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**Author(s):**

Pavlov, I.N.; Rinkevichyus, B.S.; Vedyashkina, A.V.

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## POSSIBLE WAYS TO INCREASE SENSITIVITY OF THE SURFACE PLASMON RESONANCE IMAGING METHOD

I.N. Pavlov<sup>c</sup>, B.S. Rinkevichyus, A.V. Vedyashkina

V.A. Fabrikant Physics Department, National Research University „MPEI“, Moscow, Russia

<sup>c</sup>Corresponding author: Tel.: +79267520058; Email: inpavlov@bk.ru

### KEYWORDS:

**Main subjects:** heat and mass transfer, flow visualization

**Fluid:** distilled water, salt solution, glycerol, isopropyl alcohol

**Visualization method(s):** Surface Plasmon Resonance

**Other keywords:** increasing sensitivity, evaporation, crystallization, mixing, cooling

**ABSTRACT:** *The paper shows the results of application of the surface plasmon resonance imaging method for investigation of physical processes in liquid droplets. It is substantiated that there is a need to increase sensitivity of a created experimental setup. Possible ways to increase sensitivity to the change in refractive index of an investigated liquid are discussed on the basis of literature review. Developed solution is given in the conclusion.*

### 1 Introduction

Surface plasmon resonance imaging (SPRI) is a known method for flow investigations [1-3]. The development of commercial SPR instruments has made this method available to wide scientific audience and the number of publications in which the use of SPR is described is rapidly increased. SPR is in use for many purposes from food quality control to the study of nanoparticles [4]. Due to ease of use and numerous advantages (such as label-free imaging, ability of nondestructive and in-vivo testing, possibility of investigating fast processes, multipurpose use and many other) SPRI has become widespread in many areas of science. A common implementation of SPRI is based on registration of distribution of reflected light beam intensity from a glass-air interface coated with thin (~50 nm) gold film. The main limitation of this technique is its lower sensitivity to change of refractive index in comparison with conventional angular or spectroscopic SPR systems [5]. The aim of this work is to find possible ways to increase sensitivity of SPRI method in order to meet the growing requirements for the parameters of diagnostic systems.

### 2 Method and Experimental Setup

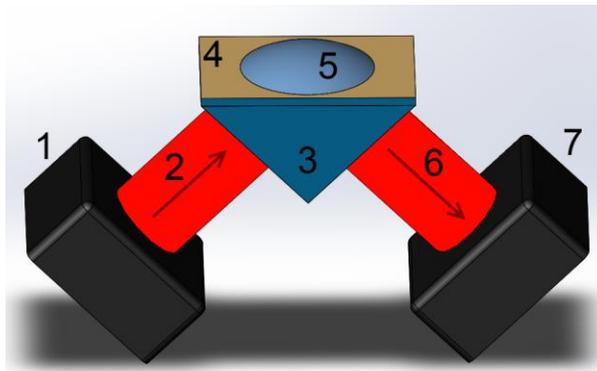
#### 2.1 Visualization Method

It is based on the surface plasmon resonance phenomena when there is no reflected light from an inner basement of a glass prism coated with a thin gold film. In this case it is generally accepted that all energy of incident light is expended on generation of surface plasmons (fluctuations of density of free electrons at the surface of metal film), i.e. efficiency of generation tends to 100%. These resonance conditions depend on polarization, wavelength and angle of incidence of exciting light, thickness of metal film and ratio of refractive indices of a glass, a metal film and an investigated medium (usually it is liquid). In case of alteration of one of these parameters (for example, change in refractive index of medium due to change in temperature, concentration or phase distribution) from resonance values the

resonance is frustrated and the reflected light can be seen on a screen. By magnitude of intensity of this light it is possible to estimate value of deviation of the altered parameter.

## 2.2 Description of the Experimental Setup

To implement this method an experimental setup was created which described in [6]. The scheme of setup is shown in fig. 1. In the setup the next modification of the method was realized. A wide collimated laser beam with wavelength of 650 nm and power of 3 mW was used as an incident light. This parallel beam with radius of 2 cm illuminated the inner basement of a BK7 glass prism covered by a thin BK7 glass plate with 50 nm gold film deposited by sputtering. Reflected light was directed on a camera lens. At the resonance conditions it was absent. And in some physical processes (heating, cooling, crystallization, mixing) occurring in a thin boundary layer of investigated liquid these conditions were violated and intensity of reflected light in corresponding places of a contact spot was registered.



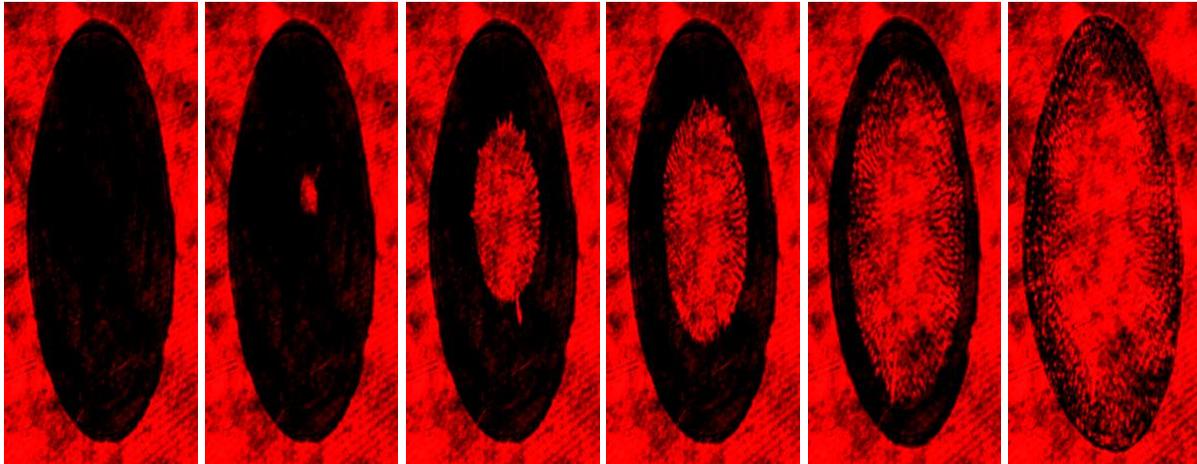
**Fig. 1. The scheme of experimental setup: 1 – optical system for formation of probing wide collimated laser beam; 2 – incident beam; 3 – glass prism; 4 – glass plate with 50-nm gold coating; 5 – liquid droplet; 6 – reflected beam; 7 – system for registration of images**

Sensitivity to change of refractive index in the setup is determined by the slope of the curve of dependence of reflectance on refractive index, sensitivity of a camera matrix, an algorithm of processing of obtained images and some other factors. In this work it is considered how to increase the sensitivity of the SPRI method. It is necessary for expanding the scope of the method and to enable the study of more subtle effects and phenomena.

## 3 Results

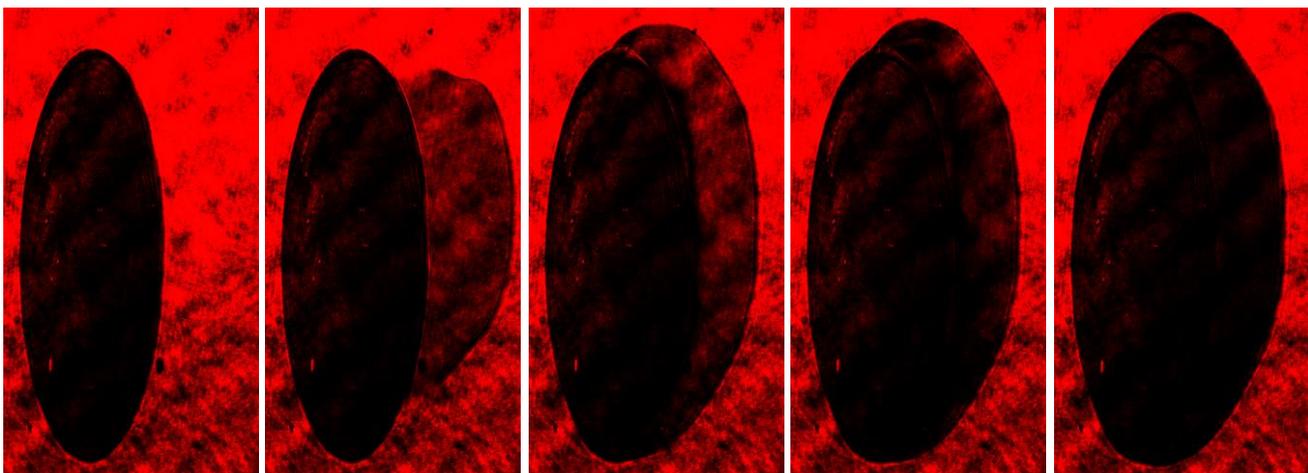
The described above experimental setup was applied for investigation of such processes in liquid droplets as evaporating, cooling, crystallization, spreading, mixing and formation of dryout patterns. Evaporation of a distilled water and isopropyl alcohol occurred in the surrounding space at room temperature (22-24 °C) and humidity 60-70%. In experiments on visualization of freezing of a drop (distilled water mainly was used in it) cooling was carried out with help of a Peltier element, the cooling surface of which touched to the upper edge of the drop. In this case, instead of usual drop shape in the form of a spherical segment, a meniscus was formed (examples of such images are shown in fig. 2). But this circumstance had little effect on the images obtained, since the processes in the lower layer of a droplet were visualized. Fig. 2 shows a series of experimental images on visualization of the crystallization process of a distilled water droplet of volume 15  $\mu\text{l}$ . At initial moment of time the

droplet is liquid and its refractive index is so that the minimum of intensity of reflected light is observed due to surface plasmon resonance. Then because of external cooling with help of a Peltier element the droplet becomes crystallized and the small bright spot appears on the image corresponding to a formed piece of ice. Then this piece grows and finally occupies the whole image of the droplet. Gray areas of the image correspond to reflection from a film-ice interface and bright areas correspond to reflection from an interface of a gold film and formed gas bubbles. It was established that the structure of a near-wall layer of droplet (quantity and sizes of bubbles) is determined by a gap between the cooling surface and the glass plate with gold film.



**Fig. 2. A series of images showing the process of crystallization of a distilled water droplet**

Also a number of experiments on visualization of the process of mixing two liquid droplets were performed. In the experiments such liquids were used as distilled water, water for injections, tap water, aqueous solutions of salt, glycerol and isopropyl alcohol. Fig. 3 shows a series of experimental images on visualization of the process of mixing fresh and salt water droplets ( $n_c$  equals 1.3320 and 1.3450 correspondingly).



**Fig. 3. A series of images showing the process of mixing two droplets: fresh (left) and salt (right) water**

From test experiments it became clear that sensitivity of the created setup to the change of refractive index in an investigated liquid is about 0.0001 and should be improved.

## 4 Possible Ways to Increase Sensitivity

### 4.1 Improvement of the Experimental Setup

Consider some possible ways to increase sensitivity of described surface plasmon resonance imaging method. Among them, first of all, it is necessary to allocate evident corrective measures to address deficiencies in the experimental setup. They include a beam quality improvement, use of different light sources (for example, a diode laser instead of a superluminescent diode [7]), use of pinhole as a spatial filter or even special intensity equalizers over beam cross section (so called  $\pi$ -shapers [8]), use of better quality polarizers, reduction of roughness of glass surfaces, choice of material for a glass plate coating, getting rid of speckles, use of a digital camera with higher bit depth of image, decreasing signal-to-noise ratio by all available means, choice of wavelength range for each problem (in [9] it is shown that use of infrared range of spectrum allows to increase sensitivity for CO<sub>2</sub> detection).

### 4.2 Improvement of an Algorithm of Image Processing

Second evident way to increase sensitivity, i.e. ability to distinguish as little as possible a change in the refractive index, is to use another processing algorithm, including image filtration, averaging, increase an image contrast in controlled ways, for example, such as used in motion microscope [10]. A new method of magnifying subtle changes normally invisible to the eye lies at its core. Authors show that amplification level of up to 50 can be achieved.

### 4.3 Method of Polarization Contrast

In recent years, a polarization contrast method for high-throughput SPR sensing has been developed [11]. It converts changes of light polarization (phase and amplitude) into changes of light intensity that is measured directly and thus it gains the same level of sensitivity as the phase modulation method. A remarkable way to implement this method is shown in [5]. Authors suggest to use not bare prism or thin gold film, but thick film as the extinction area. Compared with other direct and indirect contrast methods, this method can provide higher contrast sensor images and inert background to shield the reference channel from the influence of the sample. It has good adaptability for sensor parameter errors and thus regulating the polarization system for background extinction is easy to be realized in practice.

### 4.4 Phase-Jump Detection

Another way to improve sensitivity of SPRI is to use phase-jump detection [12]. It was noted by authors of this work that if a system amplitude parameter crosses zero, then the system total phase shows the Heaviside  $\pi$  jump as well as markedly different behavior for infinitesimally close values of external parameters. They show that the SPR phase jump and the strong dependence of the SPR phase upon system parameters can be used to improve the sensitivity of the SPR microscopy technique [13] and develop interferometric SPR imaging with monoatomic thickness and micrometer spatial resolutions. The sensitivity to a refractive index change was estimated as  $4 \times 10^{-8}$  in a model experiment with different gases.

#### 4.5 Different Plasmon Modes

Apart from using microdot or microwell arrays instead of flat gold films to improve sensitivity of the SPR method, it is possible to use different plasmon modes [14]. It was considered long-range SPR, coupled plasmon-waveguide resonance, waveguide-coupled SPR in comparison with conventional SPR for three of the most commonly applied detection methods, namely angular interrogation, wavelength interrogation, and intensity measurement. Under angular interrogation, it has been shown that the sensitivity of the CPWR biosensor is approximately 10 times poorer than that of the conventional SPR device since the former device utilizes multiple beam interference within the waveguide layer, whereas the latter employs electric enhancement of the SPR. The LRSPR biosensor enhances the electric field on either side of its thin metal film by establishing a symmetric configuration in which the dielectric constants of the dielectric buffer layer and the buffer solution are equal. Under wavelength interrogation, the dielectric constant of any media can be modulated, and hence the sensitivity of the LRSPR biosensor can be slightly increased. However, its sensitivity still remains far poorer than that of a conventional SPR. Furthermore, the LRSPR demands a symmetric configuration, and this is not easily attained when detecting environments of varied physiologies. Finally, the WCSPR biosensor provides a sensitivity which is only marginally lower than that of the conventional device by coupling the waveguide and SPR resonances.

#### 5 Conclusion

On the basis of the literature review, analysis of available opportunities and due to lack of access to equipment for the deposition of metal coatings it was decided to abandon the use of different plasmon modes and also microarrays as excitation area instead of flat gold film and attempt to use the phase-jump interferometric SPRI method with together with further improvement of the experimental setup and the processing algorithm above-mentioned. Besides, it is evident that the higher sensitivity the sensor has, the less its operating range. In order to avoid this disadvantage, it is suggested to divide reflected beam into two channels and apply different techniques of processing to the channels with providing higher sensitivity in one and wider operating range in other (like in case of accurate and coarse adjustment).

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