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Microscopic absorption measurement with enhanced sensitivity by using focused surface plasmons

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ABSTRACT

We propose a technique to enhance absorbance of a sample with ultra small volume by using surface plasmons excited in a microscopic region on a flat metal surface. In this technique, the absorbance is measured from the variation of reflected intensity at around the propagating constant of excited surface plasmons in a spatial frequency spectrum. We demonstrate that a wider and deeper dip appears in the spatial frequency spectrum by placing an absorbing droplet sample instead of a non-absorbing sample. An experimental result employing methylene-blue water solution as a sample reveals absorbance enhancement by a factor of 19 from a measurement by attenuated total reflection method.

1. Introduction

Surface plasmons (SPs) are coherent delocalized electron oscillations that exist at the interface between metal and dielectric [1–5]. Kretschmann configuration that consists of a metallic thinfilm supported by a dielectric mediums with different refractive indices is widely used to excite SPs [6–8]. In this configuration, illumination light with an appropriate incident angle and polarization is given from the dielectric with higher refractive index, and SPs are excited at the interface between the metal and the dielectric with lower refractive index [9–11].

It is reported that SP is useful for enhancing the absorbance of a sample on a substrate compared with attenuated total reflection (ATR) method [12,13]. For this enhancement, the thickness of the metallic thinfilm is chosen to have maximum absorption at the excitation angle of the SP under the condition with an absorbing sample. Since the thickness is thinner than the optimal one for a non-absorbing sample, leak of SPs is generated when the non-absorbing sample is placed on the metallic surface. Therefore, reflectance decreases by absorption of the sample. By using this phenomenon, the ratio of reflectance for the absorbing and non-absorbing samples can be remarkably enhanced from ATR method.

In the reported method, the size of the sensing region is determined by the diameter of a laser beam illuminating the metallic thinfilm [14]. Due to the diffraction nature of light, it is difficult to downsize the sensing region to the order of sub-micrometer by miniaturizing the optics, while downsized sensing region is advantageous in the micro-analysis [15]. In order to realize such a small sensing region, we propose to employ focused SP (FSP), which reveals localization in the sub-micrometer region on the metallic surface [16–18].

In this paper, we introduce a method to measure the absorption of a droplet sample by using FSP. We also demonstrate an experiment showing absorbance enhancement by using a droplet of methylene blue (MB) water solution.

2. Sensing principle

Fig. 1 shows an optical setup for exciting FSP. In this setup, collimated light is converged on a metal surface by an oil immersion objective lens with high numerical aperture. The converging light which involves plane wave components satisfying the angular and polarization conditions excites SPs propagating to the geometrical focus. Interference of these SPs results in localization in the diffraction-limited region. In our previous study, we calculated the electric field intensity of the FSP that shows a spot with the radius of 180 nm and evanescent decay in the distance of 170 nm from the metallic surface when radially polarized light with the wavelength of 632.8 nm is focused on the gold

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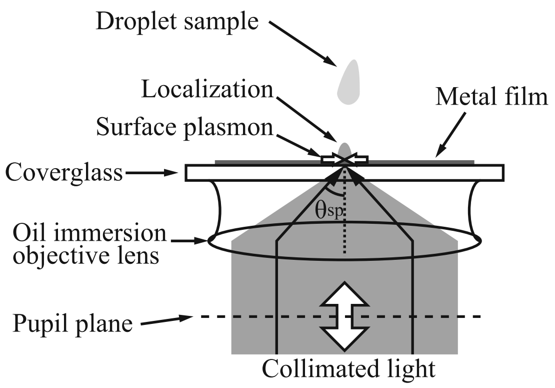

Fig. 1. The optical setup for the excitation of FSP on a flat surface of metal film. θ_{sp} is the excitation angle of SP.

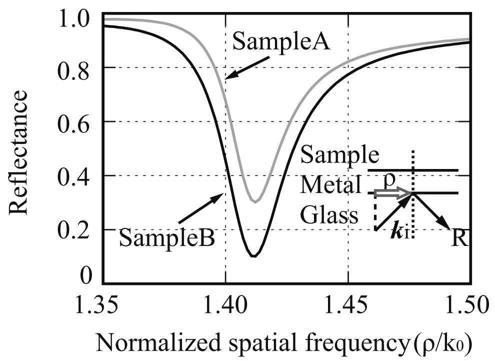

Fig. 2. The calculated reflectance as a function of spatial frequency for sample A (gray line) and B (dark line). The spatial frequency is normalized by k_0. In this calculation, plane wave illumination to the metal film is assumed as shown in the inset diagram. $\boldsymbol{k}_i$ is the wave vector of the plane wave, ρ is the projected component of $\boldsymbol{k}_i$ to the interface between metal and glass. The R is the reflected intensity. The sample A and B are assumed as non-absorbing and absorbing sample, respectively. The minimum of reflectance is caused by the excitation of SPs.

Table 1
Refractive indices at the wavelength of 632.8 nm.

Medium	Refractive index
Glass	1.78
Metal (Ag)	$0.0666 + 4.045i$
Sample A	1.33
Sample B	$1.33 + 0.002i$

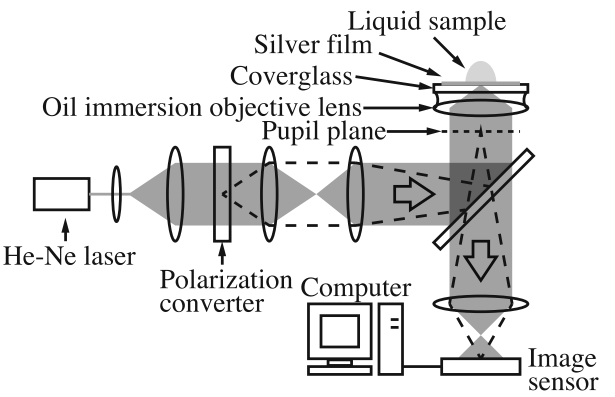

Fig. 3. The optical system of the FSP sensor. SPs are excited and focused in the optical diffraction limit. The developed FSP sensor is able to measure the light absorption of a liquid sample with ultra small volume.

thinfilm covered with water. Therefore, FSP enables the measurement of a sample with the volume of 10 atto litter [19].

In order to explain the principle of the measurement method, we show a simulation of reflected spatial frequency spectrum that is observable at the exit pupil of the objective lens. In this simulation, we assumed a 3 layered structure that consisted of a glass substrate coated with silver and a sample because a droplet sample fully covers FSP on the metal surface. We calculated curves of intensity reflectance against plane wave illumination as a function of the spatial frequency (ρ) given to the interface between the coverglass and the metal film. The transfer matrix method [20] was used for this simulation. We assumed the illumination light with the wavelength of 632.8 nm in vacuum and p polarization. The refractive indices are shown in Table 1 [21,22], and sample A and B represent non-absorbing and absorbing samples, respectively. We also assumed the thickness of the silver film as 40 nm, while the thickness providing maximum light absorption due to SP excitation with sample A is 54.75 nm. Fig. 2 shows the simulated curves. The lateral axis is normalized by the wavenumber in vacuum (k_0). The simulated curves show absorption dips due to SP excitation. One can see that the absorption dip for the sample B is wider and deeper than one for the sample A. The absorbance A_{SP} that can be defined as $-\log_{10} R/R_0$ where R_0 and R represent intensity reflectances with sample A and B, respectively, was obtained as 0.475.

To show the enhancement of the absorbance from ATR method, we calculated reflectance at the critical angle by assuming a 2 layered structure that consisted of the same glass substrate and one of the same samples. We obtained R_0 and R for ATR method as 1.000 and 0.732, respectively, therefore, the absorbance by ATR method A_{ATR} was 0.135. Consequently, SP enhances the absorbance by 3.52 times in this particular simulation. If we optimize the thickness of the metal film, we can obtain larger absorbance and enhancing factor.

3. Absorption measurement by using FSP

Fig. 3 shows the optical system for the experiment. He–Ne laser was used as a light source. The expanded light with linear polarization was converted to radial polarization by a polarization converter. The radially polarized light was focused on the substrate by an oil immersion objective lens with the numerical aperture of 1.65. The substrate consisted of a coverglass with refractive index of 1.78 and a silver film with the thickness of 40 nm. The FSP was excited at around the geometrical focus on the substrate surface. The reflected beam was collected by the same objective lens. The spatial frequency spectrum of the reflected beam, which appeared at the exit pupil plane of the objective lens was recorded by an image sensor located at an imaging plane of the exit pupil. The spatial frequency spectrum was processed by a computer to evaluate absorption of the sample.

Fig. 4 shows the pupil images with (a) pure water and (b) water containing MB with a density of 0.5 g/l, respectively. Fig. 4(c) and (d) are magnified images of the framed areas in Fig. 4(a) and (b), respectively. One can notice that the absorption arc in Fig. 4(b) is wider and darker than Fig. 4(a).

We analyzed detailed characteristics of the reflected spatial frequency by processing the recorded images. Firstly, we determined the center O of the exit pupil image by finding a ring that overlaped with the absorption pattern. In this process, a circular pattern that consisted of 1920 dots was employed as the ring and an average value of pixels in which the dots were involved were used to evaluate the matching. Then, we plotted a series of the averaged values by varying the radius ρ of a concentric ring. The lateral axis of the plot was calibrated by the theoretical value of the propagating constant of SP excited at the interface between silver and pure water. Fig. 5 shows the plot that represents dependency of reflected intensity on spatial frequency