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VISUALIZATION AND ANALYSIS OF A FLOW-RATE DISTRIBUTION IN A LIQUID-LIQUID TWO-PHASE FLUID USING SURFACE PLASMON POLARITONS

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Abstract:

Optical energy propagates along the metal surface as collective oscillations of free electrons when optical waves are irradiated with the resonant condition. The oscillations with electrical fields are called surface plasmon polaritons (SPPs). SPPs are applied to sensors since excitation condition of SPPs sensitively responses to refractive indices. Here, using SPPs, a flow of a liquid-liquid two-phase fluid is visualized, and a flow-rate distribution is derived. A channel is made on the silver-film surface deposited on a slide glass, and filled with ethanol-aqueous solution. In this condition, SPPs are excited on the silver surface by Helium-Neon laser. The laser is expanded and collimated to obtain two dimensional images. Then, water is injected into the channel at a constant pressure. SPPs disappear gradually as the water comes close to near-field of the silver surface, because SPP excitation is optimized to the ethanol-aqueous solution, not to water. The SPP disappearing is visually observed in reflected light. Changes in brightness at the centre of the channel in a recorded movie are converted to changes in distance between the water layer and silver surface, based on simulation of SPP excitation. As a result, we confirmed that the flow speed of the vicinity of the silver surface is slower than that of the distant position. This flow rate distribution corresponds to a theory that a velocity gradient is caused by the shearing force between fluid and wall surface. Presented method doesn't require any tracer particles or coloring though sample fluids are clear and colorless.

Keywords: Surface Plasmon Polariton, Fluid, Flow, Visualization

1. INTRODUCTION

Light is electromagnetic wave. The waves are reflected by metal surfaces since electric fields of the incident waves oscillate the free electrons to generate radiative electromagnetic-fields, generally. On the other hand, the incident waves propagate along the metal surface without radiation, if the waves are coupled with collective oscillations of free electrons propagating along the metal surfaces. The collective oscillations with electromagnetic fields are called Surface Plasmon Polaritons (SPPs). When SPPs are excited by light, the energy of the light is confined and concentrated on the metal surface as SPPs. Thus, intensity of the incident energy is enhanced and optical effects on the surface increase. In addition, the excitation condition depends on a refractive index in the vicinity of the metal surface. These characteristics are applied to sensors [1], Raman spectroscopy [2], photovoltaics [3], holography [4], the laser oscillation [5][6], and so on.

In this paper, using the SPPs, we visualize and analyse how the ethanol-aqueous solution in a channel is pushed out by the water. A silver film is deposited on a channel wall made of glass, and illuminated with an expanded laser-beam through the wall. When the ethanol is in the channel, the incident light is absorbed as SPPs with low reflection. The reflection increases with the water coming close to the silver surface because the water can't satisfy the excitation condition of SPPs. The reflected beam is projected on a screen so that we can visually identify the water incoming to the channel. In addition, the distance from the silver to water layer is estimated by analysing the reflectance. The water layer should come close from centre axis of the channel to walls, because a velocity gradient is caused by the shearing force between fluid and wall surface. This method of visualization and analysis using SPPs doesn't require any tracer particles or colouring though sample fluids are clear and colourless so that the visualization can be carried without any contamination. In addition, it can be applied to micro channels, since it is only necessary to make channels on the silver surface.

2. THEORY AND CALCULATION

2.1 Excitation Condition of SPPs and Visualization of Refractive Index Difference of the Sample Fluids

To excite SPPs, a silver film is illuminated with light waves through the glass as shown in Fig. 1. The silver is deposited about 50 nm thickness so that incident waves penetrate to upper surface of the silver and oscillate free electrons on the surface. Sample medium is above the silver surface. Refractive index of the glass should be larger than that of the sample. SPPs and incident waves resonate mutually and SPPs are excited when wavenumbers k_{spp} and k_{gx} are matched. These wavenumbers are expressed as,

$$k_{\mathrm{spp}} = \frac{\omega}{c}\sqrt{\frac{n_{\mathrm{s}}^2\, n_{\mathrm{m}}(\omega)^2}{n_{\mathrm{s}}^2 + n_{\mathrm{m}}(\omega)^2}}\,, \tag{1}$$

$$k_{\mathrm{gx}} = \frac{\omega}{c}\, n_{\mathrm{g}} \sin\theta\,, \tag{2}$$

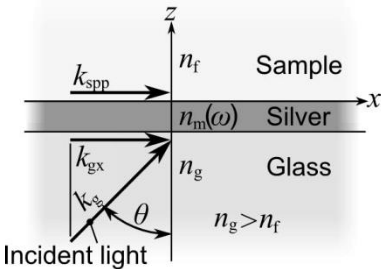


Fig. 1: A cross-sectional diagram of the optical geometry for SPP excitation. Silver is deposited on a glass surface. The silver layer is as thin as the light wave can penetrate into the fluid. n_f, $n_m(\omega)$, and n_g indicate refractive indices of the fluid, silver, and glass substrate, respectively. Note that $n_m(\omega)$ is a function of the optical frequency ω. k_{spp}, k_g and k_{gx} indicate wavenumber of SPPs, incident light, and x-component of the incident light, respectively. θ is incident angle of the light. When k_{spp} meets to k_g, SPPs are excited.

where ω and c are the optical frequency and the speed of light in vacuum, respectively [7]. Thus, excitation condition of SPPs is expressed as,

$$n_g \sin\theta = \sqrt{\frac{n_s^2\, n_m(\omega)^2}{n_s^2 + n_m(\omega)^2}}\,. \qquad (3)$$

This equation indicates that the condition of SPP excitation is affected by the index of the sample, if other parameters are fixed. When the condition is satisfied and SPPs are excited, the incident light is absorbed to silver surface and reflection of the light decreases. On the other hand, if the above condition is not satisfied, the light is reflected. Thus, we can monitor the SPP excitation by means of reflectance. The incident beam is expanded and the reflected beam is projected on screen to observe a flow two-dimensionally and visually.

2.2 Estimation of Distribution of Fluids in Direction of Normal to Metal Surface

When the fluids flow through a channel, the flow-velocity gradient occurs by viscosity of the fluid and shearing force with wall surface of the channel. In this paper, ethanol-aqueous solution is filled in a channel initially and water is injected to push out the ethanol-aqueous solution. Thus, the water comes into the channel in order from the centre to walls of the channel; consequently, the boundary between the ethanol-aqueous solution and water is curved as shown in Fig. 2(a) and the water layer comes close to the silver surface from the centre of the channel.

We simulated the distance from the silver surface to the water layer, using the model of Fig. 2(b) and Fresnel's Equations. The model consists of glass substrate, silver film, ethanol-aqueous solution, and water layers. Before the water injection, the thickness d_{ea}, the distance from the silver to water, is assumed to be infinite, and the incident angle is set to θ_{spe}, at which SPPs are excited most efficiently. In this case, the calculated reflectance I_r/I_0 was very low as shown by dotted line in Fig. 3(c) because SPPs were excited and the incident light was absorbed to silver as the SPPs. After the water injection, the thickness d_{ea} gradually decreases to 0 as the water comes close to the silver surface. The calculated reflectance I_r/I_0 gradually increased to 0.6654 with d_{ea} as shown in Fig. 3(c) because effective value of n_s satisfying a condition of SPP-excitation in Eq. (3) is gradually changed form $n_s = n_{ea}$ to n_w and SPPs disappear gradually. Using this

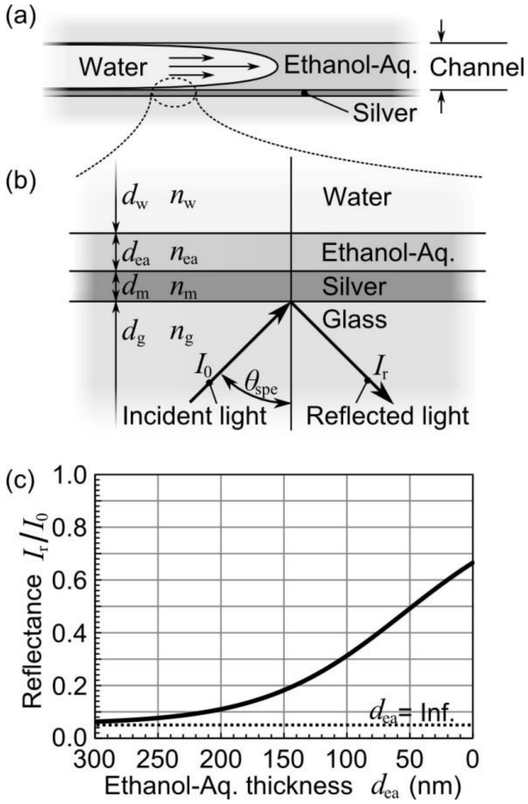


Fig. 2: (a) Cross-sectional diagram of the channel in case that the ethanol-aqueous solution is filled in the channel initially and then water push out the ethanol-aqueous solution. The flow velocity of the centre of the channel is faster than that of near wall because of the shearing force between fluid and wall surface. As a result, the boundary between the ethanol-aqueous solution and water is curved. (b) Calculation model: enlarged view of (a). It consists of glass substrate, silver film, ethanol-aqueous solution (molar fraction of 0.6), and water layers. Refractive indices n_g, n_m, n_{ea}, and n_w are 1.515, 0.124 + 4.059i, 1.355 [8], and 1.333 [9], respectively. Layer thicknesses d_g, d_m, and d_w are infinity, 42.64 nm, and infinity, respectively. Wavelength of the incident light in vacuum λ_0 (=$2\pi c / \omega$) is 632.8 nm. θ_{spe} is 71.47°, at which SPPs excite most efficiently when d_{ea} = infinity and the water layer doesn't exist. I_0 and I_r are intensity of incident and reflected light, respectively. (c) Calculated reflectance I_r/I_0, which depends on d_{ea}. Low reflectance implies SPP excitation. When d_{ea} = infinity, I_r/I_0 = 0.05309. When d_{ea} = 0, I_r/I_0 = 0.6654.

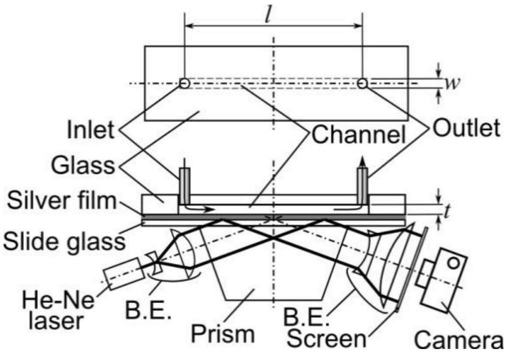


Fig. 3: Channel with optical setup for SPP excitation and observation. The channel is a grooved glass-plate. The inlet and outlet are made of Al pipes. Straight-section length l is 50 mm. Both of cross-sectional dimensions w and t are 3 mm. Silver film is deposited on the slide-glass surface, on which the channel is attached. The silver is illuminated with He-Ne laser (λ_0 = 632.8 nm) through the slide glass and the prism. The laser beam is expanded by the beam expander (B.E.). A gap between them is filled by immersion oil. Reflected beam is expanded again, projected to the screen, and recorded by the camera.

result, the thickness d_{ea} can be estimated from I_r/I_0, which can be obtained by the following experiment.

Note that the parameters of the silver film n_m and d_m, and θ_{spe} used in this calculation are estimated by experiment, in which SPPs are excited actually. Of course, the silver film is used in the following experiment.

3. EXPERIMENTAL SETUP

Figure 3 is a channel and an optical setup for SPP excitation and observation. The channel is a grooved glass-plate. The silver film is deposited on the slide glass, on which the channel is attached. He-Ne laser is expanded by beam expander to observe the channel areally. The silver film is irradiated with the expanded beam through the prism and slide glass. The incident conditions are same as the calculation described above. Reflected beam is expanded again and projected on the transparent screen. The projected image indicates a distribution of SPP-excitation efficiency on the silver surface in the channel. The image is captured by a digital camera and analysed on a personal computer. The recorded movie is 260 × 200 pixels and 0.68 fps.

Figure 4 is the flow path around the channel of Fig. 3. The ethanol-aqueous solution, the initial fluid, is injected by the syringe through the 3-way valve in advance. The water to push out the ethanol-aqueous solution is reserved in the tank. The tank is located at $h \approx 0.5$ m high from the end of the drain tube so that the water flows into the channel with a constant pressure by the potential energy. After connecting the channel and tank by the 3-way valve, and opening the stop valve, the water begins to flow into the channel. Flow rate is adjusted to 5.0 ml/min by the needle valve and flow sensor in advance. The Reynolds' numbers of the flows of the ethanol and water in the channel are 20 and 31, respectively, which are much smaller than the critical Reynolds' number (≈ 2200) [9][10].

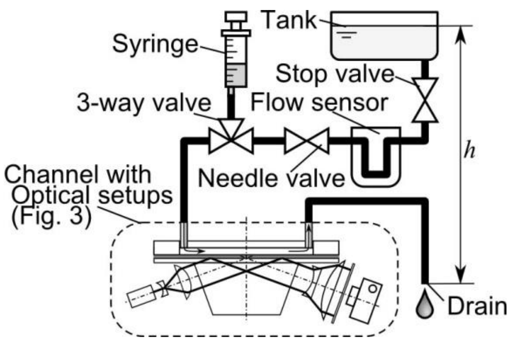
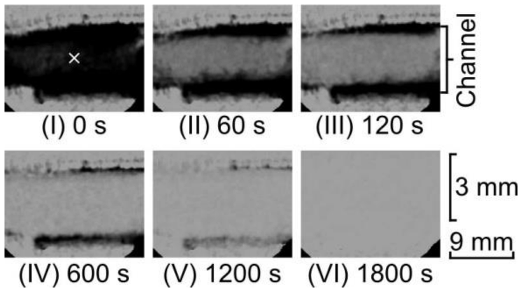


Fig. 4: Flow path around the channel. The syringe is filled with the ethanol-aqueous solution, which is injected to the channel initially. The tank is located at height $h \approx 0.5$ m. Flow sensor and the needle valve are used for control a flow rate. After connecting the channel and the tank by the 3-way valve, we open the stop valve to start experiment.



(I) 0 s (II) 60 s (III) 120 s

(IV) 600 s (V) 1200 s (VI) 1800 s

Fig. 5: Several still images of recorded by the camera. The images are processed 256-colour images. Maximal brightness 255 and minimum brightness 0 of the image pixel correspond to maximal reflectance 1 and minimum reflectance 0, respectively. Namely, whiteness corresponds to the reflectance. The reflectance increasing implies SPP extinction decreasing, which means that the water layer comes closer to silver surface. The scales in right of the images correspond to the dimensions in the channel. It was calculated from the projection angle θ_{spe}. 0 s is the time, at which the brightness began to increase. The water was injected from left to right of the channel.

4. EXPERIMENTAL RESULT

4.1 Visualization of the water incoming

The values of the pixel brightness in the recorded movie are processed as maximal brightness 255 corresponds to maximal reflectance 1 and minimum brightness 0 corresponds to minimum reflectance 0. Figure 5 is several still-images of them. The time, at which the brightness began to increase, was set to 0 s. At first, inside of the channel is totally dark. In the succeeding images, the reflectance increases from around central-axis of the channel, and it broaden toward the channel walls. This implies that the injected water layer approached the silver surface in order from the centre to the walls of the channel, and disturbed the condition of Eq. (3) in the same order. Consequently, the reflectance at the disturbed region increased and the resultant brightness increasing of the images were obtained. Meanwhile, it was difficult to obtain distinctly a flow direction, which was from the left to right of the channel in the experiment. The direction may be obtained, if the flow-path length between the 3-way valve and the channel is shortened so that the water can approach the silver surface before the boundary grows to a longitudinal direction of the channel. If the flow velocity is sped up to make the turbulent flow, the direction may be visualized as movements of the boundary between the water and ethanol.

4.2 Estimation of distance between the water and the channel wall

The black lines in Fig. 6 indicate a relationship between the elapsed time and the reflectance at the point x-marked in image of Fig. 5(I). The grey lines are the thickness of the ethanol-aqueous solution d_{ea}, in the other words the distance between the water and channel wall, derived from the reflectance I_r/I_0 and Fig. 2(c). The distance d_{ea} gradually decreased with the time. This means that the water layer came close to the channel wall with the time. The inclination also decreased with the time. This is occurred by that the water pushed the ethanol of the channel centre more rapidly than that of the near wall-surface because the flow velocity on the centre of the channel is faster than that of the walls.

5. CONCLUSION

In this paper, we visualized the water flowing into the channel filled by the ethanol-aqueous solution in the vicinity of the channel wall surface, using SPPs. The water layer coming close to silver surface affected SPP-excitation efficiency, which was visualized areally. Analysing the visualized images, the distance between the water layer and the silver surface in the direction of normal to silver surface was estimated by calculation. All of results indicated that the flow-velocity increases with the distance to the wall surface of the channel. These results qualitatively correspond to the theory that the viscosity and shear stress makes the velocity gradient. Simulations taking account of the diffusion of fluids is required to give the quantitative measurements in the future.

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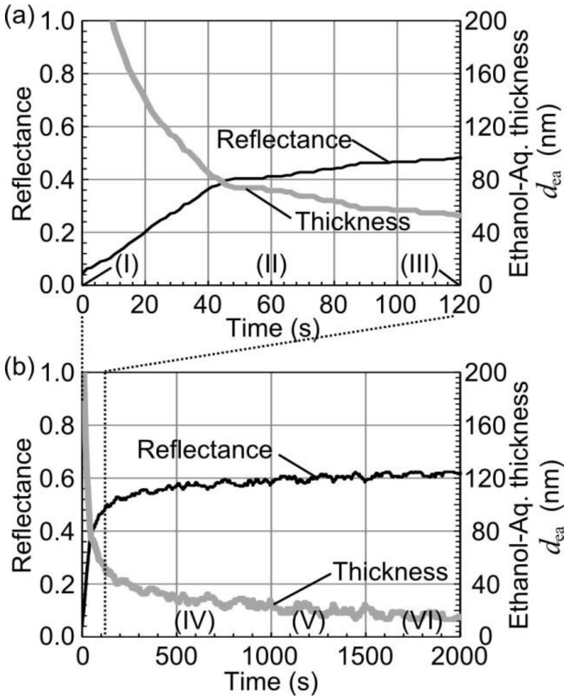

Fig. 6: (a) Reflectance and calculated thickness of the ethanol-aqueous solution layer at x-marked position in the images of Fig. 5. The time is 0 - 120 s. The thickness is derived from the reflectance, using calculation in Fig. 2. (b) Same as above, except the time (0 - 2000 s). The roman numbers (I) to (VI) correspond to the images of Fig. 5.

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