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LONG-RANGE PLASMONS IN ASYMMETRIC FOUR-LAYER STRUCTURE: THE PHASE-POLARIZATION CONTRAST METHOD

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Abstract

An asymmetric four-layer structure supporting a long-range surface plasmon mode for sensing purposes was investigated. The excitation of the plasmon modes was achieved by means of a corrugated grating. The anomalous increase in the reflectivity was observed experimentally. The phase-polarization contrast method for a long-range surface plasmon was investigated experimentally.

1. Introduction

A long search for the long-range surface plasmon (LRSP) has been made since Sarid [1] showed the possibility of existence of long-range surface plasmons propagating along a thin metallic slab embedded in a dielectric medium. It was initially thought that LRSP can exist only in nearly symmetric structures. However, it was shown in [2] that a LRSP also exists in asymmetric structures provided the modal electric field can be made to pass through zero near the middle of the metal film.

Experimental evidence of LRSP in both symmetric and asymmetric structures was given in [3, 4]. We demonstrated that the structures supporting a LRSP can be used as a basis for sensor applications [5]. We report here on further theoretical and experimental work aimed at evaluating the expectable sensitivity of LRSP structures for an evanescent wave sensor of the refractometric type.

2. LRSP along Asymmetric Structures

Surface plasmons are collective oscillations of electrons propagating along a metal–dielectric interface at optical frequencies. The associated electromagnetic field is of TM polarization. The properties of surface plasmon modes propagating along a metal film depend on the film thickness d and on the permittivity of the dielectric media at both sides of the film. When one has a metal thin film, there is one plasmon at each interface. In the case where the metal film is arranged symmetrically in a single dielectric, the propagation constants of surface plasmon modes propagating at both sides of the metal film are the same. When the film thickness d becomes smaller compared to the skin depth in the metal, the electromagnetic modal fields of these two surface waves overlap. This results in the formation of a mode with a large propagation constant β^+ (the longitudinal electric field component is symmetric and the magnetic field is asymmetric) and a second mode with a low propagation constant β^- (the longitudinal electric field component is asymmetric and the magnetic field is symmetric).

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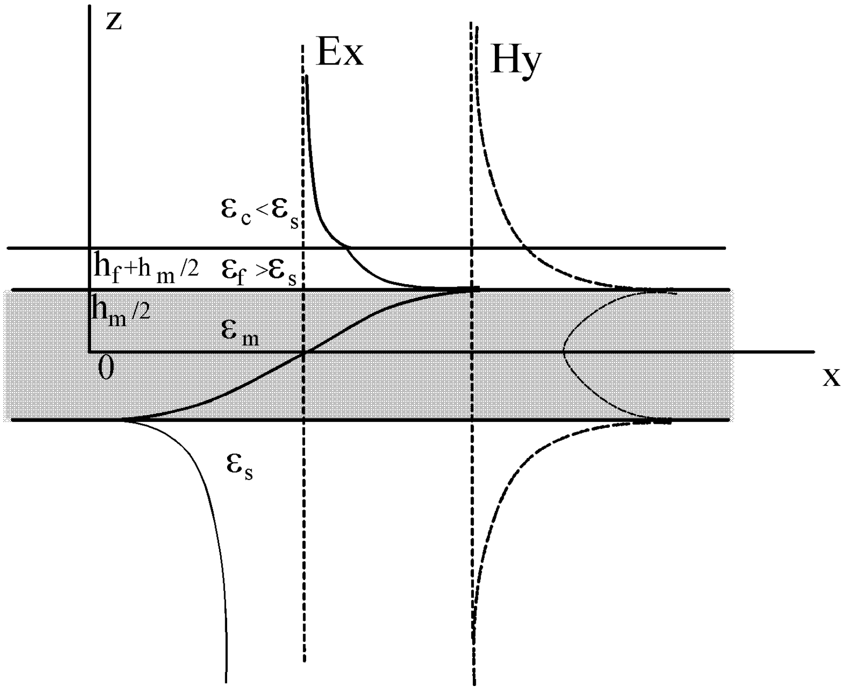

Fig. 1. Longitudinal electric field of a surface plasmon along a symmetric (or quasisymmetric) structure. The dashed lines represent a symmetric mode with large propagation constant β^+. The solid lines represent an asymmetric mode with small propagation constant β^-.

The complex propagation constant $\beta_x = \beta_x' + i\beta_x''$ of these longitudinal electronic oscillations is determined by the dispersion relationship resulting from the continuity condition for the tangential field component at both interfaces. The imaginary part β_x'' is related to metal absorption. The power carried by a surface plasmon along the smooth and flat surface decreases as $e^{-2\beta_x''}$. The values of β^+ and β^- depend on the complex permittivities of the metal film and surrounding medium and especially on d. As has been found in studies of the propagation constant in a symmetric structure [1], the damping of the low spatial frequency mode β^- (i.e., the mode with an asymmetric electric field distribution and a symmetric magnetic field distribution) decreases with decreasing thickness d. The reverse is true for the mode with the propagation constant β^+ (i.e., the mode with a symmetric electric field distribution and an asymmetric magnetic field distribution). The physical reason is the electric-field attenuation in the metal film, so that the light dissipation (Joule heat) diminishes. In the metal film, the value of E_x decreases with decreasing thickness d, because near the cut-off ($d = 0$) the field E_x is pulled into the substrate. The length L at which the intensity decreases by e times is proportional to $(2\beta'')^{-1}$, hence L increases with decreasing thickness d (for β^-). Thus the condition for LRSP is a zero field for the E_x component in the middle of the metal film. This condition can be met in asymmetric structure (where $\varepsilon_c \neq \varepsilon_s$) as well as in symmetric one (see Fig. 2 for the case where $\varepsilon_c = \varepsilon_f$). In this case, a dielectric medium with lower permittivity should be inserted between the metal film and the dielectric of higher permittivity.

In this section, the parametric dependence of the thickness of the metal film h_m on the dielectric layer thickness h_f (Fig. 2) leading to a LRSP in an asymmetric structure is studied. There is no clear definition of what kind of long-range plasmon mode it should be. It is just one of two modes whose electron driving field has a zero crossing within the metal film. Both the zero-point position of the E_x component on the z axis and the resulting propagation losses depend strongly on the dielectric from the high index side of the metal film. The thickness of dielectric h_f and its refractive index n_f leading to the largest propagation length L

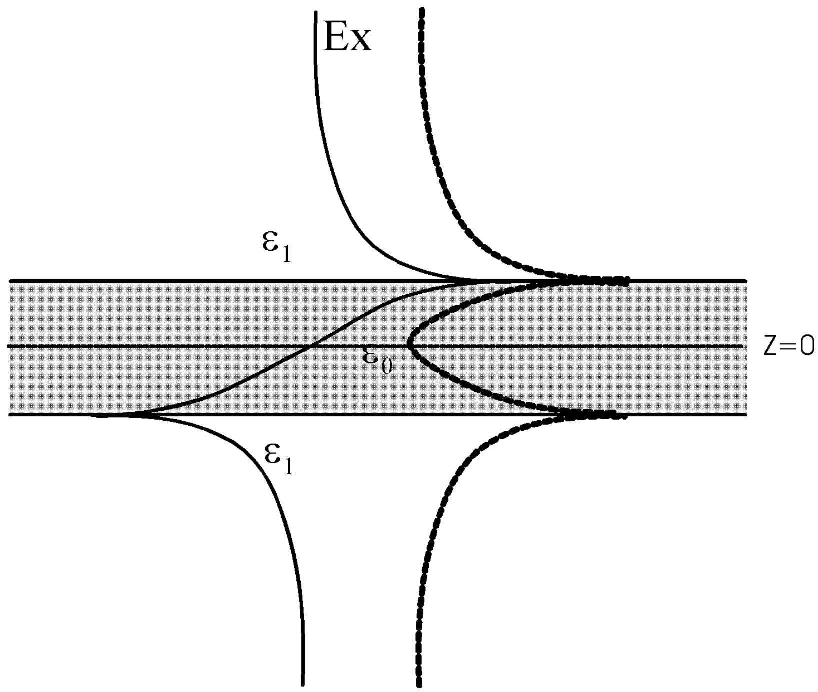

Fig. 2. LRSP field along an asymmetric structure.

are defined by the four-media dispersion relation (1). The latter can be obtained by matching the tangential field components at three boundaries of the structure. By analogy with the symmetric case and with the aim of obtaining an analytical expression for the sensing characteristics in a closed form, we define here the LRSP structure as that ensuring zero crossing of the longitudinal electric field in the middle of the metal film. As will be shown below, this choice is physically plausible, and systematic numerical modeling shows that the actual field profile leading to the least propagation losses is very close to this condition.

As assumed above, minimum losses can be obtained when the electric field component E_x has a nonsymmetric distribution (Fig. 1) in the metal layer. In this case, the tangential magnetic field component H_y has a symmetric distribution (see Fig. 2):

$$H_y\big|_{z=h_m/2} = H_y\big|_{z=-h_m/2}.$$

This is not true for its normal derivative:

$$\left.\frac{\partial H_y}{\partial z}\right|_{z=h_m/2} = -\left.\frac{\partial H_y}{\partial z}\right|_{z=-h_m/2}.$$

After dividing the second expression by the first one and by the metal permittivity ε_m, we obtain the following expression:

$$\left.\frac{1}{\varepsilon_m H_y}\frac{\partial H_y}{\partial z}\right|_{z=h_m/2} = -\left.\frac{1}{\varepsilon_m H_y}\frac{\partial H_y}{\partial z}\right|_{z=-h_m/2}. \tag{2.1}$$

The symmetric solution of the Maxwell equations is a hyperbolic cosine function in the metal film. Therefore, substituting the expression for H_y into Eq. (1) gives the following relationship for the lower interface ($z = -h_m/2$) in the metal film:

$$\left.\frac{1}{\varepsilon_m H_y}\frac{\partial H_y}{\partial z}\right|_{z=-h_m/2} = \frac{k\sqrt{n_e^2-\varepsilon_m}}{\varepsilon_m}\tanh\left(kh_m\sqrt{n_e^2-\varepsilon_m}\right), \tag{2.2}$$

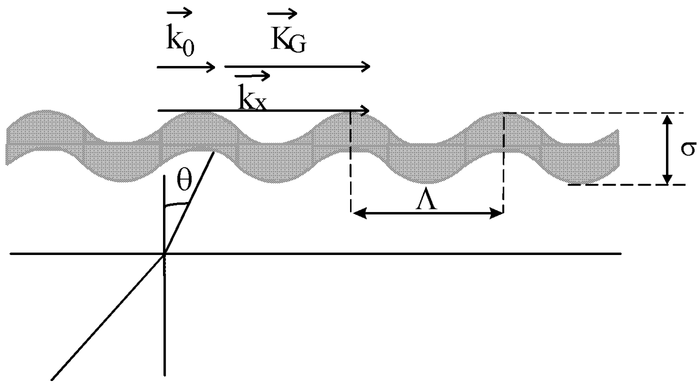

Fig. 3. Excitation of the surface plasmons using a grating.

where ε_s is the substrate permittivity and n_e is the effective refractive index of the LRSPs.

The solution of the Maxwell equations is an exponential function in the substrate. Substituting the expression for H_y into the continuity relation of the tangential component of the magnetic field at the lower metal/dielectric interface ($z = -h_m/2$) in the metal film, we obtain

$$-\tanh\left(\frac{kh_m}{2}\sqrt{n_e^2-\varepsilon_m}\right) = \frac{\varepsilon_m\sqrt{n_e^2-\varepsilon_s}}{\varepsilon_s\sqrt{n_e^2-\varepsilon_m}}\,. \tag{2.3}$$

A similar equation can be obtained at the upper metal interface ($z = h_m/2$). The product $(1/\varepsilon H_y)\partial H_y/\partial z$ is continuous at both sides of the interface since

$$\frac{1}{\varepsilon}\frac{\partial H_y}{\partial z} = jkE_x.$$

Thus we can obtain the thickness h_f of the layer of high refractive index which is needed for achieving a LRSP in an asymmetric structure:

$$kh_f\sqrt{\varepsilon_f-n_e^2} = M\pi + \arctan\frac{\varepsilon_f\sqrt{n_e^2-\varepsilon_c}}{\varepsilon_c\sqrt{\varepsilon_f-n_e^2}} - \arctan\frac{\varepsilon_f\sqrt{n_e^2-\varepsilon_s}}{\varepsilon_s\sqrt{\varepsilon_f-n_e^2}}\,, \tag{2.4}$$

where ε_f and ε_c are the permittivities of the dielectric layer and cover medium, respectively, and h_f is the thickness of the dielectric layer. Equations (3)–(4) give the parametric dependence $h_f(h_m)$ for the LRSP in the asymmetric structure, i.e., the thickness h_f which the dielectric must have to fulfill the condition of minimum propagation losses for a given thickness h_m of the metal film. This parametric dependence of the dielectric-film thickness was obtained under the hypothesis that the plasmon losses (i.e., the imaginary part of its effective index) are minimum when the E_x-field component is zero in the middle of the metal film.

3. Grating Excitation of LRSP

As it follows from the dispersion relation (4), the excitation of a surface plasmon requires a coupler (a grating coupler or a prism one). We are concentrating here on the scheme of the LRSP excitation using a grating. As distinct from the case of a grating coupled dielectric waveguide, the presence of the surface undulation represents a strong perturbation both of the modal field and light guidance properties.

The grating acts as a coupler between the surface plasmon field and incident plane wave. It diffracts light in different orders, one of which is used to couple the free-space wave to the plasmon. The spatial harmonics

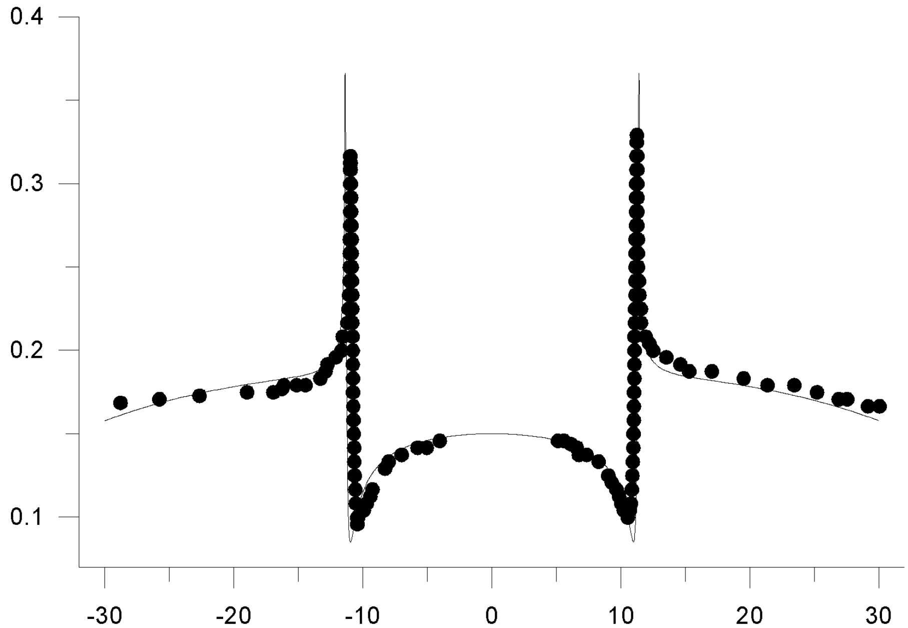

Fig. 4. Dependence of the reflectivity on the angle of incidence onto the corrugated structure. Numerical modeling (solid line) of a four-layer structure in comparison with the experimental results (points).

$\vec{k_x}$ along the x axis of the incident beam impinging on the grating of period Λ at a resonant angle Θ are given by the wave vectors $\vec{k_x} = \pm N\vec{K_G} + \vec{k_0}$, where $k_0 = \omega_0/c$, $K_G = 2\pi/\Lambda$, and N is an integer which represents the diffraction order of the grating. A large coupling efficiency is usually obtained by using the first diffraction order ($N = 1$).

A plasmon propagating along an undulated metal slab exhibits losses due to the propagation in the absorptive medium. The additional losses come from the field redistribution in the grating region. If σ represents the groove depth, the losses are proportional to σ^2.

The plasmon mode also diffracts into plane waves in all possible diffraction orders at a rate which is described by the radiation coefficient α. The latter is also proportional to σ^2.

In order to achieve the maximum coupling of the incident wave energy with a plasmon mode, we have to find a balance between the radiation losses and the dissipative losses in the thin metal film. This condition can be satisfied by the appropriate choice of σ.

4. Experimental Results

Long-range plasmon excitation was performed in thin copper films deposited on a corrugated glass substrate ($n_s = 1.512$). The gratings were first made on a glass substrate by standard holographic exposure with a He–Cd laser followed by ion beam etching to transfer the grating from the resist film onto the glass substrate. The grating period was $\Lambda = 0.37$ μm and the groove depth was 56 nm. The resulting groove profile prevents the deposition of continuous metal thin film. Because of this, the samples were annealed in a furnace at a temperature close to the glass softening one. As a result of such an annealing, the grating profile is nearly sinusoidal and allows the deposition of continuous undulated metal films. Copper films ($\varepsilon'_m = -8.24$, $\varepsilon''_m = 2.2$) with a thickness of 10 nm were deposited by RF sputtering in a "Sputron-II" installation in oil-free

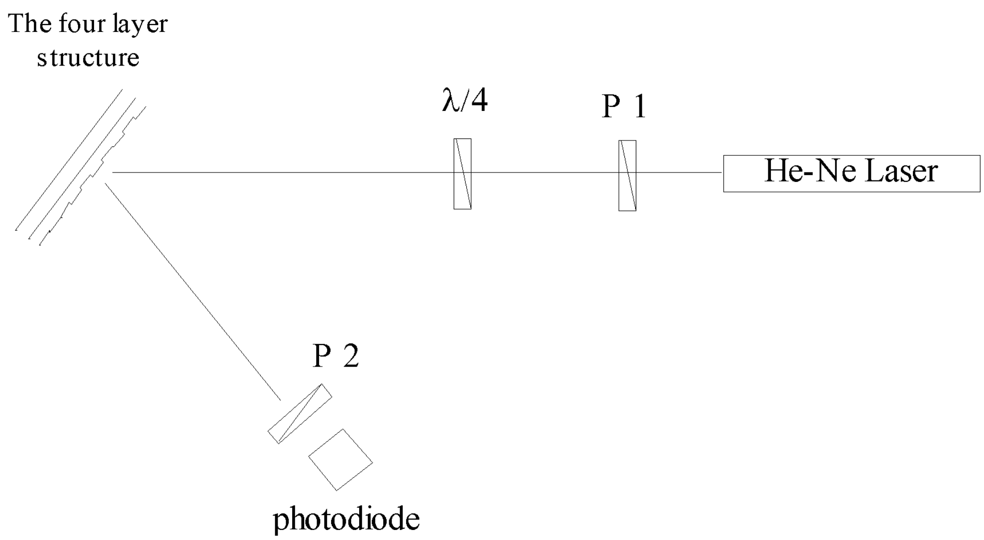

Fig. 5. Experimental set-up for measuring the phase shift.

vacuum. The copper film was covered by a 116 nm thick Si_3N_4 film ($n_f = 1.81$) [3].

The structure described was characterized at a wavelength of 632.8 nm with air as a cover medium. Figure 4 shows experimental evidence of the anomalous LRSP reflection. Two reflection bands are observed which are caused by excitation of the LRSP. In spite of its relatively low anomalous reflection maximum, the long-range plasmon propagating along a corrugated grating film seems to be attractive for use in chemical and biochemical sensors because of the narrow angular and spectral width of the resonant reflection. The four-layer structure supporting the long-range (LR) modes was shown [4] to be promising for the creation of sensors. This structure is not very sensitive to the variation of dielectric parameters of the cover media but has a good resolution. The possibility of measurement of the permittivity of ultrathin metal films was experimentally demonstrated in [5]. However, measurement of the dielectric parameters of a variety of dielectric media covered by another dielectric presents a serious problem. Any multilayer structure supporting a LR mode is optimized for the particular value of the refractive index of the cover medium. The optimal parameters include the corrugation depth, metal film thickness, and, in particular, the thickness of the dielectric layer. However, a change in the dielectric medium of the cover (for example, water instead of air) changes the optimum conditions. This is a rather important problem in sensor and other applications.

5. Experimental Evaluation of Sensitivity

As follows from the theory described above, there is an optimum thickness of the dielectric layer for any cover medium. However, in practice (for example, in biosensor applications), we often have to optimize the structure for the particular cover medium and the dielectric layer being studied.

The phase-polarization contrast method was shown to be applicable in the case of non-optimal coupling of an ordinary surface plasmon [6, 7]. We used this approach for the LRSP resonance in an asymmetric structure. A schematic diagram of the experimental set-up is presented in Fig. 5. The polarization of light coming from the He–Ne laser ($\Lambda = 0.6328$ μm) is controlled by a $\lambda/4$ plate and a polarizer (P1). This polarization is fixed in an effort to find the angular position of the sample which corresponds to the zero intensity detected (for the perpendicular position of P2 relative to P1 polarization).

Figure 6 presents the results of measurements of the angular dependence of light intensity reflected from

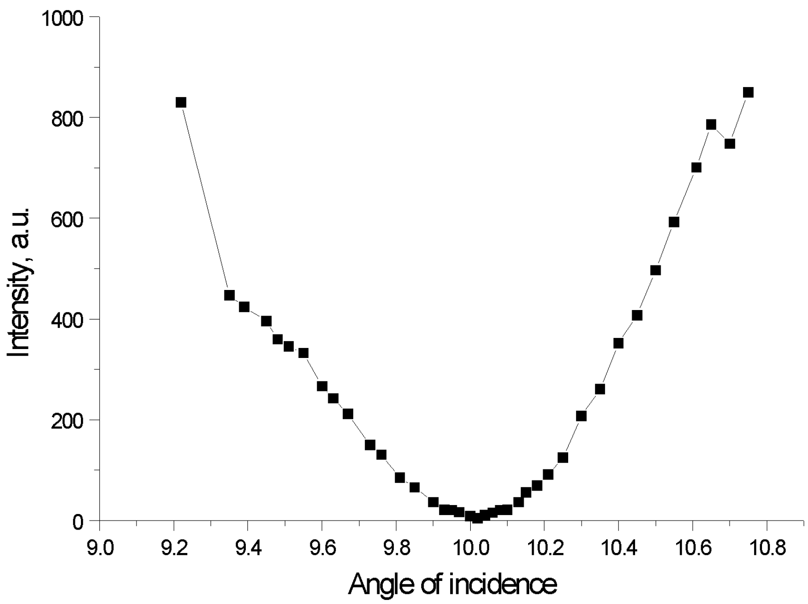

Fig. 6. Experimental results for a four-layer structure with water as the cover layer.

the surface of corrugated four-layer structure with water as the cover layer. Thus, the possibility of optimizing the LRSP structure, which was initially intended to be used with air as a cover medium, was demonstrated experimentally in the case where water was used as the cover medium.

6. Conclusions

Theoretical studies of LRSP in an asymmetric structure as applied to evanescent wave sensing were undertaken. The dispersion equations of the LRSP in the asymmetric structure were obtained. The phase-polarization contrast method was used for optimizing the four-layer LRSP structure. We have shown both theoretically and experimentally that a LRSP exhibits a pronounced effect of the anomalous reflection. This effect is of practical interest for sensor applications since it provides the reflection maximum along with high angular resolution rather than the reflection dip typical of an ordinary plasmon. This approach based on the use of the reflection maximum instead of the reflection minimum opens wide perspectives in a variety of applications for corrugated multilayer structures supporting LR modes.

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