Form 3

Abstract of Thesis

Name	(Bikas Ranjan)
Title	Particle-enhanced Raman and Hyper-Raman spectroscopies (ナノ粒子増強ラマン及びハイパーラマン分光)

Abstract of Thesis

Raman and hyper-Raman spectroscopies are widely used to study the intrinsic properties of material. Raman scattering is a weak process and hyper-Raman scattering is further weaker process, therefore, enhancement is required for ultra low detection of sample materials. Recently, plasmonic properties of metallic nanoparticles are widely used to study enhanced spectroscopy. The excitation of surface plasmon of nanoparticles can plasmonically enhance the Raman and hyper-Raman scattered light. The wavelength of surface plasmon resonance (SPR) is determined by the shape, size and dielectric properties of the particle. Plasmonic nanoparticles with large aspect ratio, such as nanorods, show two distinct SPR bands, which originate from the longitudinal and transverse resonances. The wavelength of SPR bands can be tuned by changing the length and the diameter of nanorods. The ability to tune the SPR in such particles has potential for selective enhancement of desired scattering modes. In this dissertation, I discussed the growth of nanoparticles with tunable SPR, and its two such applications - particle-enhanced resonance Raman scattering and particle-enhanced hyper Raman scattering (HRS).

In the first chapter, I introduced the basic theory of Raman and hyper-Raman scattering. Here, classical theory and quantum theory of Raman and hyper-Raman scattering with its differences and signal strength are discussed.

In the second chapter, I discussed about the electromagnetic (EM) field enhancement through plasmonic nanoparticles. Where, EM field confinement and enhancement through metal nanoparticles is reviewed. In this chapter, I also reviewed the plasmonic enhancement of Raman scattering and hyper-Raman scattering.

In the third chapter, optimization of the shape and size of nanoparticles using finite differential time domain (FDTD) calculation is presented. Based on the FDTD simulation, I synthesized silver and gold nanoparticles (nanospheres and nanorice) with tunable plasmon resonance.

In the fourth chapter, a new approach to detect ultra-low concentration of pesticides on food items using particle-enhanced Raman scattering is described. Pesticides are harmful for human health; hence, there is a need of suitable technique for the detection of low-concentration of pesticides on the food. To address the above issue particle-enhanced Raman scattering utilizing nanoparticles with tunable SPR wavelength are used to detect ultra-low concentration of pesticide molecules. In order to detect weak Raman signal, SPR of nanoparticles is controlled in such a way that it is tuned to the laser excitation wavelength which is close to the resonance Raman of the pesticide molecules.

In the fifth chapter, a new approach based on double band plasmonic enhancement HRS is demonstrated. HRS is a very weak nonlinear process, where wavelength of the scattered photons is about half the wavelength of the incident photons. In order to observe double band plasmonic enhancement of HRS, I used silver nanorods, where the length and the diameter of the nanorods are controlled in such a way that the longer SPR wavelength is tuned to the laser excitation wavelength and the shorter one to the scattered photons wavelength. Thus, the selective simultaneous enhancements of both the incident photons and the scattered photons allow us to detect weak HRS signal from crystal violet dye molecules of low concentration.

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論文審査の結果の要旨及び担当者

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論文審査の結果の要旨

This dissertation describes the use of plasmonic nanoparticles for selective enhancement of Raman and hyper-Raman spectroscopies in great details. Raman scattering is a weak process and hyper-Raman scattering is further weaker process, therefore, enhancement is required for the detection of ultra low concentration of the sample materials. This work is important and interesting in nanospectroscopy and nanoscience. Since plasmonic nanoparticles give flexibility to tune surface plasmon resonance (SPR), it could be utilized in the selective enhancement. Main specific achievements discussed in this dissertation are summarized below.

This dissertation discusses the synthesis of plasmonic nanoparticles with tunable plasmonic properties, where gold and silver nanoparticles with different geometries are synthesized and the SPR tunability in the visible to near infrared region of light spectra is successfully demonstrated. The synthesis of plasmonic nanoparticles suggested in this work is efficient and very precise. The strength of SPR tunability is demonstrated for two interesting applications - particle-enhanced resonance Raman scattering and particle-enhanced hyper Raman scattering (HRS).

A new approach to detect ultra-low concentration of pesticides on food items is demonstrated via particles-enhanced Raman spectroscopy. Here, by utilizing the strength of SPR tunability, two different kinds of pesticides are detected at ultra low concentrations by tuning the SPR and excitation laser energy to maximize the resonance Raman effect of the pesticides. This technique has shown the potential to detect different kinds of pesticides present on food items at ultra-low concentration.

At last, double band SPR enhancement of HRS by plasmonic nanoparticles is demonstrated. Here, silver nanorods are used for enhancement. The length and the diameter of nanorods are tuned in such a way that the selective simultaneous enhancements of both the incident photons and the scattered photons allowed detecting weak HRS signal from crystal violet dye molecules. Double band plasmonic enhancement of HRS gives deep insight of sample molecules and it is expected to contribute to a variety of future HRS research work.

The dissertation presents a new aspect of SPR tunablity of nanoparticles and its potential applications in field enhancement. The SPR of nanoparticles has potential to contribute to the better understanding of nanomaterials. Overall, results are well supported both experimentally and analytically, convincing referees of the author’s conclusion. For these reasons, the referees accepted that this dissertation reached the level high enough to grant a doctoral degree.