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Author(s)	Ren, Fang; Takashima, Hideaki; Tanaka, Yoshito; Fujiwara, Hideki; Sasaki, Keiji
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Two-photon excited fluorescence from a pseudoisocyanine-attached gold-coated tip via a thin tapered fiber under a weak continuous wave excitation

Fang Ren, Hideaki Takashima, Yoshito Tanaka, Hideki Fujiwara, and Keiji Sasaki*

Research Institute for Electronic Science, Hokkaido University, Sapporo, 001-0020 Japan

**sasaki@es.hokudai.ac.jp*

Abstract: A simple tapered fiber based photonic-plasmonic hybrid nanostructure composed of a thin tapered fiber and a pseudoisocyanine (PIC)-attached Au-coated tip was demonstrated. Using this simple hybrid nanostructure, we succeeded in observing two-photon excited fluorescence from the PIC dye molecules under a weak continuous wave excitation condition. From the results of the tip-fiber distance dependence and excitation polarization dependence, we found that using a thin tapered fiber and an Au-coated tip realized efficient coupling of the incident light (~95%) and LSP excitation at the Au-coated tip, suggesting the possibility of efficiently inducing two-photon excited fluorescence from the PIC dye molecules attached on the Au-coated tip. This simple photonic-plasmonic hybrid system is one of the promising tools for single photon sources, highly efficient plasmonic sensors, and integrated nonlinear plasmonic devices.

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OCIS codes: (240.6680) Surface plasmons; (310.6628) Subwavelength structures, nanostructures; (190.4390) Nonlinear optics, integrated optics.

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1. Introduction

In recent years, strong interaction between light and matter at the single-photon level has attracted considerable attention, because it plays a vital role in various applications, such as nanophotonics, quantum computation, and quantum information science. Localized surface plasmons (LSPs) near metal nanostructures have been the subject of such intensive research, because they have the ability to confine light into the nanoscale areas beyond the diffraction limit and to enhance the efficient interaction of light and matter [1–4]. They have been used in various applications, such as optical sensing [5,6], surface enhanced Raman spectroscopy

(SERS) [7,8], enhanced nonlinear fluorescence [9–11], and second harmonic generation [12,13]. However, it is still a great challenge to focus a single propagating photon into a single plasmonic nanostructure efficiently due to the huge scale mismatch among photons, metallic nanostructures and molecules. To realize the excitation of LSPs, so far various methods had been proposed, such as prism coupling [14,15], objective lens (free-space excitation) [16], silicon on insulator waveguides [17], and tapered fibers [18,19]. Among these methods, optical tapered fibers with subwavelength diameter are particularly promising in view of their highly efficient LSPs excitation.

The tapered fibers have been a very powerful tool for various optical studies such as biological sensing, optical power delivery systems, quantum optics and quantum information science, owing to their low loss, single mode guidance and the intense evanescent field outside the optical thin tapered fiber [20–22]. Besides their use as excitation methods of LSPs induced in metallic nanostructures, it also has been demonstrated that fluorescence photons from light nanoemitters, such as atoms, quantum dots, and nanodiamonds can be channeled into the guided modes of the thin tapered fibers [23–25]. Furthermore, trapping atoms around the optical nanofibers using the strong evanescent tail of the guided field has been proposed [26,27] and the spontaneous emission of atoms can be strongly modified around the nanofiber [28].

In this paper, we proposed a simple tapered fiber based photonic-plasmonic hybrid nanostructure that combined the merits of both plasmonic and photonic elements, and succeeded in observing two-photon excited fluorescence (TPF) from the pseudoisocyanine (PIC) dye molecules attached on the Au-coated tip even under a weak continuous wave (CW) excitation condition. Especially we note that Sanchez et al. [29] have also demonstrated that TPF from PIC dye molecules dispersed on a substrate was enhanced by an Au tip, where the incident pulsed laser light was focused on the tip by an objective lens and the emission from PIC dye molecules were collected by the same objective. Although in their paper, the plasmonic enhancement at the tip apex would also play an important role for improving the excitation and emission efficiencies for observing TPF, the coupling of incident light into the Au tip would be low due to the scale mismatch of the focused spot and the apex of the tip, resulting in the use of a high intense pulsed laser excitation. The simple tapered fiber based photonic-plasmonic hybrid nanostructure studied here was composed of an optical thin tapered fiber and a PIC-attached Au-coated tip. The tip-fiber distance dependence and excitation polarization dependence were measured. It was found that by the use of a thin tapered fiber and an Au-coated tip, efficient coupling of the incident light (~95%) and LSP excitation at the Au-coated tip could be realized and this effect makes it possible to induce two-photon excited fluorescence from the PIC dye molecules attached on the Au-coated tip with a weak CW excitation. In addition, because the tapered-fiber collection efficiency of the emission light from the PIC-attached Au-coated tip was estimated to be ~16%, which was higher than the emission collection efficiency of an objective lens with 0.42 NA. We consider that this simple photonic-plasmonic hybrid system is one of the promising tools for single photon sources, highly efficient plasmonic sensors, and integrated nonlinear plasmonic devices.

2. Experimental setup

The experimental setup is shown in Fig. 1. A diode laser (New Focus, TLB-6312, Wavelength ~780 nm) was introduced into a tapered fiber as a probe and excitation light. The pair components of half wave plate (HWP1) and polarization beam splitter (PBS) controlled the power of the incident light coupled into the tapered fiber. Two quarter wave plates (QWP1, 2) and a half wave plate (HWP2) were used to adjust the polarization states of the excitation light parallel to the tip axis in order to efficiently induce TPF from the PIC-attached Au-coated tip. The power of the excitation light was monitored using an optical power meter (Newport, 2935-C) connected to the output of a fused fiber coupler (95:5). In order to control the coupling conditions, the separation distance between the tapered fiber and the PIC-attached Au-coated tip was controlled by piezo manipulators (PI-Polytec, P-

621.1CD, P-621.ZCD). The origin of the distance between the tapered fiber and the tip was determined as the position where the tip was contacted to the surface of the tapered fiber. These components and samples were placed in a plastic box to keep a stable condition [30]. The transmitted light from the end of the tapered fiber was measured using a highly sensitive photodiode (Thorlabs, DET36A) and a digital oscilloscope (Tektronix, TDS5034). The emission light from the PIC-attached Au-coated tip was collected by two ways. One way was that the emission light was collected using a microscopy system set on the top of the sample (free-space collection). The emission light was collected by an objective lens with 0.42 NA, and then passed through a dichroic mirror to separate the excitation and emission light, and was incident into a multimode fiber bundle connected to a spectrometer (JASCO Corporation; iDus; Andor). The other way was that the emission light was collected by the tapered fiber (tapered-fiber collection) and was measured at the end of the tapered fiber by the same spectrometer after using shortpass filters to block the excitation light.

A tapered fiber (diameter: 200 ~1000 nm) was fabricated by heating a fused-silica single-mode optical fiber (Thorlabs, 780HP) with a ceramic heater and at the same time stretching both ends of the fiber [30,31]. We monitored the transmittance in the tapered fibers at wavelength about 780 nm during the fabrication process. The transmittances of the tapered fibers in the experiment were over 0.90.

As a metal nanostructure, a commercial silicon atomic force microscope (AFM) probe tip (Olympus, OMCL-AC160TS-C3), was coated by Au thin film using a helicon sputter (MPS-4000C1/HC1). The inset in Fig. 2 shows the image of the Au-coated tip measured by a field emission-scanning electron microscope (FE-SEM, JSM-6700FT). The curvature diameter of the Au-coated tip with the thickness of about 50 nm was ~80 nm. To confirm the localized surface plasmon resonance property of the Au-coated tip, we measured the scattering spectrum from the apex of the Au-coated tip, when a white light was incident into the tapered fiber and the Au-coated tip was contacted to the surface of the tapered fiber. Figure 2 shows the result, which was obtained from the scattered intensity spectrum at the Au-coated tip collected by the objective lens divided by the transmitted intensity spectrum from the end of the tapered fiber without the Au-coated tip. From the results, because we found that the wavelength of the excitation light (~780 nm) was close to the LSP resonance peak wavelength, we could expect the efficient excitation of LSPs at the tip apex.

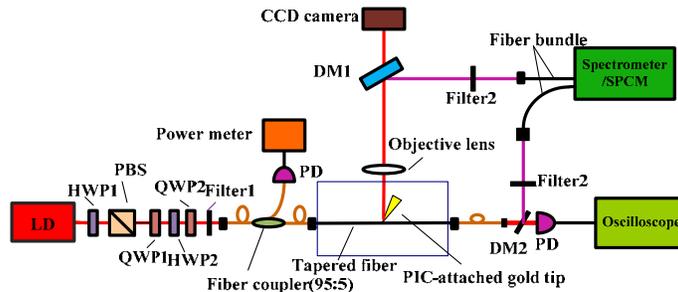


Fig. 1. Schematic of experimental setup. LD: Laser diode; HWP: Half wave plate; PBS: Polarization beam splitter; QWP: Quarter wave plate; PD: Photodiode; DM: Dichroic mirror.

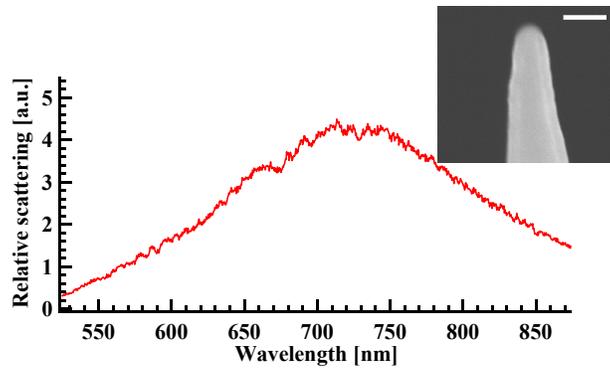


Fig. 2. Localized surface plasmon resonance scattering spectrum of an Au-coated tip; Inset: Scanning electron micrograph of an Au-coated tip by FE-SEM. Scale bar: 100 nm.

For the preparation of the PIC-attached Au-coated tip, PIC dye molecules, which have been well known as dye molecules with a large two-photon absorption cross section at the excitation wavelength ~ 780 nm [32–34], could be attached on the surface of the Au-coated tip using a self-assembly technique [35]. In this method, firstly the Au-coated tip was slowly immersed into 1 mM ethanol solution of 3-mercaptopropionic acid (MPA) at room temperature for approximately 20 min, followed by rinsing it with ethanol solution to remove excess thiol MPA molecules. The MPA was chemically bonded to the surface of the Au-coated tip, because the thiol function groups presented in MPA facilitate the attachment of the Au-coated tip. Then we dipped the Au-coated tip into 0.1 mM aqueous solution of AgNO_3 at room temperature for about 20 min in order to activate the reactive chemical group of the MPA able to bind to PIC dye molecules. After rinsing it by pure water to remove excess AgNO_3 molecules, we immersed the Au-coated tip into 0.01 mM ethanol solution of PIC dye molecules at room temperature for nearly 1 h. PIC dye molecules were attached on the surface of the Au-coated tip after rinsing it using pure water to remove unbound PIC dye molecules.

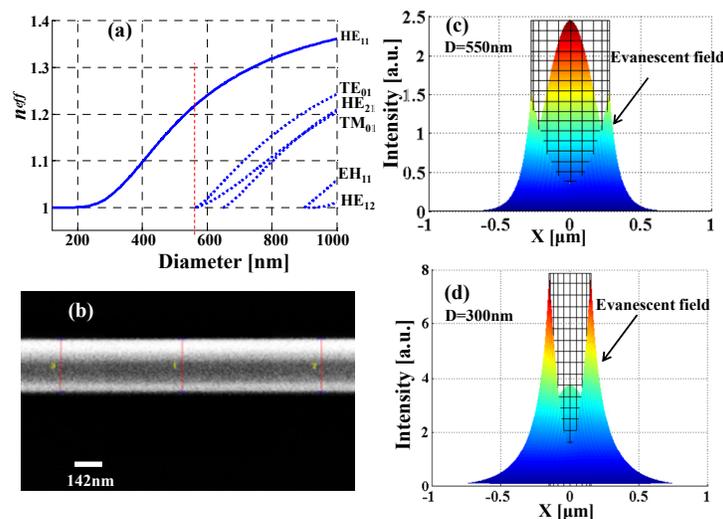


Fig. 3. (a) Numerical diameter dependence of effective refractive index n_{eff} of the guided mode at 780-nm wavelength. Solid line: fundamental mode. Dot lines: higher order modes; (b) Scanning electron micrograph of a tapered fiber by SEM. Diameter (D): 274 nm; Calculated cross-section profile of the total field intensity distribution of the electric field in the

fundamental mode HE_{11} for tapered fibers with diameter of 550 nm (c) and 300 nm (d) at 780-nm wavelength. Cylindrical mesh indicates the tapered fiber as a reference.

3. Results and discussion

First, we numerically analyzed the propagation mode properties of a thin tapered fiber. According to the exact solutions of Maxwell's equations and eigenvalue equations for waveguide modes [20–22], the diameter (D) dependence of effective refractive index of the guided mode (n_{eff}) was obtained numerically, as shown in Fig. 3(a) where refractive index of tapered region (n_1), that of surrounding medium (n_2), and the incident light wavelength (λ) were assumed to be 1.45, 1.0, 780 nm. It was found that, when the diameter of the tapered fiber (D) was reduced to 565 nm (red dashed line in Fig. 3(a)), the tapered fiber only supported the fundamental mode HE_{11} with quasi-linear polarization. Then we numerically calculated the fundamental mode distributions of a thin tapered fiber with the diameters of 300 nm and 550 nm, as shown in Figs. 3(c) and 3(d). From the results, the thick tapered fiber with the diameter of 550 nm confined major power inside the tapered fiber, while a large amount of light as an evanescent field was in the outside of the thin tapered fiber with the 300-nm diameter. These results corresponded to the previous reports [20–22] that an intense evanescent field was generated in the vicinity of a thin tapered fiber, indicating the possibility of the efficient interaction of the propagating photons in the tapered fiber with the Au-coated tip.

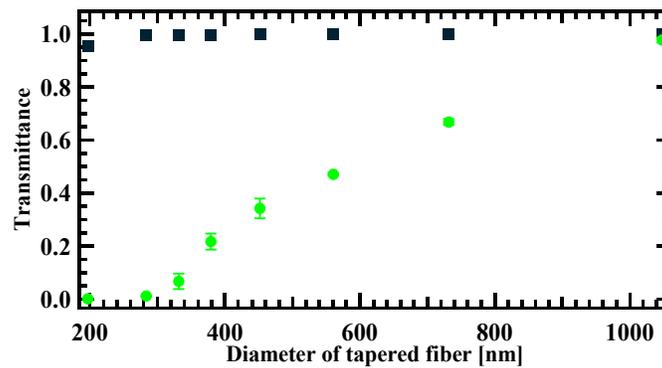


Fig. 4. The tapered-fiber diameter dependence of transmittance from the end of the tapered fiber with the PIC-attached Au-coated tip contacted to the surface of the tapered fibers (circles) and without the PIC-attached Au-coated tip (squares).

To experimentally confirm this, we measured the dependence of transmittance from the end of tapered fiber on the tapered-fiber diameter (Fig. 4). In the absence of the PIC-attached Au-coated tip (squares in Fig. 4), almost no change of the transmittance (T_l) dependent on the tapered-fiber diameter was observed, and T_l was almost equal to 1, when the diameter was changed from 1000 to 200 nm. As the same results were reported in refs [36,37], they suggested that when the diameter of the tapered fiber was larger than $\sim 0.25 \lambda$ (λ : light wavelength), the transmittance of a tapered fiber could keep high transmittance. In the two ends of untapered region, only the fundamental mode HE_{11} of the standard single-mode fiber was existed in the fiber core. When the shape of a fiber was adiabatically changed, the light was guided by the silica-air interface as the hybrid fundamental mode or higher order modes with strong evanescent field around the tapered region. Thus, when the diameter of the tapered fibers was larger than ~ 200 nm in our experiments, hybrid propagation modes of a thin tapered fiber maintained the high transmittance as shown in Fig. 4. However, when the tip was contacted to the surface of the tapered fiber (circles in Fig. 4), the transmittance (T_0) was decreased with decreasing the diameter of the tapered fiber, and T_0 almost reached to 0 as the diameter decreased to 300 nm. We note that the circles with error bars in Fig. 4 present the standard deviation of experimental measurements. These results suggested that the

dissipation ($\eta_c = T_l - T_0$) of the incident light at the tip increased when the tapered-fiber diameter decreased owing to the increase in the amount of the evanescent field. As the tapered-fiber diameter was decreased to ~ 300 nm where the dissipation ($\eta_c \sim 93.0\%$) reached to maximum, the efficient LSP excitation at the Au-coated tip would be expected. Therefore, taking into account these results and the technical reason that thinner fibers were easily broken, the diameter of the tapered fiber was chosen to be ~ 300 nm in the experiments.

Then, we also experimentally analyzed the dependences of transmittance from the end of tapered fiber and emission intensity from the PIC-attached Au-coated tip on the distance (d) between the tapered fiber and the tip. The diameter of the tapered fiber in the experiment was ~ 300 nm. The tip was approached to the surface of the tapered fiber at 20-nm interval step. Figure 5 (circles) represents the distance dependences of the transmittance from the end of the tapered fiber and the emission intensity from the tip through tapered-fiber collection. When the tip was far away from the surface of the tapered fiber, there was no change in the transmittance ($T_2 \sim 1$) and no emission from the tip. As we continued to approach the tip to the tapered-fiber surface, the transmittance decreased, and emission intensity increased. At the distance (d) of 0 nm, the transmittance reached to the minimum ~ 0.05 (T_{min}), and emission intensity reached to the maximum. Thus, at this distance, about 95% of the incident light via the thin tapered fiber was coupled into the tip ($\eta_c = T_2 - T_{min}$), resulting in maximizing the emission intensity from the tip (circles in Fig. 5(b)). Because the dissipation by the tip would depend on the spatial profile of the evanescent field around the tapered fiber, the transmittance has $T(d) \sim 1 - \exp(-\alpha d)$, where α is coefficient of an exponential function. When the transmittance was fitted by the exponential function (red solid line in Fig. 5(a)), the coefficient α was determined to be ~ 0.008 nm⁻¹. Assuming that the intensity of the incident light coupled into the tip followed $I_m \sim \eta_c \sim 1 - T(d) \sim \exp(-\alpha d)$, the TPF emission intensity would be expressed as $I_{em} \sim (I_m)^2 \sim \exp(-2\alpha d)$. By fitting the data (red solid line of Fig. 5(b)), we confirmed that the coefficient of the exponential function was ~ 0.016 nm⁻¹ which was just a twice of $\alpha \sim 0.008$ nm⁻¹ and suggested the quadratic dependence on the incident power. Thus, because the efficient interaction between the tapered fiber and the Au-coated tip would cause the enhancement of two-photon excitation process, the TPF from the PIC-attached Au-coated tip could be observed. Note that in order to confirm the reproducibility, we also measured the data using different Au-coated tips fabricated under the same conditions. When the tip was contacted to the surface, we confirmed that the deviation of the transmittance was evaluated to be ± 0.01 and the emission intensity was 130 ± 20 counts/30s at the incident intensity of ~ 200 μ W.

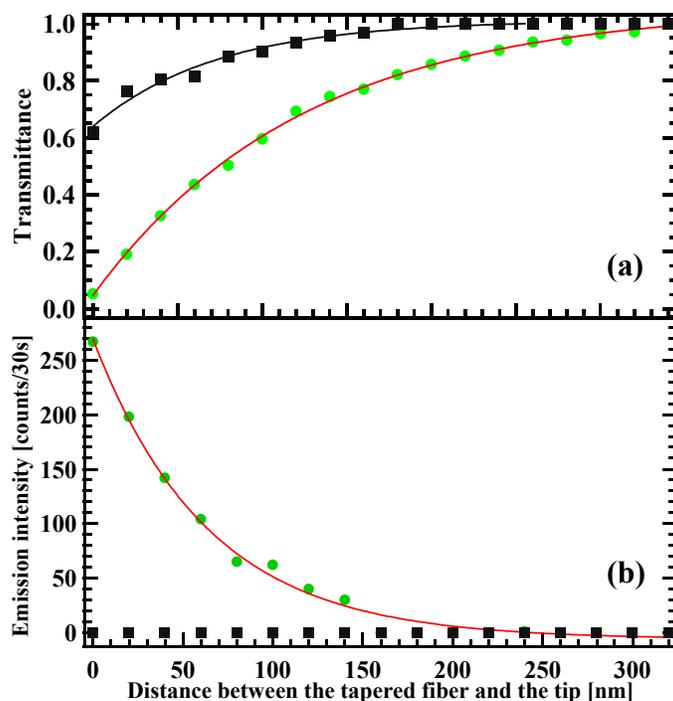


Fig. 5. (a) The transmittance from the end of tapered fiber dependent on various distance between the tapered fiber and the PIC-attached tip with (circles) and without (squares) Au coating. (b) The distance dependence of TPF intensity emitted from the PIC-attached tip with (circles) and without (squares) Au coating. The solid curves are exponential fit to the data.

In order to further prove the importance of the Au coating on the tip, similar distance dependences of the transmittance and TPF intensity from a PIC-attached silicon tip without Au coating were also measured. The silicon tip having curvature diameter of ~ 100 nm, which was almost the same as the apex size of Au-coated tip, was used in the experiment. In order to attach PIC dye molecules on the silicon tip, we used the same method as for the Au-coated tip mentioned in section 2. We compare the photon count rates from PIC-attached Au-coated tip and silicon tip excited by a CW Ar^+ laser (wavelength: ~ 488 nm). From the results, we found that the ratio of the averaged photon count rates was evaluated to be ~ 1.1 , implying almost the same number of PIC dye molecules attached on the Au-coated tip and non-coated tip. When gradually approaching the tip to the surface of a tapered fiber with the diameter of ~ 300 nm, the transmittance (squares in Fig. 5(a)) was decreased to the minimum (~ 0.62). However, although about $\sim 38\%$ of the incident light via the tapered fiber was coupled into the tip without Au coating, no emission light (squares in Fig. 5(b)) from the tip was observed even when the tip was contacted to the tapered-fiber surface. Comparing the results with and without Au coating, we considered that the enhancement caused by LSPs at the Au-coated tip would improve the evanescent coupling of incident light to the tip and also the excitation efficiency of PIC dye molecules. These results suggested that the combination of LSPs of an Au-coated tip and a thin tapered fiber could make it possible to induce nonlinear phenomena even under a weak CW laser excitation.

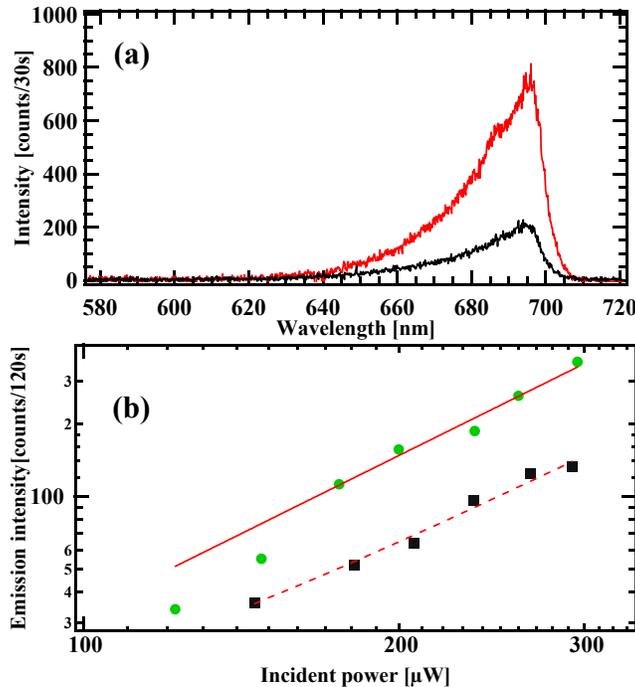


Fig. 6. (a) TPF spectra from the PIC-attached Au-coated tip collected by an objective (black curve) and the tapered fiber (red curve). (b) Log-log plot of dependence of the TPF intensity on the incident power through tapered-fiber collection (circles) and free-space collection (squares). The slope value is ~ 2.15 (solid line) in tapered-fiber collection (~ 2.04 (dashed line) in free-space collection).

On the other hand, in order to further suggest the importance of the thin tapered fiber, we evaluated the collection efficiency of the TPF emitted from the PIC dye molecules attached on the Au-coated tip into the thin tapered fiber (diameter: ~ 300 nm). Figure 6(a) represents the emission spectra measured through the free-space (black line) and tapered-fiber (red line) collections, when the tip was in contact with the surface of the thin tapered fiber. Note that the spectral shapes at the longer wavelength regions (> 700 nm) were strongly modified due to the shortpass filters to eliminate the incident light. In the experiment, we also performed the measurements using an Au-coated tip without PIC dye molecules. However, we could not observe any emission spectrum, and only after the PIC dye molecules were attached on the surface of the Au-coated tip, we could observe the emission. Thus, we concluded that the observed emission spectrum was from PIC dye molecules, not from the Au-coating. According to ref [23], the collection efficiency (η_{col}) into the thin tapered fiber could be express as, $\eta_{col} = 1 / (1 + n_r / n_g)$, where n_g and n_r are the photon emission rates into the tapered fiber (guided modes) and free space (radiation modes). Here, n_r / n_g could be written as $n_r / n_g = (n_{r,obs} / n_{g,obs}) \times (\kappa_g / 2\kappa_r\eta_r)$, where $n_{g,obs}$ and $n_{r,obs}$ are the photons detected by the spectrometer through the thin tapered fiber and free space, as shown in Fig. 6(a), and η_r is the emission collection efficiency of an objective lens. Taking into account the transmission / reflection properties of optical components (lens, filters, DM), the light-transmission factors (κ_g and κ_r) for the paths of guided and radiation modes were estimated to be $\sim 44.6\%$ and $\sim 20.7\%$. Thus, the emission collection efficiency (η_{col}) of TPF from the tip into the tapered fiber was determined to be about 15.7% , which was higher than the emission collection efficiency of the objective lens with 0.42 NA ($\eta_r \sim 4.6\%$), assuming that the emission from PIC dye molecules was homogenously emitted in the whole directions. This result suggests that the tapered fiber provides the roles of not only the efficient coupling of incident light

from the thin tapered fiber into an Au-coated tip but also the efficient collection of nonlinear fluorescence.

We also verified TPF from the PIC dye molecules attached on the Au-coated tip by measuring the excitation intensity dependence of the emission intensity. Figure 6(b) indicates the excitation intensity dependences of the emission intensities measured by the free-space (squares) and tapered-fiber (circles) collections, when the tip was in contact with the surface of the thin tapered fiber. We clearly found a quadratic dependence with the slope values of ~ 2.15 through the tapered-fiber collection and ~ 2.04 through the free-space collection. Thus, these results indicate that the two-photon excited emission from PIC dye molecules attached on the Au-coated tip was observed. From the effective radius of the field distribution at the tapered region in Fig. 3(d), the excitation power density at the tapered region was roughly evaluated to be about 61 kW/cm^2 (incident power at the tapered-fiber input: $\sim 120 \text{ }\mu\text{W}$). According to the distance dependence of the transmittance shown in Fig. 5(a), the difference in the transmittances when the tip was far away and contacted to the surface of tapered fiber suggested about 100% coupling of incident light into the Au-coated tip. Thus, the power excited at the tip apex could be estimated to be the same power of the incident light through the tapered fiber ($\sim 120 \text{ }\mu\text{W}$). Comparing with results of Tanaka et al. [32] where TPF from PIC dye molecules without metal nanostructures was induced by a strong CW excitation light in free space (incident power: $\sim 350 \text{ mW}$), the excitation intensity enhancement of TPF from the PIC-attached Au-coated tip in our simple hybrid system via the thin tapered fiber was estimated to be about three orders of magnitude larger. This enhancement was originated from LSPs of the Au-coated tip and the thin tapered fiber. Therefore, these results suggest the possibility that using this simple system via the thin tapered fiber under a weak CW excitation condition, the highly efficient LSP excitation can be achieved, resulting in the strong light-matter interaction at the Au-coated tip.

Furthermore, to clarify the LSP excitation at the apex of the PIC-attached Au-coated tip, we simultaneously measured the excitation polarization dependences of the transmitted intensity of the incident light from the end of the tapered fiber and the TPF intensity emitted from the PIC dye molecules attached on the Au-coated tip, when the tip was contacted to the surface of the thin tapered fiber. Before starting the measurements, the polarization controller was adjusted to obtain a linearly polarized light in the tapered fiber, in which the polarization state at the tapered region was determined by the use of the same method of Konishi et al. [38]. The initial polarization state of the incident light in the tapered region was confirmed by monitoring the polarization state of the scattered light through Rayleigh scattering process. The dim scattering light of tapered fiber was observed through a polarizing filter parallel to the tip axis using a microscope equipped with a highly sensitive charge-coupled device camera (CCD). When the intensity of the dim scattering points in the tapered region was maximized by adjusting the polarization controller, the polarization of incident light in the tapered region was considered to be parallel to the axis of the Au-coated tip. Then, using the polarization controller, the angle (θ) between the polarization direction of the incident light and the tip axis was controlled, and the polarization of the incident light in the tapered region was first set to be parallel to the axis of the tip ($\theta \sim 0 \text{ deg.}$). Figure 7 shows the excitation polarization dependences of the transmitted intensity and TPF intensity. The transmitted intensity of the incident light and TPF emission intensity was expected to be modified as a function of the angle (θ) between the polarization of the incident light and the tip axis. The transmitted intensity from the end of the tapered fiber would show a $\sin^2(\theta)$ intensity dependence [9,12] because the intensity of the scattering light at the tip depended strongly on the polarization of the incident light owing to the LSP excitation at the tip. As a result, because the scattered light intensity at the tip apex (I_{sca}) would be proportional to $1 - \sin^2(\theta)$, the TPF intensity (I_{em}) would follow a $\cos^4(\theta)$ dependence on the excitation polarization, $I_{em} \sim \cos^4(\theta) \sim (1 - \sin^2(\theta))^2 \sim (I_{sca})^2$. From solid lines in Figs. 7(a) and 7(b), we found that these results are well fitted by the functions of $\sin^2(\theta)$ and $\cos^4(\theta)$, and in good agreement with previous theoretical and experimental results [9,12]. From Fig. 7(b), the TPF intensity (I_{em}) was maximized when the polarization of excited field was set along the tip axis ($\theta = 0$ and

190 deg.), while the TPF intensity decreased several times when the excited field was perpendicular to the tip axis ($\theta = 100$ deg.), which well followed a $\cos^4(\theta)$ dependence. Note that due to the imperfection of linear polarization state of the incident light at the tapered region, the transmitted and the emission light were partially polarized in Figs. 7(a) and 7(b). Thus, we concluded that owing to the LSP enhancement, the two-photon excited process could be induced at the PIC-attached Au-coated tip even under the weak CW excitation.

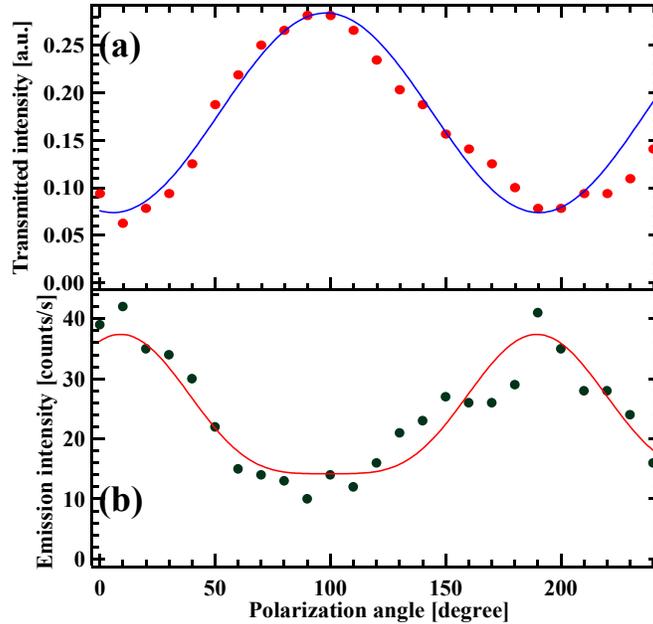


Fig. 7. (a) Transmitted intensity of the incident light from the end of the tapered fiber against the polarization of the incident light. The fitting curve in (a) shows a $\sin^2(\theta)$ function. (b) Incident light polarization dependence of the TPF intensity from the PIC-attached Au-coated tip. The fitting solid curve in (b) shows a $\cos^4(\theta)$ function. The tip was in contact with the surface of the tapered fiber.

4. Conclusions

We proposed a simple tapered fiber based photonic-plasmonic hybrid nanostructure composed of a thin tapered fiber (diameter: ~ 300 nm) and a PIC-attached Au-coated tip, and succeeded in observing two-photon excited fluorescence from the PIC dye molecules attached on the Au-coated tip even under a weak CW excitation condition (several tens kW/cm^2). From the results of the tip-fiber distance dependence and excitation polarization dependence, we found that by the use of a thin tapered fiber and an Au-coated tip, the efficient coupling of the incident light ($\sim 95\%$) and LSP excitation at the Au-coated tip could be realized, and this effect makes it possible to induce two-photon excited fluorescence from the PIC dye molecules attached on the Au-coated tip with a weak CW excitation. In addition, because the tapered-fiber collection efficiency of the emission light from the PIC-attached Au-coated tip was estimated to be $\sim 16\%$, which was higher than the emission collection efficiency of an objective lens with 0.42 NA. We consider that this simple photonic-plasmonic hybrid system is one of the promising tools for single photon sources, highly efficient plasmonic sensors, and integrated nonlinear plasmonic devices.

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