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Polarimetric detection of the photon excitation of surface plasmons

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Abstract. A polarimetric method for the detection of surface plasmons, excited by linearly polarised light, was proposed and tested. It was established that this method is not only more sensitive to variations in the parameters of the waveguiding structure but also makes it possible to increase by one – two orders of magnitude the precision of determination of the excitation efficiency and of the phase velocity of the plasmons, compared with the amplitude method for plasmon detection. The possibility of applying the method in sensor devices, in polarimetry, and in the microscopy of conducting surfaces is discussed.

1. Introduction

Surface plasmons (SPs) represent a variety of surface electromagnetic waves in the visible region and are used widely not only in the optical spectroscopy of surfaces [1], but also in microscopy [2, 3], ellipsometry [4, 5], and sensors of external perturbations [6, 7]. According to the *Science Citation Index* database, more than 300 communications concerning the nature and applications of SPs have been published annually during the last decade. Most of these communications are devoted to various sensors, methods, and devices. Industrial fabrication of a number of plasmon sensors has been mastered [8, 9]. The principal advantage of plasmon devices is the high sensitivity of their characteristics to the state of the surface, which guides SPs, and of the transition layers on the surface.

In monitoring measurement devices, plasmons are usually generated by photons using one of the schemes of the frustrated total internal reflection (FTIR) method. Linearly polarised radiation, in which the electric vector oscillates in the plane of incidence, is then generally used. Such radiation corresponds to polarisation of the SP field and ensures minimum noise and maximum image contrast in SP microscopy [10]. In this case, the excitation of SPs is accompanied only by a decrease in the reflected radiation intensity and an additional phase change [11]. Two methods are therefore used nowadays in the reflectometric detection of the SP excitation: the amplitude method [12], in which the reflected-radiation intensity is measured, and the phase method proposed in Refs [13] and [14] and implemented in Refs [15] and [16], in which the change in the phase of the SP exciting radiation is measured. A combination of these methods may also be employed [17].

The simpler amplitude method for the detection of the photon excitation of SPs is used most widely. In this method, either the frequency ω or the angle of incidence φ of the incident radiation is scanned and the relationship $R_{\mathrm{p}}(\omega)$ or $R_{\mathrm{p}}(\varphi)$ (R_{p} is the energy reflection coefficient of the structure for the p-component) is determined in measurement of the intensity of this component of the reflected radiation. The excitation of SPs is indicated by a resonance dip in the $R_{\mathrm{p}}(\omega)$ or $R_{\mathrm{p}}(\varphi)$ plot. The excitation efficiency η and the phase velocity of SPs (inversely proportional to the real part k' of the complex wave number $k = k' + \mathrm{j}k''$ of the SPs) may be determined, and the SP absorption coefficient (proportional to k'') as well as the depth of penetration of the SP field into the surrounding medium may be estimated from the shape and depth of the resonance dip and also from its position on the ω or φ axis. However, the precision of the measurement of η and k' is low (the error is of the order of $10^{-3} - 10^{-4}$), which has a negative effect on the precision of determination of an external perturbation or of the parameters of the transition layer.

We examined and tested a polarimetric method for detection of the photon excitation of SPs which makes it possible to measure η and k' with an error down to $10^{-5} - 10^{-6}$.

2. Brief theory

We shall examine the excitation of SPs by a planar linearly polarised monochromatic electromagnetic wave with non-zero p- and s-components of the field, i.e. by a wave in which the plane of polarisation (the plane in which the vector $\boldsymbol{E}$ oscillates) is tilted by an angle Θ_0 relative to the plane of incidence. The reflection of such a wave from a layered structure is known to change not only the amplitudes, but also the phases of both components of its vector $\boldsymbol{E}$ [18]. This occurs to an even greater extent in the reflection of a wave from the absorbing structure, which guides SPs. Therefore, in the photon excitation of SPs the reflected radiation acquires an elliptical polarisation in which this radiation has no definite plane corresponding to the oscillations of the vector $\boldsymbol{E}$.

Since the SP excitation in the FTIR method takes place for an angle of incidence greater than the critical value, the reflection of the radiation from the prism base is accompanied by a change in the phase and amplitude of only the p-component of the radiation (corresponding to the polarisation of the SP field), whereas the s-component hardly changes under these conditions [11, 16]. If the phase shift of the p-component arising on reflection is compensated, the reflected radiation still remains linearly polarised. However, as a result of the decrease in the intensity of the p-component,

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the plane of polarisation of the reflected radiation is tilted relative to the plane of incidence by an angle $\Theta > \Theta_0$. The greater the SP excitation efficiency η, the greater the value of Θ under these conditions. Thus the polarimetric detection of the photon excitation of SPs and the polarimetric determination of the efficiency of this excitation become possible.

We shall establish an analytical dependence of the angle Θ on Θ_0, R_p, and η. Let us designate the p- and s-component of the intensity of the incident linearly polarised wave by I_p^0 and I_s^0, respectively. The equality $\tan\Theta_0 = I_s^0/I_p^0$ is then valid (Fig. 1). Reflection of the wave from the prism base, optically coupled (via the evanescent field) to the waveguiding structure, makes the intensities of the radiation components assume the following values: $I_p^* = R_p I_p^0$ and $I_s^* = R_s I_s^0$ (R_s in the energy reflection coefficient for the s-component). Since in the case of the s-component of the incident light the SPs are excited under the conditions of total internal reflection, it follows that $R_s \approx 1$ and $I_s^* \approx I_s^0$. When the phase shift arising between the p- and s-components on reflection is compensated, we have

$$\tan\Theta = \frac{I_s^*}{I_p^*} \approx \frac{I_s^0}{R_p I_p^0} = \frac{\tan\Theta_0}{R_p} \,. \tag{1}$$

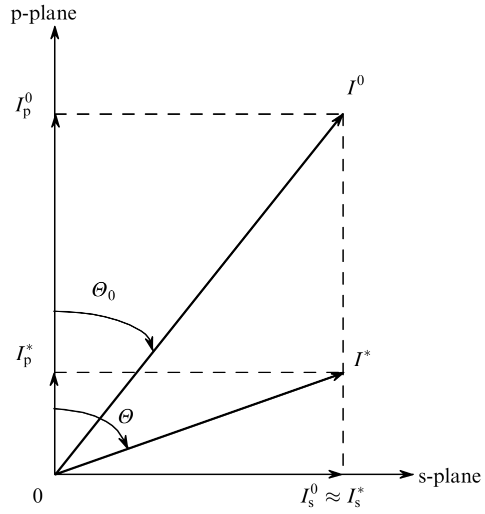

Figure 1. Rotation of the plane of polarisation of linearly polarised light in SP excitation by the angle $\Theta - \Theta_0$ (Θ_0 and Θ are the angles between the plane of incidence and the plane in which the vector $\boldsymbol{E}$ oscillates in incident and reflected light).

Having substituted the expression for the SP excitation efficiency

$$\eta = \frac{I_p^0 - I_p^*}{I_p^0} = 1 - \frac{I_p^*}{I_p^0} = 1 - R_p \,,$$

in formula (1), we obtain

$$\Theta(\eta) = \arctan\frac{\tan\Theta_0}{1-\eta} \,. \tag{2}$$

It follows from formulas (1) and (2) that we have $\Theta = 90°$ for 100% SP excitation efficiency, corresponding to $R_p = 0$ and $\eta = 1$, whereas the relationship $\Theta(\eta)$ is of the resonance type, becoming more pronounced with decrease in Θ_0 (Fig. 2).

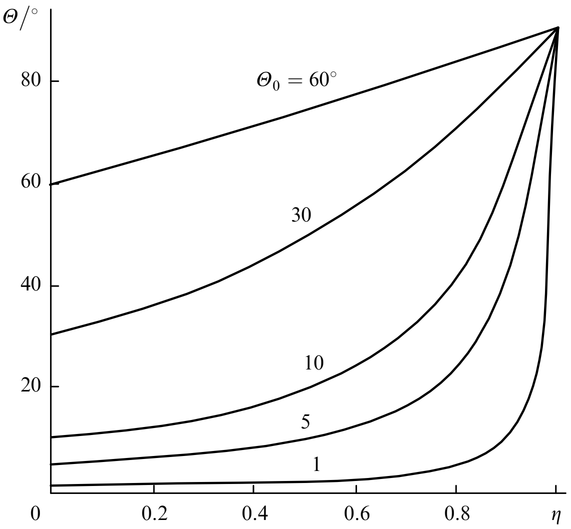

Figure 2. Dependences of the angle of tilt Θ of the plane of polarisation of the reflected light on the SP excitation efficiency η for various angles of tilt Θ_0 of the plane of polarisation of the incident light.

3. Numerical modelling

We carried out a numerical modelling of the rotation of the plane of polarisation of linearly polarised light with the wavelength $\lambda_0 = 632.8$ nm, which excites SPs in a structure which consists of a glass prism with the refractive index $n_1 = 1.51$, a transparent gold film (with $n_2 = 0.14$ and the absorption coefficient $k_2 = 3.30$) of a thickness d and deposited on the planar base of a prism placed in air ($n_2 = 1.0$). For $d = d_0 = 53.2$ nm and the angle of incidence $\varphi = \varphi_0 = 44°10'$ the SP excitation efficiency attained in such a structure is 100%.

The dependences of the angle of tilt Θ of the plane of polarisation of the reflected radiation on the angle of incidence φ were calculated initially for $d = d_0$ and various values of Θ_0 (Fig. 3). The plots in Fig. 3 show that:

(1) the angular positions of the minima of the resonance dips on the $R_p(\varphi)$ curves coincidence with the angular positions of the peak vertices on the corresponding $\Theta(\varphi)$ curves;

(2) for $\Theta_0 < 30°$, the angular width of the resonance peaks is appreciably smaller than the angular width of the resonance dips;

(3) since the error in determination of the angular position of the plane of polarisation with the aid of commercial polarimeters is not more than $1'$ [19], it follows that for $\Theta_0 \leqslant 1°$ the angle of incidence φ_0, corresponding to the maximum SP excitation efficiency, may be determined with an error not greater than $1''$, which exceeds by two orders of magnitude the error in determination of φ_0 when the amplitude method is employed to detect SPs.

Our numerical experiment thus confirms the hypothesis that, for low values of Θ_0, the angle of tilt of the plane of polarisation of the reflected radiation relative to the plane of incidence is more sensitive to the SP excitation efficiency η than is the reflection coefficient R_p.

Next, in order to estimate the sensitivity of the polarimetric method for the detection of SPs to variations in the waveguiding-structure parameters, the relationships $R_p(\varphi)$ and $\Theta(\varphi)$ were calculated for $\Theta_0 = 1°$ and various thicknesses d of the gold film (Fig. 4). An analysis of the plots $R_p(\varphi)$ shows that, when d is varied in the range

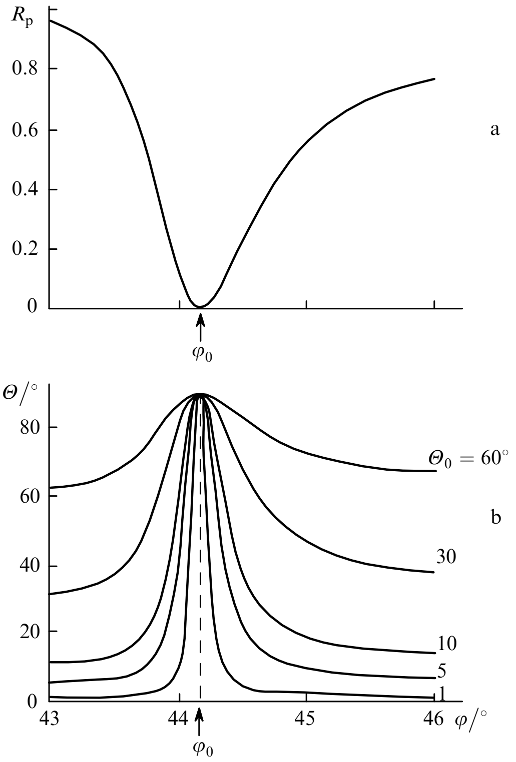

Figure 3. Dependences of the reflection coefficient R_p (a) and of the angle of tilt Θ of the plane of polarisation of the reflected light (b) on the angle of incidence φ for 100% SP excitation efficiency in the 'prism ($n_1 = 1.51$) — gold film ($n_2 = 0.14$, $k_2 = 3.30$, thickness $d = d_0 = 53.2$ nm) — air' structure and different angles of tilt Θ_0 of the plane of polarisation of the incident light with the wavelength $\lambda = 632.8$ nm.

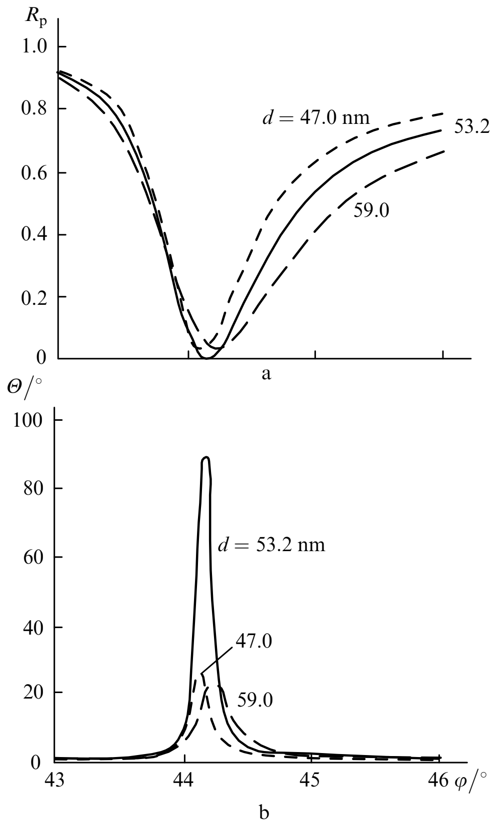

Figure 4. Dependences of the reflection coefficient R_p (a) and the angle of tilt Θ of the plane of polarisation of the reflected light (b) on the angle of incidence φ in excitation (by this radiation) of SPs in structures with various thicknesses d of the gold film (described in the caption to Fig. 3).

47.0 nm $< d < 59.0$ nm, the change in R_p at the dip minimum, corresponding to the maximum SP excitation efficiency η, does not exceed 0.05, i.e. $\sim 5\%$ of $R_{p\,\max} \approx 1$. On the other hand, the maximum angle of tilt Θ of the plane of polarisation of the reflected radiation, relative to the plane of incidence, corresponding to such variations in d, changes from 90° to 18°, i.e. by ~80%. Similar calculations performed for the same structure with variation of the refractive index n_3 of the surrounding medium showed that the sensitivity of the angle Θ to changes in n_3 also exceeds by one or two orders of magnitude (as a function of Θ_0) the sensitivity of R_p.

This allows to conclude that the sensitivity of the polarimetric method (for detection of the photon excitation of SPs) to variations in the waveguiding-structure parameters is significantly greater than the sensitivity of the amplitude method.

4. Experimental studies

There is no need to construct apparatus for the polarimetric detection of the photon excitation of SPs. It can be carried out with the aid of a commercial ellipsometer containing all the necessary components (with the exception of the waveguiding structure used as a sample): a polariser, a controlling compensator, and an analyser, which all have an axis of rotation and are provided with dials, as well as the source of monochromatic radiation and a sensitive photodetecting device.

The only component which needs to be updated is the compensator, the role of which in commercial ellipsometers is performed by a quarter-wave plate. The phase shift introduced by the plate between the p- and s-components of the radiation depends on the angle of tilt of the optical axis of the plate relative to the plane of incidence and varies in the range from zero to $\pi/2$ when the plate is rotated by 45°. However, in the photon excitation of SPs, the phase shift between the radiation components may reach 2π [4, 16], so that the wave plate having an axis of rotation must be selected as the controlling compensator for the polarimetric detection of the photon excitation of SPs with the aid of an ellipsometer. The measurements can be carried out by using the PCSA (polariser – compensator – sample – analyser) or PSCA (polariser – sample – compensator – analyser) setups, familiar in ellipsometry, with a fixed angular position of the polariser and with a rotatable compensator and analyser [18].

The polarimetric method was tested with the aid of a LÉF-3M ellipsometer provided with an He – Ne laser ($\lambda_0 = 632.8$ nm). Four connected quarter-wave plates with combined optical axes served as the compensator in the ellipsometer. A glass half-cylinder (with the 2.5 cm radius of curvature of the surface, which is much larger than the diameter of the laser beam, and with the refractive index $n_1 = 1.51$) played the role of an FTIR prism and of a mechanical base for the waveguiding structure. The choice of a prism in the form of a half-cylinder makes it possible to avoid the shifts of the light spot on the waveguiding structure when the angle of incidence of the radiation is varied. A transparent continuous copper film of a thickness $d \approx 50$ nm (estimated from the transmission coefficient) was deposited by the vacuum thermal vaporisation method on the planar face (base) bracing the half-cylindrical surface. The optical constants of the film deposited from the vapour phase, determined by the ellipsometric method by using a control sample, proved to have the following values: $n_2 = 0.21$ and $k_2 = 3.35$. The half-cylinder with the copper film deposited on it was placed in air with the base on the spacing slab at the centre of the object stage of the ellipsometer (air was the surrounding medium).

We determined initially the $\psi(\varphi)$ relationship, where $\psi = \arctan(R_p/R_s)$ is the polarisation angle in ellipsometry, by the familiar method of ellipsometric detection of the SP modes in thin metal films [20]. The $\psi(\varphi)$ relationship obtained is presented in Fig. 5a. The minimum angle $\psi = 6°27'$ is attained for the angle of incidence $\eta = 44°06'$ and it corresponds to the maximum SP excitation efficiency $\eta = 98.8\%$ in the investigated structure. If necessary, the $\psi(\varphi)$ relationship can be readily converted (bearing in mind the equality $R_s \approx 1$) into the $R_p(\varphi)$ relationship.

Polarimetric measurements of the dependence of the angular position Θ of the plane of polarisation of the reflected radiation on the angle of incidence φ were then performed for the same structure. The method of 'null measurements' with a fixed position of the polariser and controllable angular positions of the compensator and analyser was employed.

Fig. 5b presents a family of $\Theta(\varphi)$ curves obtained for different fixed angles of tilt Θ_0 of the polariser relative to the plane of incidence. The shape of these curves agrees well with the corresponding calculated relationships, confirming the high sensitivity of the orientation of the plane of polarisation of the reflected radiation to the SP excitation efficiency. Analysis of the curves in Fig. 5 also shows that, for an error of measurement of R_p of 1% and an error of measurement of Θ of 1′, the angle φ_0 may be determined with an error down to 10′ when the amplitude method is used and with an error not worse than 30″ when the polarimetric method is employed (for $\Theta_0 < 30°$). Thus our experiments confirmed the potential capabilities of the proposed method.

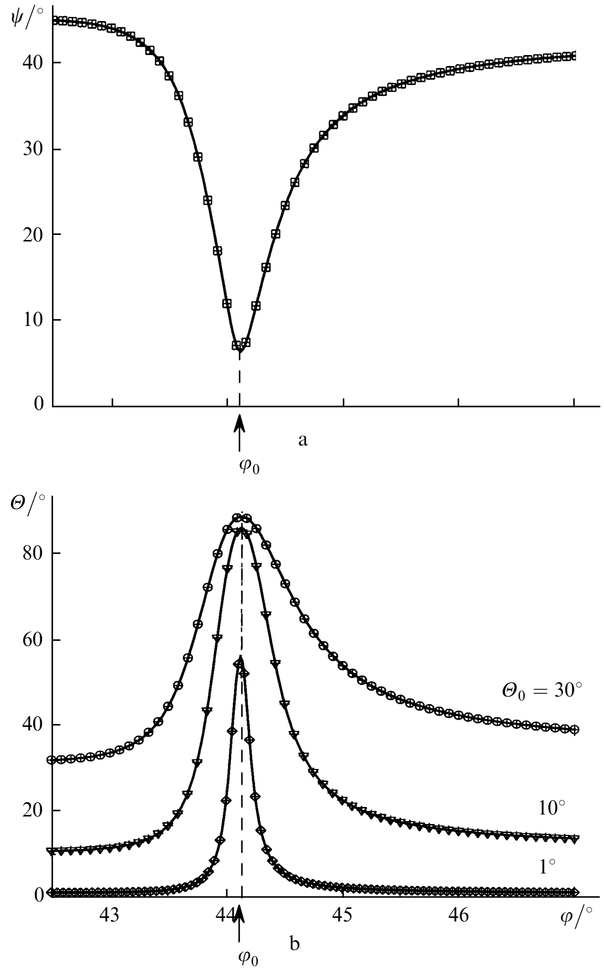

Figure 5. Dependences of the ellipsometric angle ψ (a) and of the angle of tilt Θ of the plane of polarisation of reflected light (b) on the angle of incidence φ, measured on reflection of linearly polarised light with $\lambda = 632.8$ nm from the 'prism ($n_1 = 1.51$) — copper film ($n_2 = 0.21$, $k_2 = 3.35$, thickness $d = 50$ nm) — air' structure for various angles of tilt Θ_0 of the plane of polarisation of incident light.

5. Conclusions

When the phase shift between the p- and s-components (arising on excitation of SPs by linearly polarised light) is compensated, the plane of polarisation of the reflected radiation is rotated by the angle $\Delta\Theta = \Theta - \Theta_0$, where Θ_0 and Θ are the angles between the plane of incidence and the plane in which the vector $\boldsymbol{E}$ oscillates in incident and reflected light. The angle $\Delta\Theta$ may serve as a measure of the efficiency of the photon excitation of SPs, which makes it possible to employ a new method for the detection of SPs, namely the polarimetric method. The high sensitivity of the angle $\Delta\Theta$ to the SP excitation efficiency may be used productively in the design of a new generation of plasmon sensors, measuring devices, and SP microscopes. We also note that the strong dependence of the angle $\Delta\Theta$ on the orientation of the plane of polarisation of the incident radiation permits application of the proposed method in order to increase the sensitivity of polarisation microscopy and saccharimetry.

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