



Title	Metasurface-assisted plasmonic tip for plasmon nanofocusing and its application for nanospectroscopy
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Citation	大阪大学, 2023, 博士論文
Version Type	
URL	https://hdl.handle.net/11094/91924
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Abstract of Thesis

Name (YADAV RAVI)	
Title	Metasurface-assisted plasmonic tip for plasmon nanofocusing and its application for nanospectroscopy. (メタサーフェスを有するプラズモニック探針を用いたプラズモンナノフォーカスとナノ分光法への応用)
Abstract of Thesis Near-field scanning optical microscopy (NSOM) is one of the vital techniques in nano-optics because it allows performing nanoimaging of samples using light, far beyond the diffraction limit of light. In conventional NSOM, a nanolight source is created at the apex of a plasmonic tip by exciting the localized surface plasmons at the apex of a plasmonic tip. Although it's a good technique for nanoscale characterization, it has two major limitations. One of the limitations is the generation of huge background signals during nanoscale measurements, and the other is the sample photodegradation in a large area during nanoscale measurement. The root cause of both limitations is because of the direct irradiation on the sample with a diffraction-limited focus spot during nanoscale measurement, which is a must for generating a nanolight source at the apex of a plasmonic tip, and it is impossible to eliminate this illumination in conventional NSOM. In order to eliminate the limitations, recently, plasmon nanofocusing-assisted NSOM emerged as an efficacious technique that allows generating a nanolight source at the apex of a plasmonic tip through the process of plasmon nanofocusing. In the process of plasmon nanofocusing, surface plasmons are excited at the surface of a plasmonic tip far from the tip apex by transferring energy from the photons of propagating light through a plasmonic coupler at first, and later they propagate towards the apex of a plasmonic tip and once they reach there generates a nanolight source through the process of adiabatic compression. As the propagating light and the nanolight source is far from each other, one can eliminate the problems of background signals and the sample's photodegradation during nanoscale measurements using plasmon nanofocusing-assisted NSOM. In plasmon nanofocusing-assisted NSOM, grating-assisted plasmonic tip is commonly used for generating a nanolight at the apex for nano-optical measurements. This tip design uses a grating coupler for exciting plasmons at the surface of a plasmonic tip by transferring energy from photons to plasmons. Although the grating-assisted plasmonic tip generates a nanolight source at the apex, the intensity of the nanolight source is weak. The primary reason for a low-intensity nanolight source is that the plasmons excited at the grating coupler propagate in two opposite directions, which means only half of the transferred energy propagates toward the apex. In addition to the bi-directional plasmons propagation, attenuation of plasmons energy during propagation at the surface also reduces the intensity of the nanolight source. Plasmons' energy attenuation depends mainly on the materials and the geometry of the plasmonic tip. In my research, I have focused on overcoming the limitations of bidirectional propagation by introducing the unidirectional plasmonic coupler. For this purpose, I have proposed the concept of unidirectional plasmons propagation using metasurface at the surface of a plasmonic tip. I have proposed three metasurface-assisted plasmonic tip designs, which are capable of generating nanolight source at the apex through the process of plasmon nanofocusing. I have demonstrated nanoscale Raman and fluorescence measurements using these metasurface-assisted plasmonic tips. To further improve the plasmonic tip's properties, I have investigated the dependence of nanolight source properties on the tip's materials and the tip's geometry.	

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 focuses on an improved development of the existing technique of tip-enhanced Raman spectroscopy (TERS). TERS is a powerful nanoscale analytical technique, which provides both chemical and topographic properties of materials at the nanoscale. It is a plasmonics-based scattering-type near-field scanning optical microscopy (NSOM) that utilizes a sharp metallic nanotip, which is illuminated with incident light to generate a strong and nanometrically confined light field at the apex. This confined and enhanced near-field light at the apex of the nanotip amplifies Raman scattering or photoluminescence (PL) from molecules located directly underneath the apex, providing a true nanoscale resolution to optical microscopy. Owing to the high sensitivity and high spatial resolution, TERS microscopy has been utilized to characterize chemical properties of several types of nanomaterials. However, since the apex of the nanotip is directly illuminated with diffraction-limited focused incident light, there is always a considerable amount of unwanted background scattering from the sample within the focal volume. Researchers have been struggling to eliminate this unwanted background so that a pure near-field signal only from a nanometric volume of the sample can be obtained. Lately, plasmon nanofocusing-assisted NSOM has become a popular technique as it enables one to perform background-free characterization of samples, because in this technique, one needs to illuminate a plasmon coupler fabricated far from the apex rather than illuminating the apex itself. In addition to the background-free characterization, this technique also allows one to drastically suppress sample photodegradation during PL measurement. Plasmon nanofocusing typically utilized a grating structure as a plasmon coupler, the primary role of which is to match the momentum between incident photons and the excited plasmons to facilitate energy transfer. The excited plasmons then propagate towards the apex and generate a nano-sized light source through adiabatic energy compression. In the first part of this dissertation, the candidate optimizes the tip design, including that of the plasmon coupler, and then utilizes plasmon nanofocusing-assisted NSOM to demonstrate a drastic suppression of photodegradation in near-field PL imaging. Though grating-assisted tips serve the purpose of generating nano-sized near-field-light-source, one of the main concerns with grating coupler is that plasmons propagate in two opposite directions after excitations, which significantly affects the performance of the plasmonic tip. In this dissertation, the candidate designed and implemented a metasurface as the plasmon coupler to overcome the issue of bi-directional propagation, and demonstrated how chiral-polarized incident light can control the direction of plasmon propagation in the process of nanofocusing. Chapter 1 of this dissertation discusses the theoretical background of Raman and PL spectroscopies and nanoscale measurement in NSOM. In Chapter 2, the candidate reviews some basics of plasmon nanofocusing and the suppression of photodegradation using plasmon nanofocusing with grating coupler. Metasurfaces are discussed in Chapter 3, where the candidate demonstrates unidirectional plasmon propagation at the surface of a metasurface-assisted plasmonic tip. In Chapter 4, the dissertation explores different designs of metasurface and			

different configurations for better plasmon nanofocusing. Here the candidate shows some experimental results of Raman and PL measured with newly designed and fabricated tips. At the end, in Chapter 5, the candidate discusses some issues related to attenuation of plasmon energy during its propagation on the surface of the plasmonic tip and numerically optimizes various parameters that affect the intensity of near-field light at the tip apex.

Overall, this dissertation shows good potential for a new design of plasmonic nanotip for NSOM-based microscopy, particularly TERS and tip-enhanced PL, where a metasurface is fabricated at the shaft of a metalized nanotip, far from the apex. This metasurface serves as a unidirectional plasmon coupler for background-free near-field measurement and microscopy. It demonstrates near-field measurements of both Raman and PL signals through plasmon nanofocusing by utilizing the designed and fabricated metasurface-assisted tips.

The reviewers of this dissertation found consistent consideration and strategy to demonstrate the strength of metasurface-assisted plasmonic tips for plasmon nanofocusing and its applications in TERS and TERS-based nanoimaging. The reviewers think that this dissertation contributes the progress of applied physics. After thorough considerations and discussions, the reviewers have accepted that this dissertation has reasonable achievements and it can be accepted for a doctoral degree.