS spectra recorded at monolayer GO on SiO_2, (c) D peak mapping of the GO monolayer film.

References

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