



Broadband and Large-Area Field Enhancement for SERS via Hybrid Plasmonic-Dielectric Cavity Coupling

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Abstract

By integrating a TiO_2 cavity layer beneath a hexagonally arranged gold nanoparticle array, we show modulation of LSPR characteristics without requiring sub-nanometer control over nanoparticle morphology. Finite-difference time-domain (FDTD) simulations, supported by experimental results from surface-enhanced Raman spectroscopy, confirm the performance of this design. While the primary LSPR exhibits a modest red shift as the TiO_2 thickness increases from 110 nm to 160 nm, a secondary broadband resonance emerges near 1300 nm. This cavity-induced enhancement spans a broad spectral window ranging from 900 nm to 1500 nm.

Hypothesis

A high-index dielectric spacer between nanoparticles and a gold film forms a hybrid cavity-plasmon mode through Fabry-Pérot cavity-like field distributions.

Introduction

Subwavelength confinement of electromagnetic energy of plasmonic nanostructures amplifies light-matter interaction via localized surface plasmon resonance (LSPR), enabling advances in sensing, photonics, and more. We take inspiration from a Fabry-Perot cavity, where optical waves are confined between two parallel reflectors and pass through under resonance. Varying cavity thicknesses provides scalable resonance tuning and near-field enhancement. Raman enhancement is proportional to 10^4 of the electric field intensity.

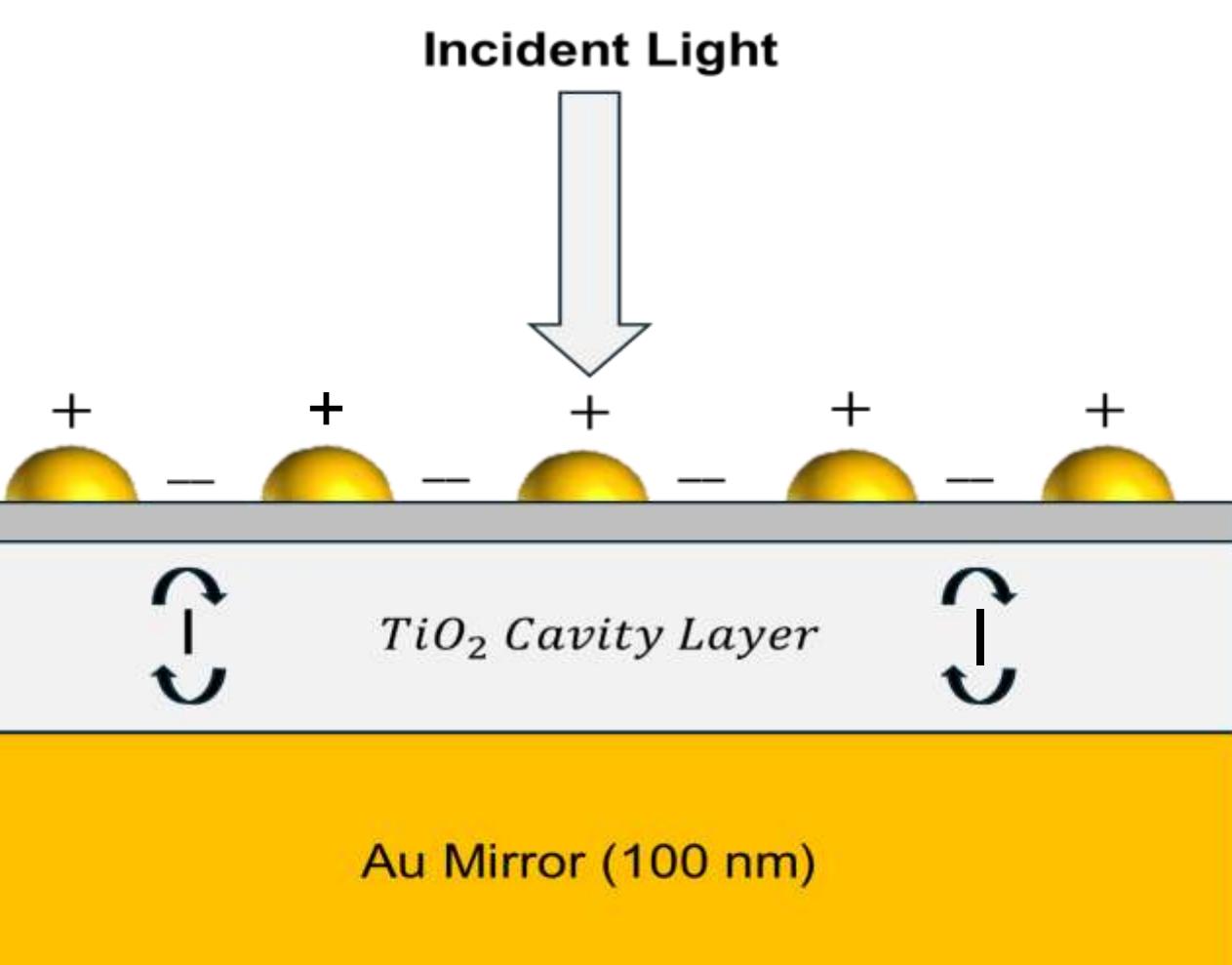
Methodology

FDTD/Surface Enhanced Raman Spectroscopy (SERS):

$$\phi_{prop} = \frac{2\pi n(2d)}{\lambda} = 2\pi \rightarrow d = \frac{\lambda}{2n}$$

- FDTD simulations to field distribution at cavity thicknesses of 110nm, 130nm, and 160nm
- Raman spectroscopy with 785nm excitation wavelength normal incident onto top surface at 1% and 5% laser power
- Range of 0-2500 cm^{-1}

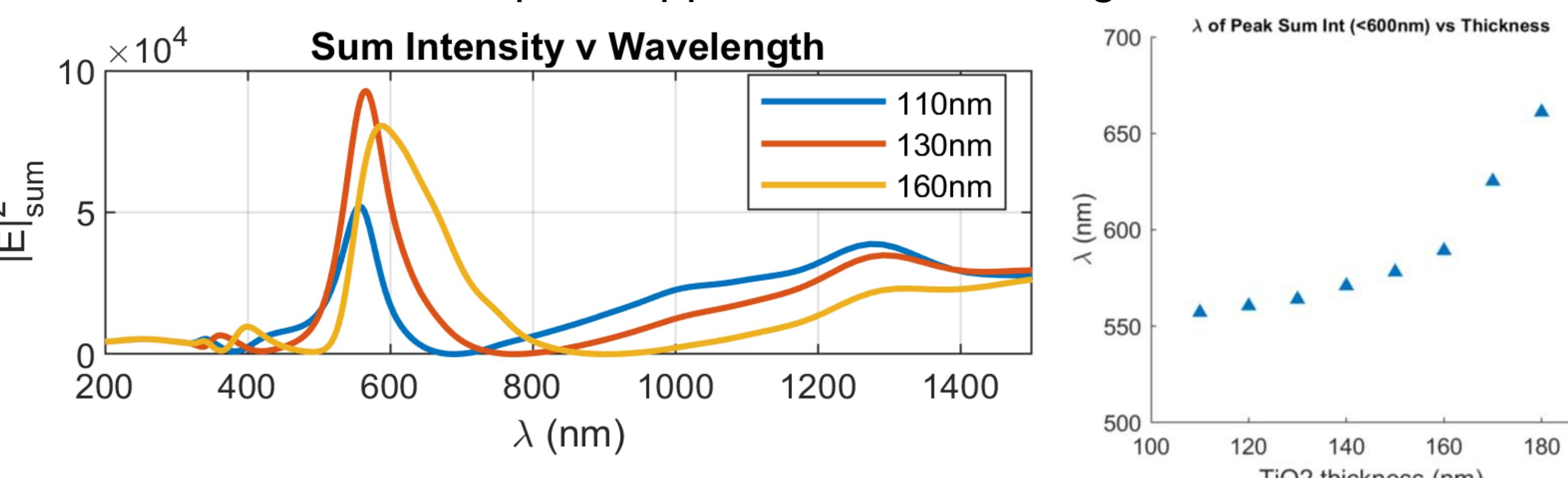
Simulation/Experimental Results



- TiO_2 layer functions as a Fabry-Pérot cavity
- 100nm Au film acts as a mirror while AuNPs partially reflect and transmit light
- Field confinement and constructive interference within cavity layer form hybrid cavity-plasmon mode where near field intensity is greatly amplified and localized at resonance

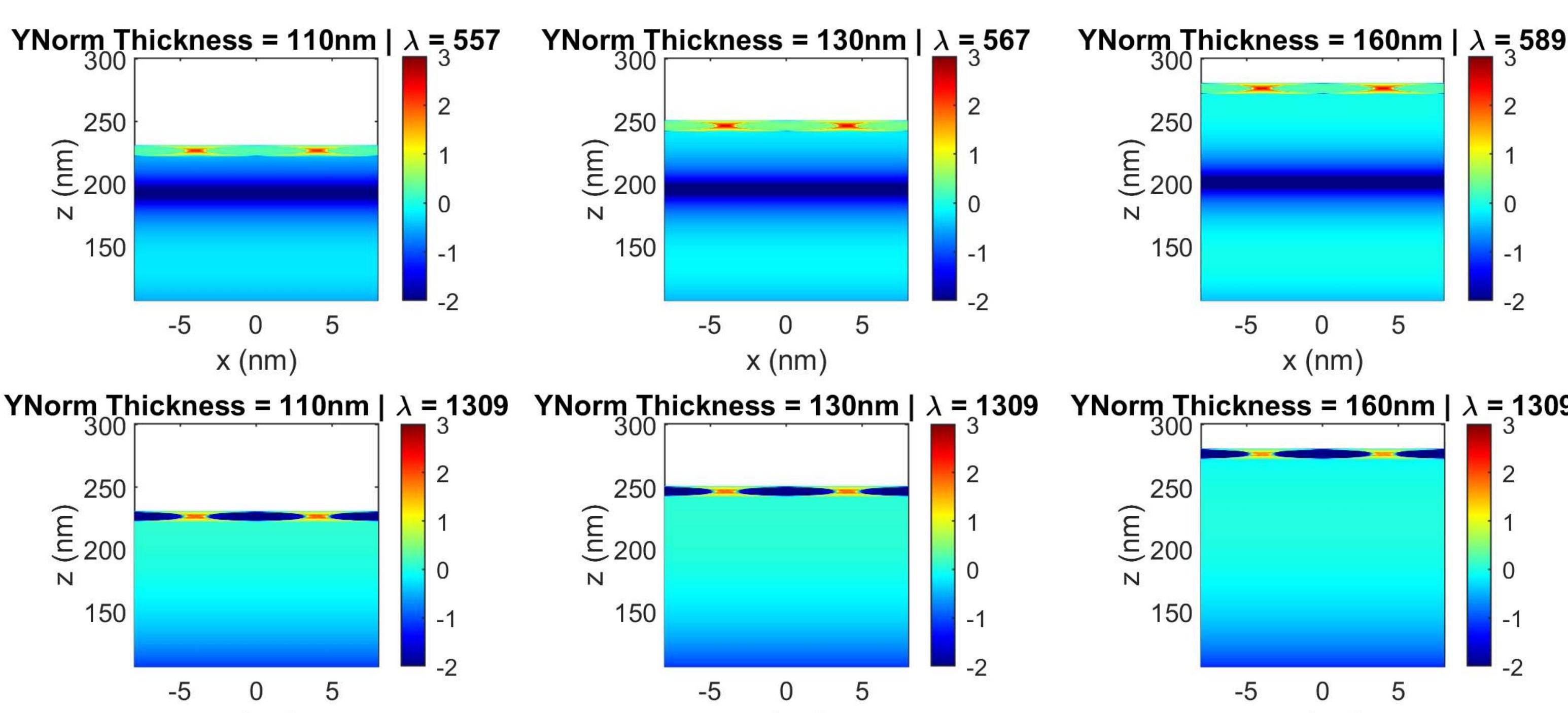
FDTD Sum Intensity Results and Analysis

- Increasing thickness corresponds to red-shifted peak wavelength
- 160nm cavity provides largest electric field intensity at 785nm
- Broadband resonance peak appears in the NIR region

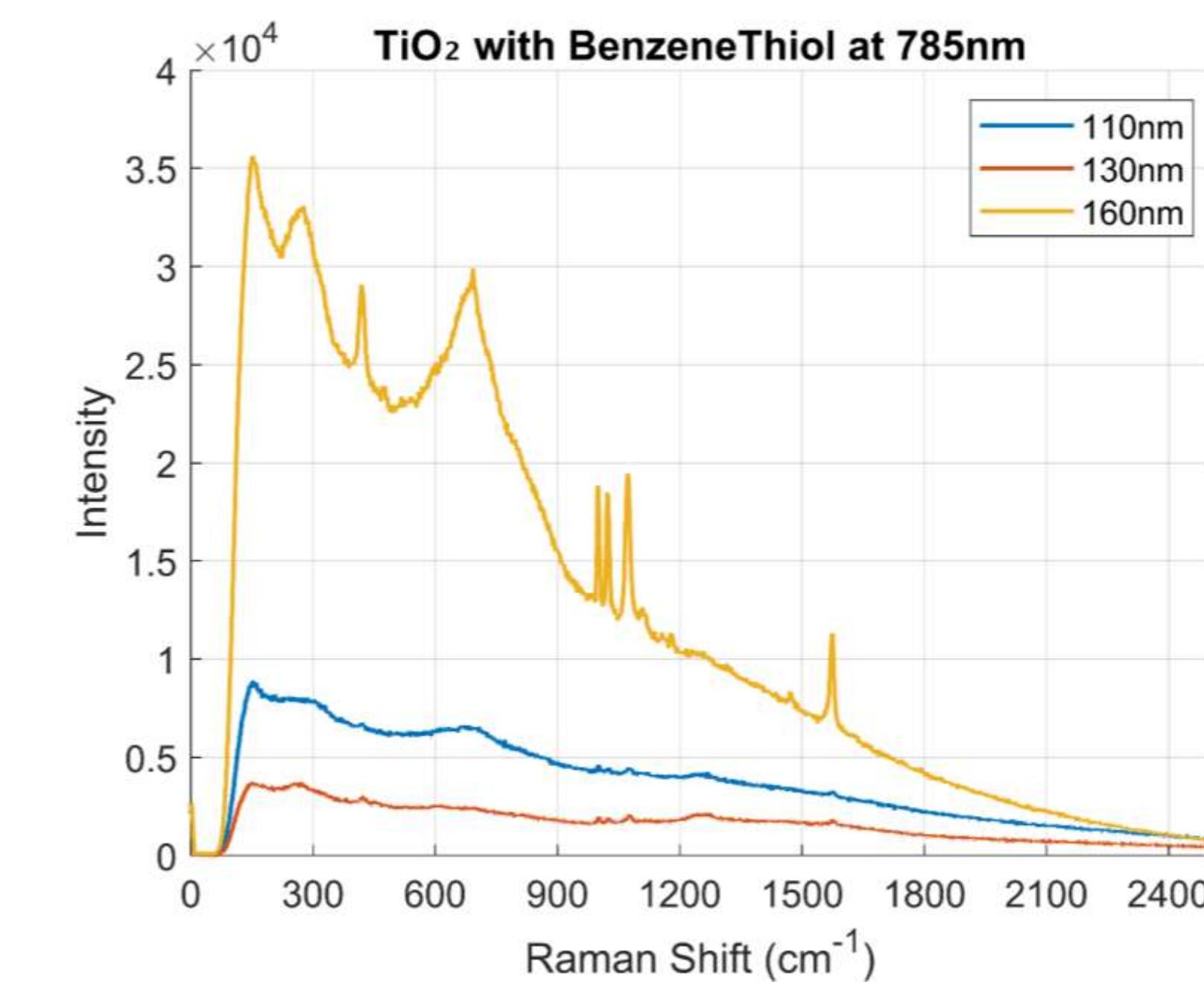


FDTD Field Profile Results and Analysis

- Field profiles demonstrate the tunability of the resonance mode by varying the thickness of the dielectric layer
- 130nm thickness corresponds with the largest field enhancement due to resonance alignment with LSPR from the gold nanoparticle layer
- Second mode resonance at shorter wavelength (~660nm)
- Fundamental mode at the longer wavelength (~1300nm)



SERS Experiment Results



- 160nm cavity shows largest Raman enhancement for 785nm laser line
- Raman peaks of Benzenethiol identified at ~420, 1020-1060, and 1600 cm^{-1}
- Peaks differ in oscillation strength due to different excitation frequencies at 785nm laser line

Conclusions/Future Work

Conclusions:

TiO_2 cavity layer offers resonance tunability and electromagnetic field enhancement. Varied dielectric thicknesses corresponded to different excitation wavelengths and near-field intensities. Raman peaks of Benzenethiol showed considerable Raman enhancement. 160nm cavity thickness offers $\sim 10^4$ Raman enhancement compared to no cavity layer at off-resonance wavelength

Future Work:

Simulate/test thinner TiO_2 thicknesses for resonance tunability. Take Raman measurements at 532nm and 633nm to verify resonance effects at different excitation wavelengths.

Acknowledgments

We are grateful for generous donation from Tom Walker for Raman spectrophotometer. B.L., S.C., and A.L. are thankful to our collaborators, Al-Amin Dhirani et al., at the University of Toronto. A.A. is thankful for National Science Foundation (NSF) for financial support.