TO THE VELOCITY OF LIGHT PROPAGATION IN METALS

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Experimentally measured angular dependences of the internal and external reflections of polarized radiation from a metal (gold) film on the surface of a glass semicylinder are used to calculate the refractive indices of both the semicylinder material and the metal. The results of measurements of total internal reflection are used to determine the losses of light intensity at the semicylinder surfaces and in the semicylinder material bulk. The direct and inverse photometry problems have been solved for light incident onto the gold film from both the air and the glass side. A good agreement between theoretical results and experimental data has been obtained. The issue of determination of the critical angle of the attenuated total internal reflection and the angular position of the plasmon resonance has been examined. On the basis of the results of our experimental studies, a positive answer to the fundamental issue – the validity of the relativity postulate about the finiteness of the velocity of light propagation in media – has been obtained.

1. Introduction

Fresnel derived his formulas in rather a specific way as long ago as in 1823. After the Maxwell theory appeared, these formulas were obtained following a logical procedure, namely, by solving the system of relevant equations. The further improvement of the experimental technique and the methods of studying the absorbing media invoked an issue concerning whether the available formulas are valid for application.

If the dielectric permittivity becomes complex-valued, the refractive index $\tilde{n} = n - ik$ becomes complex as well. Its real part n is a ratio between the phase velocities of light propagation in vacuum, c, and in a given medium, θ, i.e. $n = c/\theta$. The imaginary part k is associated with the absorption coefficient and, according to the Bouguer–Lambert law, is determined by the relation $\alpha = 4\pi k/\lambda$, where λ is the light wavelength. Hence, the former quantity should be responsible for light refraction, whereas the latter one for light absorption. However, the absorption factor k can also affect the refracted beam path [1].

When studying the dependence of the refractive index of a medium on the medium thickness, almost all experimenters obtained curves that were identical by their shape for different metals and semiconductors [2–5]. When the film thickness diminished to certain minimal values, the magnitude of the refractive index drastically grew, although it would have tended to unity, the refractive index of air (vacuum). This problem was put forward even in the encyclopaedia "Solid State Physics" [6]. The solution was obtained only in work [7]. It was found that a correct choice of the root pair n and k would ensure that the refractive index would approach unity, if the medium thickness diminished to a minimally possible value.

A different situation is observed for precious metals and some other substances. Not aiming at doing a historical review with a purpose to establish who has calculated the optical constants n and k first, we would like to mention Drude's works, which left a deep trace in the history of physical optics, in particular, metal optics. At the end of the nineteenth century, he obtained numerical values for the real and imaginary parts of the refractive index of some metals at $\lambda = 589$ nm. It turned out that, for a number of metals, the n-values were less than unity at this wavelength, and sometimes this effect was rather pronounced. The corresponding n-values are listed in the Table.

Experimenters obtained similar values for n and k in various spectral ranges. For instance, the values of n for Cu, Ag, and Au obtained in work [8] are less than unity in a wide spectral interval. Drude characterized such a result as "interesting": "Such small values of the refractive indices for silver, gold, copper, and, especially, alkaline metals have to draw your attention. They indicate that light propagates much more quickly in those metals than in a void". T.P Kravets, who edited

Optical constants of some metals at $\lambda = 589$ nm

Metal	n	k
Silver	0.18	3.67
Gold	0.37	2.82
Copper	0.64	2.62
Sodium	0.044	2.42
Potassium	0.048–0.068	1.5–1.86
Rubidium	0.131	–
Cesium	0.321	1.2

the Russian edition of "Optics" by P. Drude, marked in this occasion: "According to the principle of relativity, a motion with a velocity that exceeds c is impossible. However, the availability of refractive indices smaller than unity does not contradict it: here, we are dealing with the velocity of phase propagation at a stationary motion, but the velocity of a signal does not exceed c". T.P Kravets noticed that all that is valid for a strictly periodic, i.e. monochromatic, wave, which is not confined in time. "An issue on the propagation of a time-confined wave train in a dispersion medium has been considered in detail by Sommerfeld. Such a light signal, while propagating, deforms continuously. Its front runs forward at a velocity that is close to that of light in vacuum, whereas the propagation velocity of the other part of signal is close to the group velocity. Meanwhile, the concept of the latter becomes complicated very much in the anomalous dispersion range" (T.P Kravets's comment on p. 314 in Drude's monography).

M. Born and E. Wolf indicated in their monography [9] that the velocity becomes complex-valued and loses its physical sense in the case of absorbing media. This issue has not been elucidated in monography [10, p. 61] by A.V. Sokolov, where it is asserted that, in the case of p-polarized wave, the energy flows in a different direction, which does not coincide with the direction of phase propagation. It is of interest how those two directions converge into a single one, when the wave transmits from metal into insulator? Nevertheless, the key issue is whether or not c exceeds the velocity of signal propagation at $n < 1$? This question could be answered – and, by the way, the versions of the refraction law examined in work [1] could be verified – if one could monitor the variation of the propagation direction of a light beam, when the latter passes through a metal prism ($n > 1$, if the beam deviates toward the prism basis, and $n < 1$, if to the prism vertex). Unfortunately, the refractive index is slightly different from unity in the x-ray range, where absorption is low. At the same time, the best metal prisms would induce deviations of a few hundredths of seconds for a light beam in the visible spectral range. Even the application of modern technique gives anybody no chances to carry out such experiments successfully.

By applying Sokolov's formula [10, p. 52]

$$\theta = \frac{c\sqrt{2}}{\sqrt{n^2 - k^2 + \sin^2\varphi + \sqrt{\left(n^2 - k^2 - \sin^2\varphi\right)^2 - 4n^2k^2}}}, \quad (1)$$

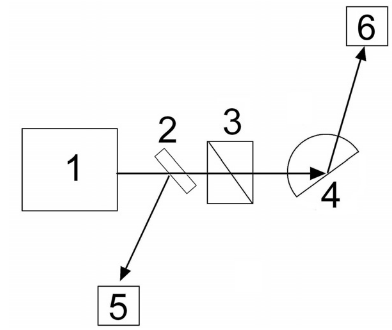

Fig. 1. Scheme of experimental installation: light source (*1*), glass plate (*2*), polarizer (*3*), glass semicylinder (*4*), reference photodetector (*5*), photodetector of reflected light (*6*)

let us determine the light velocity, provided that $n = 0.18$ and $k = 3.64$ (these are the optical constants for gold at the wavelength $\lambda = 6328$ Å). We obtain $c_1 = 16.0 \times 10^8$ m/s for the incidence angle $\theta_1 = 0°$ and $c_2 = 16.7 \times 10^8$ m/s for $\theta_2 = 89°$. Those values are more than five times larger than the velocity of light propagation in vacuum. Therefore, this is a matter to be thought about.

The aim of this work is to find experimentally which of two inequalities, $\theta > c$ or $\theta < c$, is valid for a gold film.

2. Experimental Part

Our researches consisted in measuring the reflection coefficients for light polarized either in the incidence plane or normally to it. The scheme of installation is shown in Fig. 1.

As a light source, we used a helium-neon laser with a wavelength of 632.8 nm. A glass plate split the beam into two ones: a reference one, the intensity of which was accepted as a base, and a beam that stroke the specimen. The intensity was measured with the help of silicon light-emitting diodes put into spheres. The polarization plane was determined by a polaroid which provided a degree of light polarization of 99%. The error of polarization azimuth measurements did not exceed 0.1°. The angle of incidence was determined by means of a goniometer with an accuracy not worse that 0.017°.

The subjects of inquiry were gold films deposited onto a glass semicylinder 110 mm in diameter and 20 mm in height. A large curvature radius of the cylinder provided the minimal dispersion of the beams and a high accuracy of measurements.

Gold films were deposited onto a plane part of the semicylinder by the method of thermal sputtering in

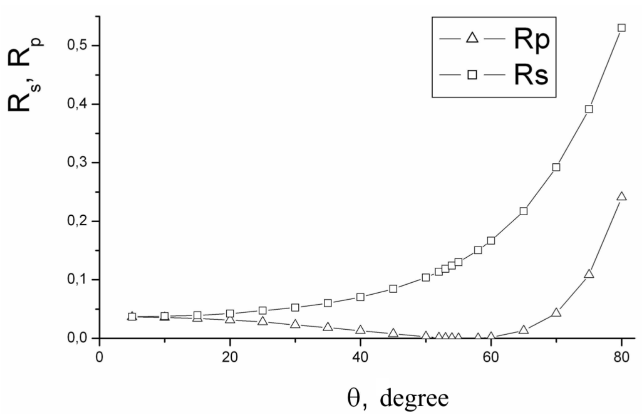

Fig. 2. Angular dependences of the reflection coefficients for s- and p-polarized light incident onto glass from air. Points are experimental values, solid curves are fittings

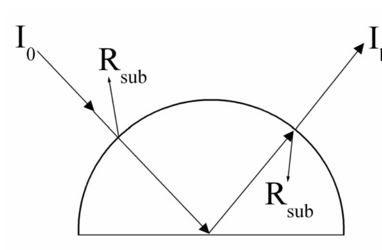

Fig. 3. Beam path through the semicylinder

vacuum. The semicylinder was maintained at room temperature. The residual pressure of atmospheric gases was about 10^{-6} mmHg. The thickness of the gold film was determined with the help of a quartz resonator.

To take the losses of intensity – when light passes through the semicylinder – into account, we measured first the coefficients of light reflection from the plane surface of the semicylinder for two polarizations (Fig. 2). Those data were used to determine the refraction coefficient of glass; we obtained a value of about 1.474. Making allowance for the character of a beam path (Fig. 3), we calculated the correction to light intensity losses by the formula

$$R = I_b/I_0, \tag{2}$$

where I_0 and I_b are the intensities of the incident and output beams, respectively.

If needed, the phenomenon of total internal reflection can be applied to determine the absorption coefficient of glass by the formula

$$\alpha = \frac{2\ln(1-R_{\rm sub}) - \ln R}{d} = \frac{1}{d}\ln\frac{(1-R_{\rm sub})^2}{R}, \tag{3}$$

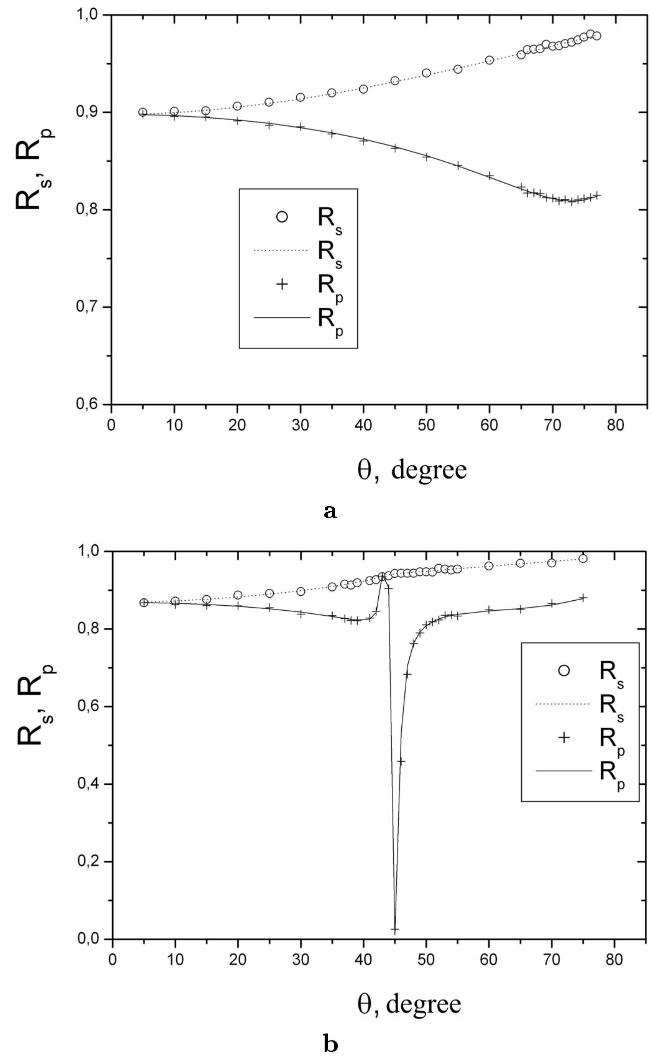

Fig. 4. Angular dependences of the reflection coefficient for two light polarizations. Reflection from a gold film ($n = 0.17$, $k = 3.65$, $d = 45.9$ nm) sputtered onto a glass substrate ($n_{\rm sub} = 1.474$). Light falls from the film (a) and the glass (b) side. Points denote experimental data, solid curves are the results of calculations

where $R_{\rm sub} = \left(\frac{n-1}{n+1}\right)^2$ is the reflection coefficient from the glass–air interface, n is the refractive index of glass, and d is the cylinder diameter. For the semicylinder used by us, $\alpha = 2$ m^{-1}. It means that the intensity of light became e times lower at a distance of 0.5 m in the glass layer, which evidences for a low quality of the applied material. However, in no way did this fact affect the results of our researches.

The optical constants of gold films were determined by analyzing the angular dependences of light reflection coefficients from the film side – they are given in Fig. 4,a

– with the use of the formulas from work [11]. For the known optical constants of a specific gold film, one can calculate the angular dependences of R_s and R_p in the case where light falls from the glass side. The results of our calculations, as well as the data of experimental measurements for a film 45.9 nm in thickness are exhibited in Fig. 4,*b*. A good agreement between theoretical and experimental results testifies that the direct and inverse photometric problems were solved correctly. It also means that the correction on light reflection from the glass surface and on absorption in the glass layer was made properly.

3. Results and Their Discussion

Let us recall that the phenomenon of total reflection occurs at the interface between two media (1 and 2), provided that light falls at an angle that is larger than the critical value and the optical density of the other medium (medium 2) is lower, i.e. if

$$n_1 > n_2. \tag{4}$$

If the absorption coefficient of the studied film $k \neq 0$, no total reflection takes place, but light also does not propagate in medium 2. This case is coined as attenuated total reflection. On the plot, the critical angle can be determined by the maximal slope in the angular dependence of R_p (see Fig. 5).

In work [12], the formula $\sin\theta = n/n_s$ was suggested to calculate the critical angle. Here, n is the real part of the complex refractive index of a film, and n_s is the refractive index of the output medium. In such a case, when light passes through the glass–gold–air system, no total reflection is possible at the glass–gold interface ($\sin\theta > 1$), whereas the angle would be 11.5° at the gold–air interface. However, our experimental result contradicts this conclusion. This contradiction can be resolved, if we consider the Descartes–Snell law

$$n_1 \sin\alpha = \tilde{n}_2 \sin\beta = n_3 \sin\gamma, \tag{5}$$

where n_1, $\tilde{n}_2$, and n_3 are the refractive indices of the media; α and β are the angles of incidence at the medium interface; and γ is the angle of refraction at the gold–air interface. In the case where there is no total reflection at the interface between the first and the second medium, the last equality gives rise to

$$n_1 \sin\alpha = n_3 \sin\gamma. \tag{6}$$

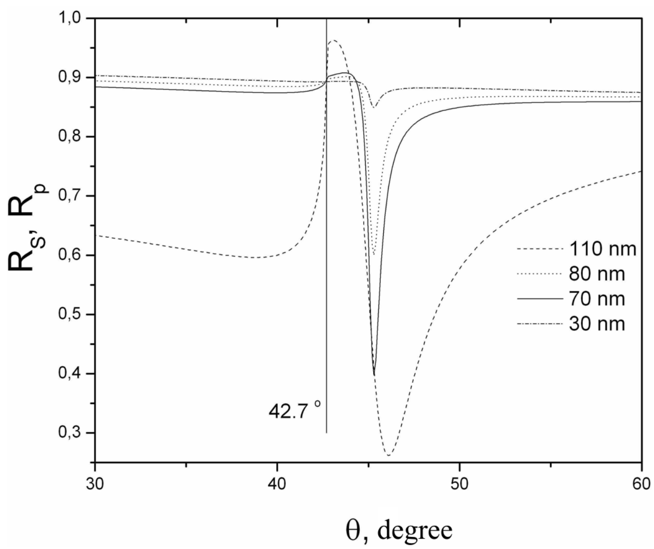

Fig. 5. Angular dependences of the reflection coefficient for p-polarized light incident on a gold film ($n = 0.17$, $k = 3.65$) from glass ($n = 1.474$) for various film thicknesses

The condition of total reflection, when the beam slides along the interface, and $\sin\gamma = 1$, brings about

$$\alpha = \arcsin(n_1/n_3), \tag{7}$$

where n_3 is the refractive index of air, and n_1 the refractive index of glass. Hence, the critical angle depends on the refractive indices of environments rather than on the optical constants of the film. Calculations by formula (7) give the value $\alpha \approx 42.7°$, which agrees with experimental data. The maxima near the critical angle in the curves depicted in Fig. 5 are induced by attenuated total reflection. The minima at about 44°, which follow them, are owing to the surface plasma resonance, when the incident wave energy transforms into the energy of surface plasmons.

The increase of the film thickness is accompanied by a decrease of the plasmon energy. It is confirmed by the gradual disappearance of plasmon-induced minima. Hence, surface plasmons are formed at the gold–air interface, and the increase of the film thickness leads to a reduction of the intensity of light that reaches this interface.

As the film thickness grew, sharp maxima in the vicinity of the critical angle got smoothed out and ultimately disappeared completely. It would not have happened, if total reflection had occured at the glass–gold interface. Hence, it occured at the gold–air interface. Quite logical seems the explanation that, with increase in the gold layer thickness, a smaller and smaller part of the energy reached the latter interface and a half larger part returned back to the photodetector. One more explanation follows from formulas (5) and (6). Since the critical angle equaled 42.7° (this value is a result of formula (6)), then, according to formula (5), there was no total reflection at the glass–gold interface. Hence, inequality (4) was fulfilled, i.e. gold is optically denser than glass. Respectively, the velocity of light propagation in gold is lower than that in glass, and the more so in air or vacuum. Therefore, $\theta < c$, and the postulate of the relativity theory is not violated.

4. Conclusions

When changing over from transparent to absorbing media, the refractive index of the medium becomes complex-valued, as well as the angle of refraction and the velocity of light propagation. It was shown in work [1] that the formula used for the calculation of the real part of the complex angle of refraction and the formula obtained in monography [10] for the angle of refraction as an angle between the plane of equal phases and the plane of equal amplitudes brought about different results. Moreover, the beam refraction was demonstrated to depend on both the refractive index and the absorption coefficient of the medium [1]. Formula (1) gives the value for the velocity of light propagation in gold that considerably exceeds the velocity of light in vacuum, which contradicts the results of this work. At the same time, the Fresnel formulas for absorbing media, which include the known optical constants of media, are the exact solution of the direct photometry problem – this fact is evidenced for by the results exhibited in Fig. 4. Those formulas allow the position of the surface plasmon resonance to be calculated exactly in terms of the angular dependences of the reflection coefficient for p-polarized light. All the abovementioned testifies that the optical constants n and k are only auxiliary quantities which allow the results of the interaction between light and an absorbing substance to be reproduced, but they have no precise physical meaning. At last, here is the answer to the main question of this work: the velocity of light propagation in gold does not exceed the velocity of light propagation in vacuum, so that the postulate of the relativity theory is not violated.

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ДО ПИТАННЯ ПРО ШВИДКІСТЬ ПОШИРЕННЯ СВІТЛА В МЕТАЛАХ

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Р е з ю м е

За виміряними кутовими залежностями коефіцієнтів зовнішнього та внутрішнього відбивання поляризованого випромінювання від металевої (золото) плівки на поверхні скляного напівциліндра обчислено показник заломлення матеріалу напівциліндра та металу. Визначено втрати інтенсивності світла на поверхнях напівциліндра та в товщині шару з результатів вимірювання повного внутрішнього відбивання. Розв'язано пряму і обернену задачу фотометрії під час падіння світла на золоту плівку як з боку повітря, так і з боку скла, і одержано добре узгодження розрахунків з експериментальними даними. Розглянуто питання визначення кута затухаючого повного внутрішнього відбивання та кутового положення плазмонного резонансу. За допомогою відзначених експериментальних досліджень отримано відповідь на фундаментальне питання – виконання постулату теорії відносності про граничність швидкості поширення світла в середовищах.