

Title	Tip Design and Fabrication for Tip Enhanced Raman Spectroscopy in Visible Range
Author(s)	Maouli, Imad
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Osaka University

## Abstract of Thesis

Name (Imad Maouli)

Tip Design and Fabrication for Tip Enhanced Raman Spectroscopy in Visible Range
可視先端増強ラマン分光のための先端の設計と作製

## **Abstract of Thesis**

Tip enhanced Raman spectroscopy (TERS) is a near-field spectroscopic technique, which achieves nano-scale spatial resolution far beyond the diffraction limit of light. In order to achieve nano-metric resolution, TERS utilizes metallic nano-tip. However, the fabrication methods of nano-tips are not reproducible in the optical region. Here, I aimed to overcome the TERS tips related problems by optimizing the tip designs using numerical simulation, fabricating the designed tips, measuring experimentally the plasmonic resonance scattering of these tips, and taking TERS images of CNT in order to demonstrate their plasmonic properties. Firstly, I did detailed finite differential time domain (FDTD) calculation in order to optimize nano-tip design and material for high enhancement and high resolution in the optical region. As results, changing the length of the silver layer on SiO<sub>2</sub> tip allowed the control of the plasmon resonance of the tip with in the visible range. The FDTD simulations also confirmed that the resolution depends on the curvature of the tip. I demonstrated a fabrication method using focused ion beam (FIB) milling to realize nano-tips with different lengths of silver layer that is deposited on SiO<sub>2</sub> AFM tips using thermal evaporation system. Applying dark field microscopy, I experimentally measured the plasmonic resonance of the fabricated tips. In addition, I performed TERS imaging of CNT with different excitation wavelengths in order to demonstrate plasmonic properties of the fabricated tips. As a result, The TERS images showed a high enhancement when the excitation wavelength was in near resonance with the fabricated tips. The last part of my thesis was to realize a tip for polarization sensitive TERS measurement. I optimized a tip design using FDTD calculation for polarization study. I fabricated the polarization sensitive tip by using FIB milling, and then thermally evaporate silver on the milled AFM tip. After the fabrication, TERS imaging of CNT were taken using three different polarized lights.

The results presented in this study are beneficial to TERS and its applications. We could fabricate tips with a specific resonance, therefore, the TERS in the visible range is promising with our technique, since we could overcome some of the tip related problems, and obtain reproducible results. I also developed polarization sensitive tip for polarization TERS imaging and analysis.

主 査 教 授 河田 聡 副 査 教 授 井上 康志 副 査 教 授 菅原 康弘 副 査 教 授 川田 善正(静岡大学工学部機械工学科)			(職)	氏 名		
副 查 教 授 菅原 康弘 資文審查担当者		主査	教 授	河田 聡		
i文審査担当者		副査	教 授	井上 康志		
以後10月1日   別本 数 授 川田 姜正 (	論文審査担当者	副査	教 授	菅原 康弘		
时 五		副査	教 授	川田 善正 (静岡)	大学工学部機械工学科)	

## 論文審査の結果の要旨

This dissertation describes the optimization and fabrication of tunable plasmonic tips for tip enhanced Raman spectroscopy (TERS). TERS utilizes sharp metallic tip. Tip confines the light to few nanometers, and enhances the optical signal. The local plasmon resonance (LPR) of this tip depends on its geometry, and material. Usually the tips are obtained by coating a thin metal layer on semiconducting AFM cantilever using thermal evaporation. It is very difficult to control the geometry and the length of the metal on the tip, and hence the control and reproducibility of LPR becomes challenging, which makes TERS not reproducible. Therefore, in this work, novel design tips are proposed. The main achievements of this dissertation are presented in a complete research that includes the simulation, fabrication, and characterization of tunable plasmonic tips and TERS imaging. These achievements are summarized as below.

-Novel designs of tips are proposed for better LPR wavelength tuning, enhancement and reproducibility for tip-enhanced Raman spectroscopy (TERS). The Finite differential time domain (FDTD) calculation is performed to design and categorize these tips. The designs are based on real aperture-less tip used in AFM-TERS. The experiment feasibility of the fabrication is discussed as well.

-The experimental fabrication method is based on focused ion beam (FIB) milling. The tunable plasmonic tips were fabricated with desired metallic lengths. The LPR of the fabricated tips are tuned by changing the length of metal. The LPR of the fabricated tips are measured using dark field microscopy.

-The TERS images of carbon nanotubes are successfully taken by three different fabricated plasmonic tips and three different incident wavelengths in order to selectively enhance the Raman scattering for every desired incident wavelength in TERS.

-To summarize, the dissertation shows simulation and experimental results, which have the same tendency. Moreover, it provides LPR measurements of tunable plasmonic tips and shows the tunable enhancement in TERS imaging. Thus, The simplicity, accuracy and the reproducibility in the dissertation convince the referees. For these reasons, the referees accepted that this dissertation reached enough level to grant a doctoral degree.