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論　文　内　容　の　要　旨

氏　名　（　馬越　貴之　）	
論文題名	Background-free near-field optical imaging via plasmon nanofocusing (プラズモンナノフォーカスによるバックグラウンドフリー近接場光学イメージング)

論文内容の要旨

Near-field scanning optical microscopy (NSOM) has been recognized as a powerful optical nano-analytical tool due to its high spatial resolution, far beyond the diffraction limit of light. Since its invention more than two decades ago, NSOM has revealed various features of sample at nanoscale through many kinds of optical analysis such as Rayleigh scattering, Raman scattering, infrared absorption and fluorescence. In NSOM, a metallic tip plays a key role since near-field light is generated at the metallic tip as nano-light-source via plasmon oscillation. Depending on the tip geometry, plasmon behavior at the tip apex remarkably changes, which largely affects NSOM measurements such as sensitivity, spatial resolution, and near-field polarization. In this dissertation, I discuss fabrication of the plasmonic tip to control plasmonic property and implementation of NSOM measurements with the fabricated tips.

In Chapter 1, I begin this dissertation with introduction of the fundamentals of NSOM, where I include importance of the plasmonic tip.

In Chapter 2, I discuss current situation of NSOM. One of the issues is sensitivity because detection volume is nanometrically small in NSOM. In order to obtain high sensitivity, I propose a tip fabrication method to control plasmon resonance. Strongly enhanced near-field light is obtained by resonantly oscillating plasmon at the tip apex. The plasmon resonant wavelength can be controlled by changing length of the plasmonic tip. In my experiment, I utilized photoreduction to control the size of silver nanoparticles at the apex, where the particle size was successfully controlled by changing laser exposure time. I achieved better sensitivity with the tip fabricated by photoreduction in tip-enhanced Raman spectroscopy.

Not only by enhancing signals but also by reducing noise, the high sensitivity can be achieved. Because the near-field light is excited by illuminating the tip apex with incident laser, huge scattering noise by the incident laser is always accompanied with the near-field signal. In Chapter 3, in order to eliminate the scattering noise, I applied plasmon nanofocusing, which allows to induce the near-field light through plasmon propagating far from the tip apex. With this technique, ideally no scattering noise is generated since a plasmon coupler located far from the apex is illuminated with the incident laser. I have designed and fabricated a plasmonic tip for plasmon nanofocusing, and performed NSOM imaging with plasmon nanofocusing on the fabricated tip, where ordinary noise reduction techniques was not required due to the feature of plasmon nanofocusing.

In Chapter 4, taking the advantage of broadband feature of the plasmon nanofocusing, I have developed NSOM to new level of optical nano-analytical technique, which is a main work in this dissertation. Because plasmon nanofocusing is inherently broadband phenomenon compared to plasmon resonance which happens only at a certain resonant wavelength, it is possible to create broadband near-field light. The broadband near-field light can be a powerful probing tool to evaluate sample with various frequency responses at nanoscale spatial resolution. I have investigated a plasmonic tip design to control the plasmonic property for broadband plasmon nanofocusing. I then fabricated a plasmonic tip with optimized structure for broadband plasmon nanofocusing, and induced broadband near-field light covering the visible region, which was confirmed through optical measurements. Further, I conducted NSOM imaging of carbon nanotubes (CNTs) with the broadband near-field light. Due to the deep insight of the broadband optical nano-analysis, I revealed energy band structure and chirality of CNTs at nanoscale. The broadband NSOM imaging is a novel and promising technique that allows to analyze fundamental property of sample with wide frequency range at nanoscale, which will contribute to interdisciplinary research fields from material to biological study.

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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 background-free near-field optical microscopy with plasmon nanofocusing technique. The near-field optical microscopy has been recognized as an attractive analytical tool which enables optical analysis at nanoscale. However, the background signal by incident illumination to a metallic tip has been a major issue to detect weak near-field signal from nanometric volume. Contributions in the dissertation will make near-field optical microscopy more powerful and even bring it up to a new level due to the background-free nature of plasmon nanofocusing. Main works in this dissertation are summarized below.

- This dissertation is dedicated to fabrication of metallic tips that are an essential component in near-field optical microscopy to control properties of near-field light. One of the important works on tip fabrication is tip fabrication for efficient plasmon nanofocusing toward background-free near-field optical microscopy. Through optimization of plasmon coupler structure as well as surface roughness of metallic coating on tip, efficient plasmon nanofocusing on fabricated tips was achieved. The fabrication method suggested in this work is efficient and precise, which enabled almost 100% reproducibility in plasmon nanofocusing. This gives a strong impact in related research fields.

- High-contrast near-field optical imaging was demonstrated with plasmon nanofocusing on fabricated tips. Construction of experimental setup for plasmon-nanofocusing-based near-field optical microscopy and its implementation were also important works to show a potential of this technique as application.

- At last, with unique properties of plasmon nanofocusing, a brand-new analytical method was suggested, which is broadband near-field optical imaging with white near-field light induced by plasmon nanofocusing. There has been no method which achieves broadband analysis with near-field optical microscopy at nanoscale. In the dissertation, broadband near-field optical imaging of carbon nanotubes was successfully demonstrated, where one can find its deep insight that will contribute to a variety of research fields.

Thus, this dissertation has shown great potentials to benefit diverse research fields through its background-free high-contrast imaging as well as broadband nano-analysis. Overall, we referees found consistent consideration and strategies to achieve plasmon-nanofocusing-based near-field measurement and make it practical and reliable as a powerful nano-analytical tool. Therefore, the referees have accepted that this dissertation has remarkable achievements and it certainly deserves a doctoral degree.