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Sensors

Peculiarities of implementation of spectral SPR effect in high-conductive metal thin films for creating SPR gas sensors

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Abstract. In this paper, we consider the conditions for excitation of surface plasmon resonance (SPR) in thin films of a number of high-conductive metals (gold, silver, copper, and aluminum) with the aim of using the features of the SPR spectral effect to create chromatic sensors of gas environment. A new approach to assessing the feasibility of SPR implementation in films of these metals in different regions of the visible spectrum is proposed. This approach is based on the analysis of spectral characteristics of surface plasmon-polariton attenuation length at metal/dielectric interface. The relationship between the attenuation length and the optical constants and imaginary part of the permittivity of the metals, as well as a number of essential parameters of the SPR effect is considered and a certain correlation between them is demonstrated. It is established that for exciting SPR, the attenuation length of a surface plasmon-polaritons in metal films should exceed 2 μm. The results of the presented theoretical analysis are compared with the experimentally obtained spectra of plasmon-scattered light in the visible light wavelength range.

Keywords: surface plasmon resonance, high-conductive metals, plasmon-polariton, attenuation length, wave vector, SPR spectral effect.

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1. Introduction

Developing sensors for detecting biomolecular interactions in solutions as well as volatile molecules in a gas phase based on the surface plasmon-polariton resonance effect [1, 2] has been an actual task for many decades [3–13]. Recently, close attention has been focused on a possibility of using various non-traditional materials as plasmon-carrying coatings. Nevertheless, traditional basic coating options based on the main types of high-conductive metals remain relevant. Each of these metals has its own physical properties, spectral region for SPR implementation and, hence, limitations for developing SPR sensors. A spectral method for detecting SPR is used for such sensors. In [14], an issue of choosing the type of metal and wavelength in spectral sensors based on SPR was raised for the first time. Aluminum, cadmium, copper, indium, gold and silver were considered as possible materials. It was shown that the sensitivity of the SPR signal to changes in the thickness of the dielectric film on the metal surface at registration by the value of the minimum angle is the best for silver.

Later, a number of reviews considered physical limitations and technological methods for using the main types of high-conductive metals, such as gold, silver, copper and aluminum, in SPR devices. In [15], an overview of technological possibilities for creating ultra-thin films capable of generating surface plasmon-polaritons (SPP) in the infrared and visible spectral ranges was presented covering noble and other metals, conducting metal oxides and nitrides, highly doped semiconductors, graphene, *etc*. It was noted that for frequencies in the visible and near IR ranges, noble metals such as gold, silver and aluminum are the best candidates for plasmonics, since their plasma frequency is in the ultraviolet region, which ensures a negative real part of the permittivity. In [16], sensitivity of a specified series of noble metals as well as combinations of their layers to changes in the refraction characteristics of an external aqueous medium was assessed by numerical modeling. These calculations were based on analysis of spectral and angular characteristics of the so-called figure-of-merit (FOM), which is represented by a combination of several parameters of the plasmon resonance curve. The study of double metal layers is grounded by the need to use protective coatings for a number of metals. The authors also showed that use of titanium as a sublayer provided better response resolution as compared to chromium. The work [17] also presents an analysis of a number of plasmonic materials (noble and high-conductive metals, silicon carbide, doped semiconductors, metal nitrides, graphene, *etc*.), which takes into account the trade-off between such parameters as propagation length and degree of confinement of surface plasmon-polaritons. The analysis of these materials is based on a two-dimensional representation of two FOM, which are derivatives of the specified parameters. The proposed method made it possible to compare different plasmonic materials in the context of their specific applications. The authors demonstrate that silver has the best properties overall, having plasmons with the longest propagation length (except in the UV range, where aluminum has an advantage).

It should be noted, however, that interpretation of graphical data, obtained using the above-described methods, for evaluating the plasmonic characteristics of materials based on the introduced FOM parameters appears to be quite difficult and not always sufficiently clear. In this paper, we propose a new approach to analyze the conditions for realization of the spectral SPR effect in thin films of high-conductive metals, based on spectral analysis of such a physical parameter as the surface plasmon-polariton attenuation length [18] at the interface between a metal and a dielectric. To substantiate the relevance of considering this parameter in the context of the problem at hand, we demonstrate its correlation with the optical constants and the imaginary part of the metal permittivity, as well as a number of essential parameters of the SPR effect itself.

In our previous studies [19–21], we proposed a new version of an optoelectronic sensor for detecting gas molecules based on the spectral SPR effect in the chromatic mode with colorimetric registration of the R, G, B components of reflected light using a thin 40-nm silver film as a sensitive element in the Kretschmann geometry. We also evaluated the use of thin films of gold, silver, copper, aluminum, and a silver-gold bimetallic layer to implement the spectral SPR effect. For each of these metals, the spectral width of the SPR excitation region in the visible range was determined, and the sensitivity of the optical response to appearance of an external dielectric layer was assessed.

The aim of this work is to analyze the conditions and features of implementation of the spectral SPR effect in thin films of various high-conductive metals to create a chromatic SPR sensor of gas media as well as to consider an acceptable criterion for excitation of plasmon-polariton resonance in these metals in the visible light wavelength region based on an estimate of the attenuation length (or energy propagation length) of surface plasmon-polaritons in the metal.

2. Methodology

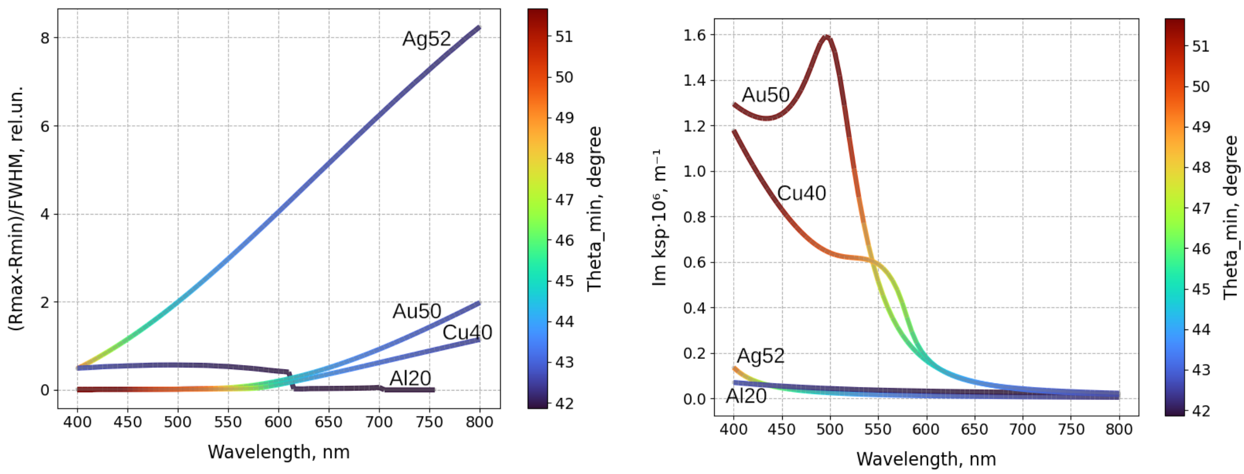
This paper examines thin films of gold, silver, copper, and aluminum deposited on the base surface of a glass prism in the Kretschmann geometry as plasmon-generating coatings. The wavelength range under consideration is limited to the visible range, in which chromatic SPR sensors of gas environments actually operate. Color webcams were used to record optical signals, followed by colorimetric analysis of the color pattern of the reflected and scattered beams under illumination by a white light source.

For calculating spectral functions of the reflection coefficient $R(\lambda,\theta)$ of the metal films in given regions of wavelengths and light incidence angles, as well as some plasmonic parameters, both traditional formulas for light reflection from a multilayer structure using Johnson matrix multiplication [22] and a freely distributed software WinSpall 3.02 [23] were used.

The values of the optical parameters for all the specified metals in the required wavelength range required for calculations were obtained by experimental measurements of film samples on glass/silicon substrates using a Semilab SE-2000 spectral ellipsometer. We recall that the complex permittivity of a metal is related to the metal optical constants n and k by the relation $\varepsilon_m = \varepsilon' + i\varepsilon'' = (n+ik)^2$. For these measurements, we had also previously determined the optimal thicknesses of the corresponding plasmon-generating coatings. It was achieved by model calculations of reflectance spectra upon excitation of SPR in the specified metals in the Kretschmann geometry for a 45-degree glass prism with a refractive index of 1.51. Based on the criteria of achieving the largest depth of minimum reflection and the smallest half-width of the resonance curves in the presence of an external gas environment and for visible light wavelengths of 400…700 nm, the optimal thicknesses were ultimately selected within the following limits: 45…50 nm for gold, 40…43 nm for copper, 50…56 nm for silver and 17…20 nm for aluminum. Note that optical elements with the listed metal layers on glass substrates with the thicknesses close to those indicated above were manufactured for experimental test measurements carried out in this work.

3. Approaches to criteria for realization of SPR in various metals

It is known that the spectral range for occurrence of the SPR effect in thin films of high-conductive metals is determined by the optical parameters of the metal used and the outer dielectric layer. A simplified condition for existence of plasmon-polariton resonance at a metal-dielectric interface is usually considered to be $\varepsilon'_m < -\varepsilon_d$ [24, 25], where $\varepsilon'_m < 0$ is the real part of the metal's permittivity and ε_d is the permittivity of the external medium. This condition works successfully for silver. It can also explain the spectral behavior of gold films in contact with an aqueous medium, which exhibit the SPR effect in the region above 550 nm [20, 21]. However, in other cases, particularly for copper and aluminum, this condition is not sufficient. According to the above condition, the SPR for aluminum should exist throughout the visible range in any environment, but in reality, this does not occur. The reason for such discrepancy is obviously that this approach neglects the imaginary part of the metal's permittivity, which defines the magnitude of plasmon attenuation in the metal.



(a) (b)

Fig. 1. Ratio of the reflection coefficient minimum depth to the half-width of the resonance curve Δ*R*/FWHM (a), and the imaginary part of a plasmon-polariton wave vector Im k_{sp} (b) as functions of the wavelength and the SPR minimum angle (the latter is shown in different colors according to the given color scale). The numbers on the graphs indicate the thicknesses of the corresponding metal films in nm.

Let us consider this assumption in more detail. To do this, we calculate the values of the following essential characteristics of SPR: the reflection coefficient *R* at the minimum and maximum, the full width at half maximum (FWHM) of the resonance curve, and the imaginary part of the surface plasmon-polariton wave vector k_{sp}, which depends on the values of the metal's optical parameters (for a given dielectric) for all the metals considered. For more information, we combine the first three values into a single general quality parameter in the form Δ*R*/FWHM, where $\Delta R = R_{max} - R_{min}$ is the depth of the resonance curve minimum. Since each metal is characterized by only a specific set of pairs of the *n* and *k* values, which depend on λ, it is more convenient to use the wavelength λ and the angle of light incidence θ as two universal variables in calculations. Fig. 1 shows 3-dimensional diagrams for the resonance curve quality parameter Δ*R*/FWHM and the imaginary part of the SPP wave vector Im k_{sp}. Note that in these diagrams, the minimum resonance angle θ_{min} is shown on the angle color scale (in fact, this is the third coordinate represented by the curve color).

In principle, the presented diagrams help us qualitatively separate those regions in the (λ, θ_{min}) scales where SPR is successfully excited (having sufficiently high Δ*R*/FWHM ratios greater than 0.5 on the one hand and low attenuation of SPP on the other hand) from the regions where the SPR is very weak or not excited at all. We can see from Fig. 1a that the resonance curves for silver have a high slope and a deep minimum in the entire visible spectrum range, somewhat broaden at decreasing the wavelength and shift on the angle of the resonance minimum from 42 to 47 degrees. For gold and copper, sufficiently high-quality resonance curves are realized only in the wavelength range above 600 nm. Below this wavelength, the quality parameter value is close to zero (this means that the resonance curve no longer exists). For aluminum, resonance curves of acceptable quality are obtained only in the short-wave region up to approximately 610 nm. Above this wavelength, the value of the quality parameter sharply decreases to zero, which also indicates disappearance of the resonance curve.

The above-mentioned features of the behavior of Δ*R*/FWHM clearly correlate with the behavior of the imaginary part of the surface plasmon-polariton wave vector (Fig. 1b). Indeed, a comparison of both parameters curves clearly shows that the minima of the resonance curves for Au and Cu disappear just when Im k_{sp} begins to increase sharply, which takes place close the above-mentioned wavelength of 600 nm. For silver, an almost perfect correlation of both parameters is observed throughout the entire visible spectrum. The high quality of the resonance curves corresponds to the minimum value of the imaginary part of k_{sp}. For aluminum, the aforementioned correlation of both characteristic parameters is well observed in the short-wavelength range up to ~ 610 nm. Only in the longer-wavelength region we note absence of such correlation for this material (however, this special case will be discussed below).

Recall that the imaginary part of the wave vector k_{sp} characterizes attenuation of surface plasmon-polaritons as they propagate from the point of generation. It follows from the established correlation between the parameters Δ*R*/FWHM and Im k_{sp} that high resonance characteristics of an SPR curve are directly related to low attenuation of the SPP, while weak resonance characteristics indicate relatively high attenuation of the SPP.

Therefore, the magnitude of the imaginary part of the wave vector of surface plasmon-polaritons can serve as a unique indicator of feasibility of the SPR effect in certain wavelength ranges and for certain incidence angles for different metals. On the other hand, we take into account that Im k_{sp} is nothing more than half of the inverse value of such a parameter as the attenuation length L_{sp} of the SPP in a metal. We conclude therefore that the assumption of the need to take into account the imaginary part of the metal permittivity is valid and allows us to propose a new approach to determining conditions for SPR excitation.

As a basis for calculations, we take the attenuation length L_{sp} (or energy propagation length) of the SPP in a metal. This length is the distance limiting the region of active localization of the plasmon-polariton wave in the metal, measured from the place of its excitation to the place where it attenuates by a factor of e. The expression for this length looks as follows [18, 26]:

$$L_{sp} = \frac{1}{2\,\mathrm{Im}k_{sp}}, \qquad (1)$$

where $\mathrm{Im}\,k_{sp}$ is the imaginary part of the wave vector k_{sp} of the SPP:

$$k_{sp}(\omega) = \frac{\omega}{c}\sqrt{\frac{\varepsilon_m(\omega)\varepsilon_d}{\varepsilon_m(\omega)+\varepsilon_d}}. \qquad (2)$$

By substituting $\varepsilon_m = \varepsilon' + i\varepsilon''$ and $\omega/c = 2\pi/\lambda$, we obtain the following expression for the wave vector:

$$k_{sp}(\lambda) = \frac{2\pi}{\lambda}\sqrt{\frac{\varepsilon_d\left(\varepsilon'^2 + \varepsilon'\varepsilon_d + \varepsilon''^2\right) + i\varepsilon''\varepsilon_d^2}{(\varepsilon' + \varepsilon_d)^2 + \varepsilon''^2}}. \qquad (3)$$

We write it in a simpler form: $k_{sp}(\lambda) = \frac{2\pi}{\lambda}\sqrt{a+ib}$. At $b << a$ (the validity of this condition was verified for all the considered cases), this expression can be transformed to an ordinary complex number:

$$k_{sp}(\lambda) = \frac{2\pi}{\lambda}\left(\sqrt{a} + i\frac{b}{2\sqrt{a}}\right).$$

Hence, the imaginary part of the wave vector is

$\mathrm{Im}k_{sp} = \frac{\pi}{\lambda}\frac{b}{\sqrt{a}}$, where

$$a = \frac{\varepsilon_d\left(\varepsilon'^2 + \varepsilon'\varepsilon_d + \varepsilon''^2\right)}{(\varepsilon' + \varepsilon_d)^2 + \varepsilon''^2}, \quad b = \frac{\varepsilon''\varepsilon_d^2}{(\varepsilon' + \varepsilon_d)^2 + \varepsilon''^2}. \qquad (4)$$

Substituting expressions (4) into (1), we obtain the plasmon-polariton attenuation length for given specific parameters λ, $\varepsilon'(\lambda)$, $\varepsilon''(\lambda)$ and ε_d. Calculated spectral dependences $L_{sp}(\lambda)$ for all the considered metals are shown in Fig. 2 for the values of the refractive index of the external environment $n_d = 1$, 1.5, and 1.8.

As can be seen from Fig. 2, the $L_{sp}(\lambda)$ values for Au and Cu films are close to zero in the entire short-wave part of the specified range. This indicates strong attenuation of SPP, or, in other words, absence of propagation of the plasmon-polariton wave energy along the surface. For silver, this parameter is also initially close to zero in the shortest wavelength region. However, it begins to increase sharply approaching $\lambda \approx 400$ nm and acquires very high values over a wide range of wavelengths. For aluminum, the L_{sp} value varies relatively little in the entire range under consideration, but, unlike the other metals, it does not tend to significantly decrease in the short-wavelength region. We will consider all these trends in subsequent analysis.

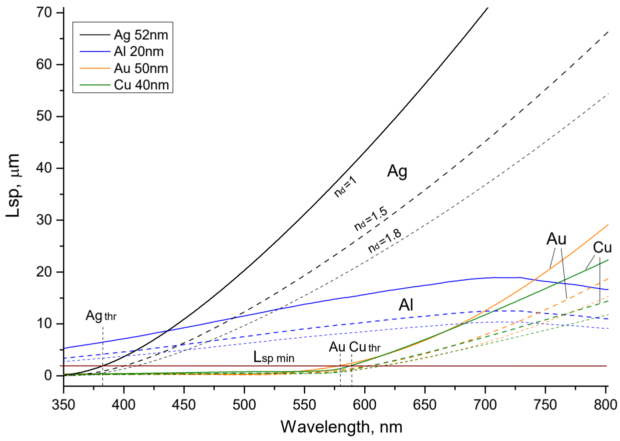


Fig. 2. Attenuation length L_{sp} in the wavelength range of 350…800 nm for the specified metals at $n_d = 1$ (solid curves), 1.5 (dashed curves) and 1.8 (dotted curves). The horizontal line $L_{sp\,min}$ indicates its minimum possible value for excitation of plasmon-polariton resonance in metal films. The vertical dotted lines show the corresponding threshold wavelengths, below which SPR is not excited, in silver, gold and copper, respectively.

We estimate the minimum possible limiting value of L_{sp} for implementation of plasmon-polariton resonance in metal films. To do this, using a model calculation of the spectral functions of reflection, we first determine the wavelengths below which the minimum of a plasmon resonance curve begins to disappear (the value of the quality parameter ΔR/FWHM becomes less than 0.2): ~ 380 nm for silver, ~ 560..580 nm for gold, and ~ 590..600 nm for copper. Note that these data are in good agreement with our previous theoretical and experimental results obtained in [20, 21]. We then draw a straight line along the horizontal axis (see Fig. 2) so that it intersects the $L_{sp}(\lambda)$ curve for silver (at $n_d = 1$) at the above-mentioned level $\lambda_{thr} = 380$ nm. We see that this line also intersects the curves for gold and copper in the range of 580…590 nm, which is consistent with the wavelength thresholds given above. As a result, we reasonably obtain a value of $L_{sp\,min} \cong 2$ µm. Note that this value corresponds to the $\mathrm{Im}\,k_{sp}$ value of approximately $0.25 \cdot 10^6$ m^{-1} (see Fig. 1b).

Note that the above estimate was made for $n_d = 1$, *i.e.* when the external medium is air or a highly diluted gas. The thickness of the outer layer is not included in the expression (1), which is true provided the outer layer is sufficiently thick compared to the metal film. However, if the medium becomes denser, *e.g.* with the refractive index in the range $n_d = 1.5…1.8$, the value of L_{sp} decreases by approximately the same factor (see Fig. 2). In this case, the threshold wavelengths λ_{thr} for Ag, Au, and Cu indicated by the vertical dotted lines in Fig. 2 slightly shift to the right. Namely, as n_d increases from 1 to 1.8, the threshold for silver shifts from 383 to 410 nm, and for gold and copper from 580 and 590 to 615 nm, respectively. The small magnitude of this shift indicates that presence of an external environment, even with significant density, has almost no effect on initial feasibility of SPR in certain regions of the spectrum for the metals under consideration, only slightly (by 3-5%) shifting their SPR excitation thresholds toward longer wavelengths.

On the other hand, assessment of the sensitivity of the attenuation length to a change in the media refractive index by $\Delta n = 0.001…0.01$ (*e.g.*, under exposure to gases) shows that it is very small for the considered metals: the value of $\Delta L_{sp}/L_{sp}$ reaches no more than 1.5% on average. As expected, it is somewhat higher in the short-wavelength part of the spectrum for all the metals except aluminum. For aluminum, this sensitivity value does not change at all across the entire spectrum. These data indicate that the environment will have practically no effect on the performance of a chromatic SPR sensor in gas sensor measurements.

The data presented above show that spectral behavior of the attenuation length could largely clarify the reasons for occurrence or absence of the observed SPR effect in films of the metals under consideration. For example, for silver, the presented dependences indicate that SPR can be fully excited in the entire visible wavelength range from blue to red. The $L_{sp}(\lambda)$ curves for gold and copper indicate that these metals allow SPR to be excited only in the long-wavelength region of the visible spectrum above 550 nm. For aluminum, the L_{sp} value varies from 7 to 17 µm, theoretically ensuring the possibility of SPR excitation in the entire visible and even IR range. However, this is not true in practice, and more data are needed to clarify this situation, as discussed below.

It should be recognized that the observed behavior of L_{sp} is predetermined by the initial spectral dependences of the optical parameters of metals $n(\lambda)$ and $k(\lambda)$. However, the refractive index $n(\lambda)$ still exerts a predominant influence. This assumption can be substantiated as follows. When calculating L_{sp}, the numerator b for the imaginary part of the wave vector $b/2a^{1/2}$ in the expression (4) is determined by the value of ε'' (the remaining terms are close to unity and have a very small effect). In turn, $\varepsilon'' = 2nk$, *i.e.* it is determined by the product of both optical constants. The trend of the extinction coefficient k is approximately the same for all metals (Fig. 3a) and has no distinctive features. However, the value of n in the short-wavelength region has its own peculiarities (Fig. 3b). It is small for aluminum and negligible for silver. At this, copper and gold have significantly higher values in this region. Therefore, it is obvious that small n values play a decisive role on a possibility of SPR excitation in the short-wavelength region for Ag and Al and do not enable it for Au and Cu. At the same time, the values of n and ε'' for aluminum (Figs 3b and 3c) begin to increase sharply in the middle and long-wavelength regions. This is definitely the key factor that makes it impossible to excite SPR in aluminum in these regions. Indeed, as shown by model calculations and confirmed by actual experiments, the upper limit of SPR existence for aluminum is approximately 550 nm for 17…20 nm thick layers and 650…700 nm for very thin layers of the order of 9-10 nm.

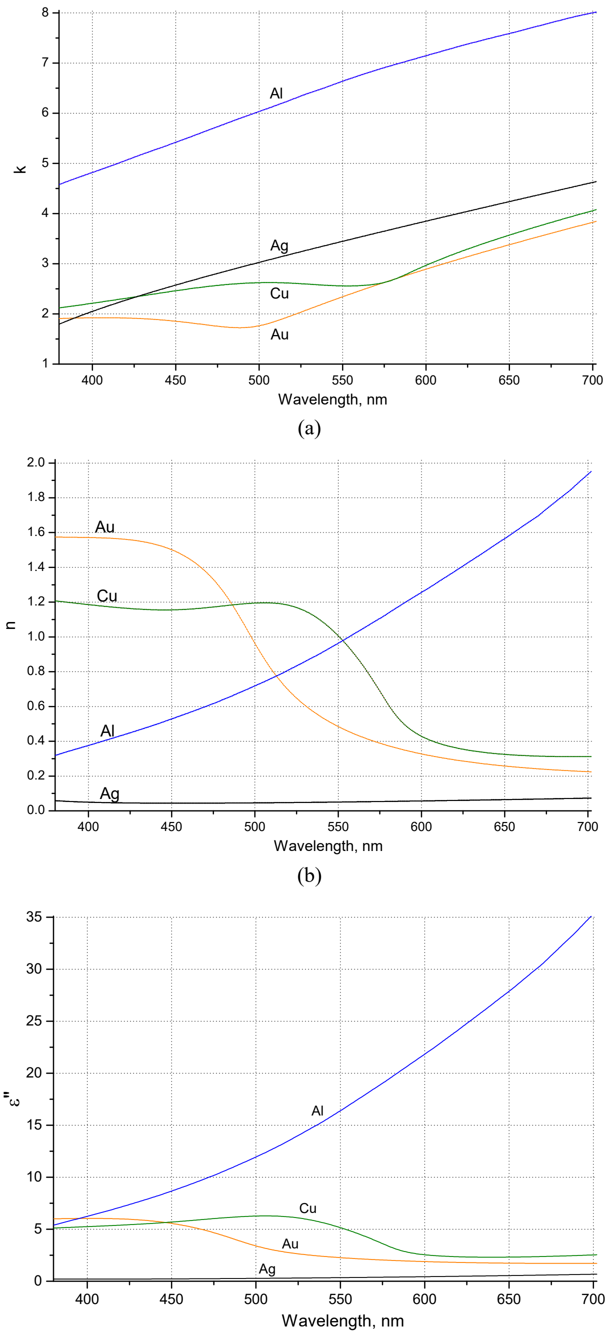


Fig. 3. Spectral dependences of the refractive index n (a), extinction coefficient k (b), and imaginary part of the permittivity ε''_m (c) for Al, Au, Ag and Cu films.

At the same time, no lower limit of SPR existence, at least down to 300 nm, is observed for this material.

When studying specific features of SPR implementation in different high-conductive metals, the effective depth of localization of the surface plasmon-polariton field within the metal film, as well as the penetration length of the electric field of a SPP wave outward into the external environment, are also relevant parameters to consider. The expressions for describing these parameters are known [25, 27]:

$$D_m = \frac{\lambda}{2\pi}\sqrt{-\frac{\varepsilon'_m + \varepsilon_d}{\varepsilon'^2_m}}, \; D_d = \frac{\lambda}{2\pi}\sqrt{-\frac{\varepsilon'_m + \varepsilon_d}{\varepsilon^2_d}} . \qquad (5)$$

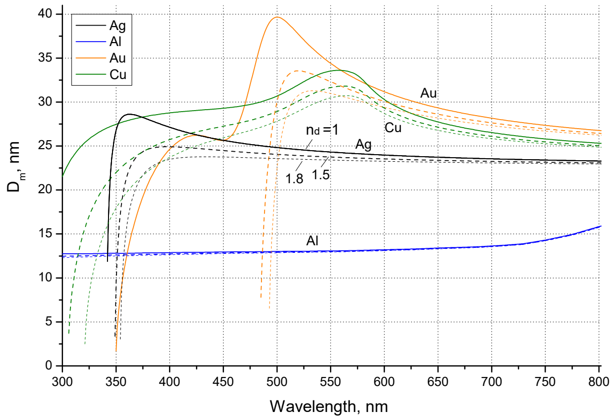
Here, D_m and D_d are the distances from the interface in a direction perpendicular to the metal and dielectric, respectively, at which the electric field intensity decreases by a factor of *e*. Fig. 4 shows the calculated values of these both parameters in the wavelength range of interest.

The unusual behavior of the D_m curves for all the metals (except Al) in the wavelength region below 400 nm is noteworthy. Here, the localization depth of the surface plasmon-polariton field in the metal rapidly approaches zero, indicating a sharp weakening of plasmon generation. When compared with the near-zero value of L_{sp} in the same region (see Fig. 2), these two factors together indicate the impossibility for these metals of both generation and propagation of a SPP wave in this range. An exception is aluminum, where SPR is successfully excited in this region, and the depth of electric field localization in the metal here, as in the rest of the spectral range, is approximately 13 nm, which is approximately half of that for all the other considered metals. Since plasmon-polaritons are excited at the interface between the metal and the external dielectric, and sufficient amount of the incident light energy must reach this interface, the thickness of the aluminum layer must be approximately half of that for the other metals to excite SPR.

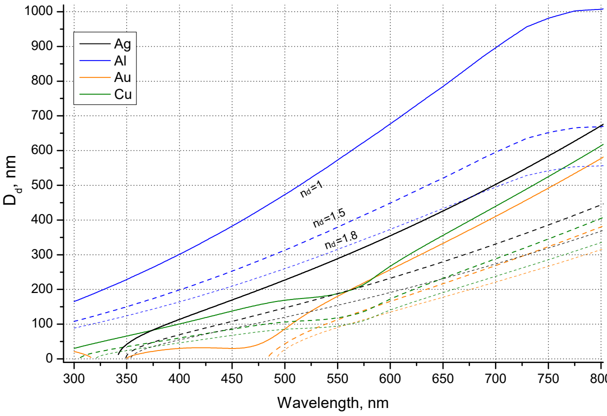
As can be seen from Fig. 4a, influence of the external environment with n_d varying from 1 to 1.8 significantly affects the dependence $D_m(\lambda)$ only for gold, shifting the plasmon generation threshold to longer wavelengths. However, as we know, SPR in gold is excited at wavelengths above 500 nm. Therefore, we can confirm the previously made conclusion that, from a sensory point of view, influence of the environment on operation of a chromatic SPR gas sensor for all the considered metals may be neglected.

As for the penetration length of the field D_d into the outer layer (essentially, this is an evanescent wave), this value characterizes the sensory capabilities of plasmon-generating coatings to sense molecules at a certain distance from the surface of the metal film (see Fig. 4b). It is evident that in this regard, silver significantly exceeds the capabilities of gold and copper over a wide spectral range. However, aluminum is the most sensitive material, exceeding other metals by more than 2-3 times. This demonstrates the potential for aluminum to be highly suitable for solving sensory tasks in the short-wavelength range.

The influence of the external environment with an increase in its refractive index naturally leads to a decrease in the evanescent wave region by a number of times approximately equal to the refractive index of the medium n_d. The peculiarities of such influence are again manifested only for gold: the sensitivity to external molecules in presence of a medium with $n_d \geq 1.5$ is possible only at wavelengths above 500 nm.



(a)



(b)

Fig. 4. Characteristic depth of localization of the electric field of a plasmon-polariton wave inside a metal film D_m (a), and in an external medium D_d (b) for a dielectric medium with refractive indices $n_d = 1$ (solid curves), 1.5 (dashed) and 1.8 (dotted).

4. Experimental patterns of scattered radiation upon SPR excitation in various metals

This section is devoted to a visual demonstration of the results presented in the previous sections, which examined the conditions for excitation of the SPR effect in the chromatic regime for the specified series of metals in contact with a gaseous environment. This demonstration can be accomplished through direct experimental measurements of the spectra of plasmon-scattered light observed from an outer surface of the metal layer upon excitation of SPR in films of the metals under consideration.

First, we briefly consider the relationship between the SPR effect and the appearance of scattered radiation. Absorption of light energy of a certain wavelength incident at a certain angle, by a metal film induces resonant oscillations of the electron gas and leads to generation of a surface plasmon-polariton wave. However, this process is directly related to the decay of surface plasmons into photons of the same frequency due to scattering by surface and bulk inhomogeneities within the metal. Therefore, emission of the scattered light as well as quenching of the specularly reflected beam during SPR excitation result from the resonance of surface currents

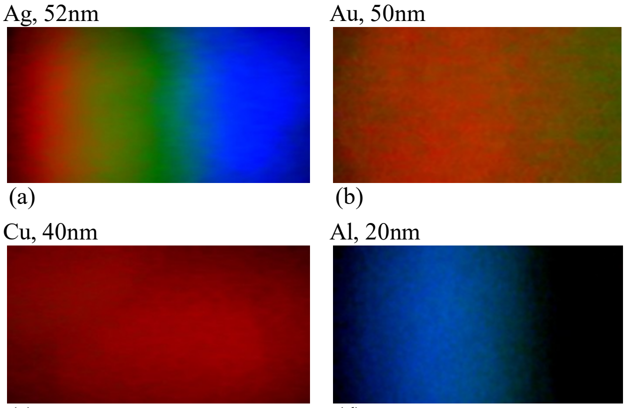


Fig. 5. Experimental spectral patterns of plasmon-generated scattered light from silver (a), gold (b), copper (c), and aluminum (d) films with indicated thicknesses. The range of light incidence angles is nearly from 42 to 46 degrees (from left to right along the spectral pattern).

at the metal-dielectric interface. It should be emphasized that the scattered light is not the light excited by plasmon-polaritons. This emission is caused by an additional mechanism of light re-emission from a rough metal surface, which functions as a diffraction grating. Although the re-emitted light obeys the law of interference amplification of radiation from neighboring protrusions, one may state that it has the same spectral composition as the original SPR excitation light.

Fig. 5 shows patterns of real experimental spectra of plasmon-photon radiation recorded from the front surface of metal films in the Kretschmann geometry using a Logitech C525 webcam under illumination by a white LED light source (Cree XTE Star 1-5W White). The horizontal rotation of the images corresponds to a range of light incidence angles of about 42..46 degrees.

As we can see, the color gamut of these scattering spectra quite accurately reflects the frequency ranges of SPR excitation in the films of the aforementioned metals, which were determined by analyzing the attenuation length of SPP (see Fig. 2). Namely, as found above, the SPR excitation range for silver begins at approximately 380 nm and covers the entire visible region up to 700 nm and more (Fig. 5a). The spectral region of SPR excitation for gold is narrower as compared to silver, limited to the red and partially green wavelength range (Fig. 5b). For copper, the region of excitation of the SPR is even narrower, being located only in the long-wave red part of the spectrum (Fig. 5c). In its turn, the range of SPR effect implementation for aluminum includes the entire blue region (Fig. 5d).

It should be noted that the above color patterns can be influenced by spectral characteristics of the radiation source and photodetectors used in the webcam [28]. In this study, in addition to the devices mentioned above, two more types of color webcams and two types of white light sources were tested. The obtained results showed that despite some differences in details, the overall qualitative pictures of the plasmon-scattered spectrum did not change.

The described features are important for creating gaseous medium sensors based on the spectral SPR effect with recording of optical response signals in the form of both polychromatic spectra and individual monochromatic bands of scattered and/or reflected radiation.

5. Conclusions

The conditions for realization of the spectral SPR effect in thin films of various high-conductive metals (gold, silver, copper, and aluminum) are considered. It is shown that the well-known condition $\varepsilon'_m < -\varepsilon_d$, although necessary, is not always sufficient for existence of plasmon-polariton resonance at the metal-dielectric interface. As an addition, it is proposed to consider the attenuation length (or energy propagation length) of surface plasmon-polaritons in the metal. It is shown that analysis of the spectral dependences of the attenuation length enables reliable determination of the spectral regions where SPR in the aforementioned metals can be excited. Moreover, it was found that to excite the SPR, the attenuation length in metal films must exceed 2 µm. This evaluation criterion works well for Ag, Au, and Cu. For aluminum, the features of the spectral dependence of the imaginary part of permittivity are additionally used in addition to this criterion.

The study of the spectral characteristics of the depth of localization of electric field of surface plasmon-polaritons inside the metal film as well as the length of field penetration outward into the dielectric confirms the conclusions made above. On the other hand, it shows that for all the considered metals, presence of a gaseous medium with a refractive index of up to 1.8 has practically no effect on the operating settings of chromatic SPR sensors. It is shown that aluminum is the most sensitive material with more than 2-3 times superiority in the evanescent wave region over other metals, which indicates its high potential for use in optical sensor elements for the short-wave region.

The results of the presented theoretical analysis, disclosing the conditions for implementation of the SPR effect in the chromatic regime for the specified metals, are confirmed by a visual demonstration of experimentally obtained spectral patterns of plasmon-scattered light in the visible wavelength range.

Acknowledgments

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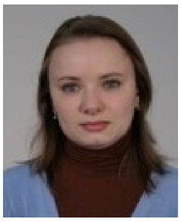

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Особливості реалізації спектрального ефекту ППР у тонких плівках високопровідних металів для створення ППР сенсорів газового середовища

А.І. Білецький, О.Л. Кукла, Ю.М. Ширшов, О.С. Кондратенко, О.М. Федченко

Анотація. У роботі розглянуто умови збудження поверхневого плазмон-поляритонного резонансу (ППР) у тонких плівках ряду високопровідних металів (золото, срібло, мідь та алюміній) з метою використання особливостей спектрального ефекту ППР для створення хроматичних сенсорів газового середовища. Запропоновано новий підхід для оцінки можливості реалізації ППР у плівках зазначених металів у різних областях видимого діапазону спектра, що базується на аналізі спектральних характеристик довжини загасання поверхневого плазмон-поляритону на межі поділу метал/діелектрик. Розглянуто зв'язок довжини загасання з оптичними постійними та уявною частиною діелектричної проникності металів, а також рядом суттєвих параметрів власне ефекту ППР, та продемонстровано їх певну кореляцію. Установлено, що для збудження ППР довжина загасання поверхневого плазмон-поляритону у плівках металів має перевищувати 2 мкм. Результати представленого теоретичного аналізу зіставлені з експериментально отриманими спектрами плазмон-розсіяного світла в діапазоні довжин хвиль видимого світла.

Ключові слова: поверхневий плазмон-поляритонний резонанс, високопровідні метали, плазмон-поляритон, довжина загасання, хвильовий вектор, спектральний ефект ППР.