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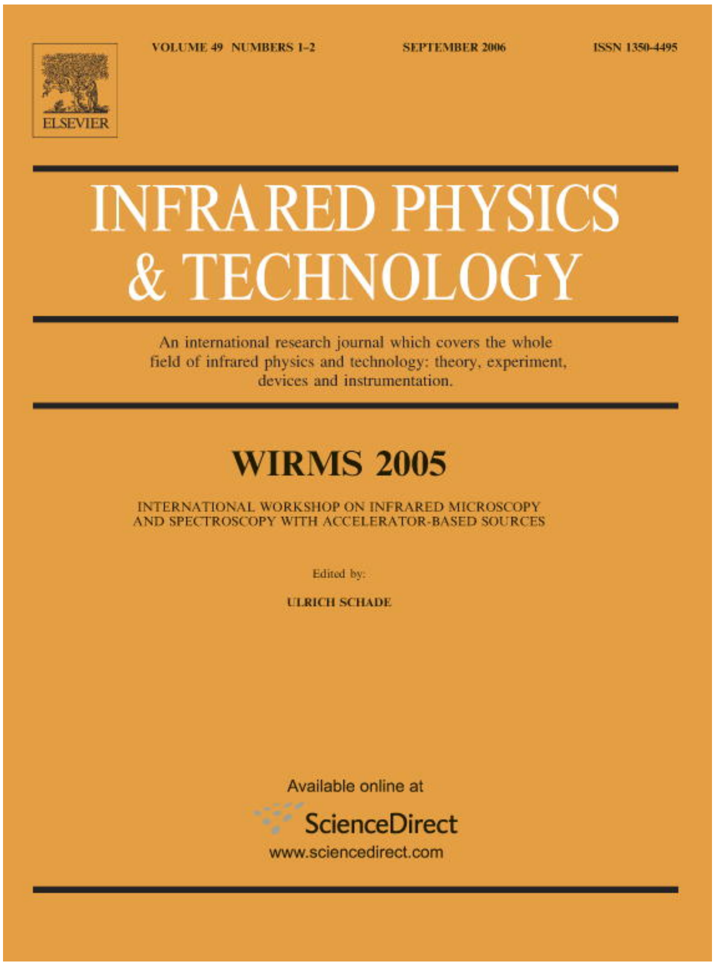

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Absorption of surface plasmons in “metal–cladding layer–air” structure at terahertz frequencies

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Abstract

Absorption of surface plasmons (SP) guided by metal surface covered with a transparent layer (“cladding layer”) at terahertz frequencies has been studied both experimentally and by computer simulations. It was found that presence of the layer may increase SP absorption by about four orders of magnitude as compared with the ideal metal surface. Experiments performed with aluminum and copper metal specimens using free-electron laser radiation (90 cm^{-1}) proved that SP absorption increases with the layer formation. But this increase turned out to be not as large as predicted and SP propagation length along the uncovered metal surface was at least one order of magnitude smaller than it should be according to Drude model.

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Keywords: Surface plasmons; Terahertz spectral range; Free-electron lasers; Laser spectroscopy

1. Introduction

Surface plasmons (SP) are used in surface science of metals, bringing good results in thin film spectroscopy in the visible and middle infrared spectral ranges [1–3]. Up to the present SP-spectroscopy technique was mainly practiced on spectral frequencies of the most intensive lines of CO_2- [4] and CO-lasers [5] as well as methanol vapor lasers [6]. As soon as *free-electron lasers* (FEL) were built scientists could study another important feature of SP – their dispersion in broad areas ranging from plasma (~50,000 cm^{-1}) to *terahertz* (THz) frequencies [7].

It was demonstrated in papers [2,8], devoted to spectroscopy and optics of SP, that combination of bulk and surface waves has good perspectives for the infrared interferometry of metal surfaces and thin films on them. These are especially important for optical constants determination in the THz spectral range, where other well-known methods such as far-IR reflectometry and Fourier-spectroscopic ellipsometry practically cannot be used for this purpose due to very high reflectivity of metals [9,10].

As preliminary analysis based on Drude model gives very pessimistic forecast for the SP measuring techniques at THz frequencies we decided to pose the problem of finding conditions under which THz SP have reasonable both the absorption coefficient $\alpha = 1/L$ and the refractive index κ (kappa). Having solved the problem we can employ the methods of SP-measurements used in middle infrared and thus extrapolate the techniques to the THz range.

To influence SP characteristics we chose the method by cladding the metal surface with a transparent layer of thickness d. This method is practiced in the visible and middle infrared spectral ranges to control SP characteristics when performing optic measurements [1,3]. The

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functioning of this method at THz frequencies is the subject of this paper.

2. Computer modeling

SP propagation in Metal—Dielectric-1—Dielectric-2 (M-D1-D2) structures was mainly investigated by those engaged in SP spectroscopy both at the visible [11] and the middle infrared ranges [12]. It was stated [12] that as the dielectric layer thickness d increases the SP refractive index κ increases monotonically until it reaches its value for the interface "M-D1". As for SP absorption it is almost linearly proportional to d up to the critical value $d_c = \lambda/\lfloor 4\sqrt{n_l^2 - 1}\rfloor$ (where n_l—is the refractive index of "cladding layer", λ—is the radiation wavelength in free space), reaches maximum at $d = d_c$ and decreases to the value of SP absorption at the interface "M-D1" as d trends to infinity.

To test whether this statement is true for lower optical frequencies we performed computer simulations for evaporated aluminum (Al) covered by a germanium (Ge) layer. Complex value of SP refractive index $\kappa = \kappa' + \mathrm{i}\cdot\kappa''$ (where i—is the imaginary unit) was determined numerically by solving the SP dispersion equation for a three-layer structure [1]. Optical constants for Al were taken from [10], while the value of n_l for Ge layer was supposed to be equal 4.0 for all wavelengths considered (as n_l varies only in the third figure after the decimal point for λ changing from 10 to 110 μm) and the absorption coefficient is negligibly small (less than 10^{-3}).

First we calculated characteristics of SP propagating along the ideal "Al–vacuum" interface. Table 1 contains modeled values of both real-(κ') and imaginary-(κ'') parts of SP's refractive index κ as well as those of L, calculated for different λ. The second and third columns of the Table 1 are filled up with Al optical constants (n and k), the forth and the fifth columns—with real (ε_1') and imaginary (ε_1'') parts of Al dielectric complex permittivity $\varepsilon_1 = (n + \mathrm{i}k)^2$, which for $\lambda = 110$ μm were calculated using Drude model (the experimental data are not available in handbooks), the ninth and the tenth columns—with values of SP field penetration depth into Al (δ_1) and vacuum (δ_2). One can see that as the radiation wavelength increases SP acquire more pronounced photon character, namely at THz frequencies its phase velocity approaches the speed of light, field's penetration depth (δ_2) increases up to centimeters, while propagation length reaches 4 m. The conclusion from this calculations is that in the case of bare metal surface it is impossible to determine SP propagation length L with accuracy of 10% using the two-prism method [1] and a specimen of reasonable size (10–15 cm).

Then we turned to calculations of L-dependencies upon the thickness d of Ge layer covering the aluminum substrate. Examples of these curves calculated for $\lambda = 10$ μm and $\lambda = 110$ μm are presented in Figs. 1 and 2, correspondingly. One can see that at the longer wavelength the effect of SP propagation length decrease at $d = d_c$ is more pronounced. Therefore it is possible to reduce L at Thz frequencies nearly by four orders of magnitude (from 4 m to 0.475 mm) using the thin-film cladding technique.

To underline the fact that the technique is more effective on longer wavelengths we calculated relationships of the extreme values of L for the structure "Al–Ge layer of thickness d – air" for different λ. The results of calculations are presented in Table 2, where L_0—is the SP propagation length at $d = 0$, $L_{\min}$—is the value of L at $d = d_c$, L_∞—is the value of L at $d \to \infty$. The column $L_0/L_{\min}$ illustrates the fact that the Ge layer presence on Al increases SP absorption more radically at larger λ.

It is worth to note that the small selective absorption into the Ge layer causes the decrease of L not more than by 1%.

3. Experimental technique and set-up

3.1. The simplified two-prism method for measuring L at THz frequencies

To determine κ'' for middle infrared SP the L value is usually measured, as these SP characteristics are related by the expression: $L = \lambda/(2\pi\kappa'')$ [5]. The most convenient way to determine L is the two-prism method when one has to measure SP intensity I_1 and I_2 at least in two points x_1 and x_2 along the track:

$$L = \frac{x_1 - x_2}{\ln(I_1/I_2)}.$$

We used the simplified two-prism method, when the output prism is removed and the SP is transformed into a bulk wave due to diffraction at the specimen edge. In this case the distance run by SP is varied by shifting the input element along the SP track.

Table 1
Dispersion of SP characteristics, calculated for the interface "Evaporated aluminum–vacuum"

λ (μm)	n	k	ε_1'	ε_1''	κ'	κ''	L (cm)	δ_1 (μm)	δ_2 (μm)
0.6	1.4	7.65	−56.6	21.4	1.007808	0.003	0.002	0.012	0.76
1.0	1.35	9.6	−91.3	26.1	1.005100	0.001467	0.005	0.016	1.57
5.0	8.67	48.6	−2287	843	1.000193	0.000071	0.561	0.017	40.5
10.0	25.3	89.8	−7424	4544	1.000049	0.000030	2.65	0.018	160
20.0	60.7	147	−17925	17845	1.000014	0.000014	11.4	0.022	602
32.0	103	208	−32655	42848	1.000006	0.000007	34.5	0.024	1521
110.0	–	–	−31902	231622	$1.0+10^{-7}$	0.000002	413	0.040	20701

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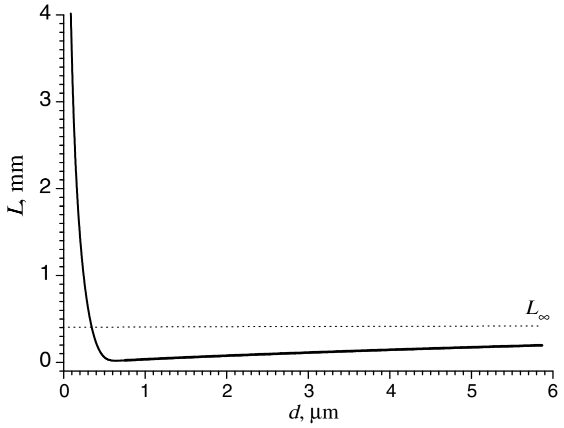

Fig. 1. Dependence of SP propagation length L on the thickness d of germanium layer covering the aluminum substrate calculated for radiation with $\lambda = 10$ µm.

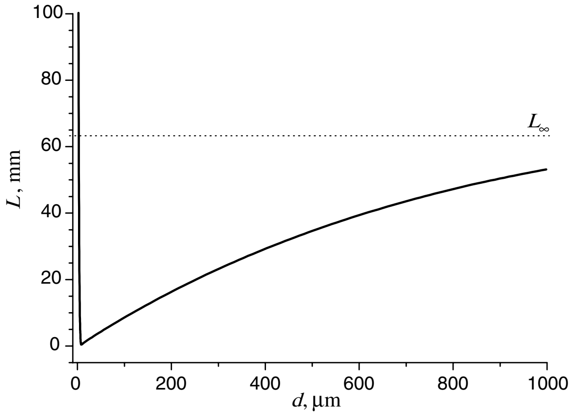

Fig. 2. Dependence of SP propagation length L on the thickness d of germanium layer covering the aluminum substrate calculated for radiation with $\lambda = 110$ µm.

Table 2
Dispersion of the extreme values of the SP propagation length L calculated for the structure "Evaporated Al–Ge layer of thickness d – air"

λ (µm)	d_c (µm)	L_0 (mm)	L_{min} (mm)	L_∞ (mm)	L_0/L_{min}	L_∞/L_{min}
0.63	0.029	0.017	8×10^{-5}	0.0002	209	93
1.0	0.049	0.053	2.5×10^{-4}	0.0007	212	83
5.0	0.311	5.61	7.6×10^{-3}	0.0870	736	65
10.0	0.600	26.5	0.021	0.4140	1280	64.15
32.0	2.100	345	0.100	5.3887	3545	64.01
110.0	7.20	4132	0.475	64.5608	8700	64.00

3.2. Description of the experimental set-up and the specimens

The scheme of the experimental set-up is presented in Fig. 3. Laser radiation is focused by the lens 1 onto the aperture 2, where it diffracts and with efficiency of several percents is converted into SP propagating along the specimen (substrate – 3, nontransparent metal film – 4). Having run the distance a SP arrive to the edge of specimen, where they transform into a bulk "edge diffracted" wave. The bulk wave with intensity proportional to the SP energy at the edge is registered by a photo detector 5 placed in the horizontal plane at a distance b from the specimen.

In our experiments FEL's radiation arrived at the set-up input in the form of macro pulses with duration of 3 µs (10 W) and repetition period of 1 s. The measured radiation intensity (normalized by independently measured beam's intensity) was detected, averaged over eight sequential pulses and finally memorized by a digital oscilloscope.

The specimens represented themselves nontransparent mirrors with Al films evaporated in vacuum (thickness more than 100 nm) on optically polished glass substrates with dimensions of 30 × 150 × 5 mm. A specimen was mounted onto a specially designed attachment enabling us to measure and control the distance a with precision of 0.1 mm. For SP excitation on aluminum specimens we used an ordinary "razor blade" as a diffraction element (the "aperture" method of excitation), while in experiments with copper specimens—a right-angle prism with a sharp front edge usually used in the two-prism method. The blade sharp edge (or the prism edge) was fixed over the specimen surface at the distance of 2 mm and this gap remained the same in all the experiments performed. FEL radiation beam with $\lambda = 110$ µm and 10 mm in diameter was focused on the slit (formed by the blade or the prism and the specimen surface) by a polyethylene lens with the focal length of 15 cm. In the case of the blade input element the optical axis of the lens was inclined relatively to the horizon by approximately 10 angle degree to avoid straight incidence of the laser radiation onto the photo detector. A Ga-doped germanium photo resistor, cooled by liquid helium and provided with 2 mm horizontal diaphragm on the entrance window of cryostat was used as a detector. The signal to noise ratio was on the level of 100.

To change the distance a run by SP the specimen was moved with the step of 0.1 mm towards the detector. To

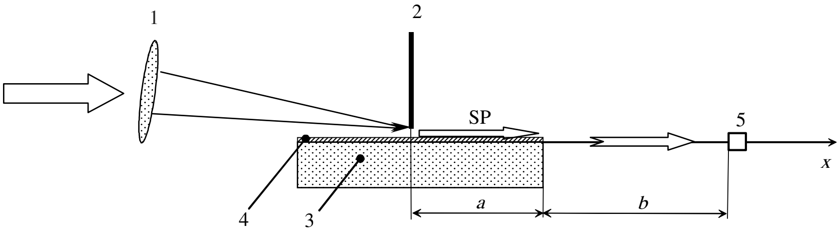

Fig. 3. Scheme of the experimental set-up.

take into account the change of distance b due to the specimen motion during an experiment we recalculated the electric signal meter readings on the assumption that the bulk "edge diffracted" wave intensity was inversely proportional to the distance covered by the wave in air.

4. Results of the experiments and their consideration

The experiments were performed at the Korea Atomic Energy Research Institute (KAERI) where a new THz FEL was recently built [13].

SP propagation distance measurements were done with aluminum and copper samples (bare and covered by a germanium layer of various thicknesses). Results of the experiments with aluminum are presented in Fig. 4. One can see that SP intensity on the bare aluminum surface (curve 1) decreases by about 50% at the distance of 10 cm that corresponds to the SP propagation length $L \approx 15$ cm and $\kappa'' \approx 1.16 \times 10^{-4}$. The germanium layer 100 nm thick (curve 2) brings down L to 10 cm, that corresponds to $\kappa'' \approx 1.75 \times 10^{-4}$.

Results of the experiments with copper films are presented in Fig. 5. One can see that experimental points are oscillating with increase of the distance a. These oscillations take place due to the interference of the bulk wave produced by SP at the specimen edge and the incident radiation, which partially passes through the input prism and propagates parallel to the surface of specimen. But averaging curves have clear exponential form and the damping factor is obviously proportional to the Ge layer thickness d. Calculations gave the following results: $L \approx 19.5$ cm, $\kappa'' \approx 9 \times 10^{-5}$ for $d = 0$; $L \approx 12.2$ cm, $\kappa'' \approx 1.5 \times 10^{-4}$ for $d = 0.2$ μm and $L \approx 8.5$ cm $\kappa'' \approx 2.1 \times 10^{-4}$ for $d = 0.5$ μm.

These values of L are unexpectedly small. They are inconsistent with the results for L obtained from Drude model for aluminum dielectric permittivity (ε_1) calculations [10]. In this case, for example, simulations for aluminum give L value of about 4 m (bare aluminum), while the experiments gave $L \approx 15$ cm.

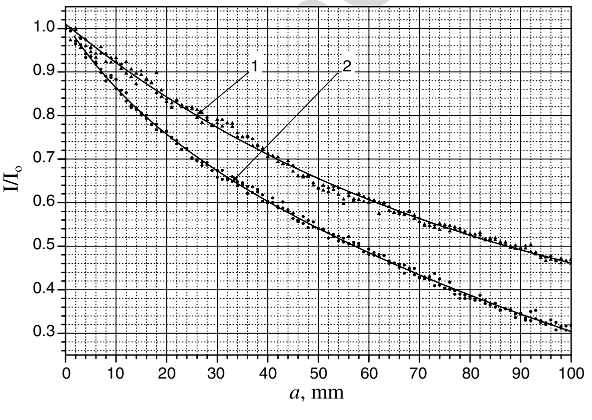

Fig. 4. Relative SP intensity I/I_0 versus distance a run by the SP for clean aluminum surface (curve 1) and the surface covered with 100 nm thick germanium layer (curve 2).

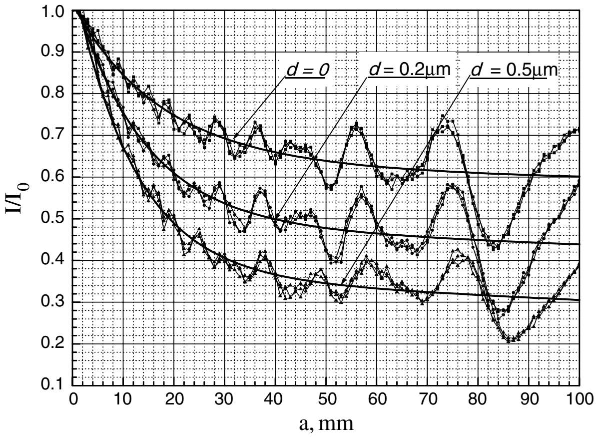

Fig. 5. Relative SP intensity I/I_0 versus distance a run by the SP along clean and covered germanium layer of various thicknesses copper specimens.

We are not the first to discover this discrepancy. A number of investigators, equipped with water vapor and methanol lasers, came across with this fact and could not give any reasonable explanation to it [14–17]. More sophisticated investigations of the phenomenon should be done to explain it.

However we think that the discrepancies may be the result of: (1) difference in values of the plasma and electron collision frequencies for crystal and evaporated aluminum; (2) radiation losses due to the roughness of the metal's surface; (3) presence of an oxide layer on the metal's surface etc.

5. Conclusion

Surface plasmons (SP) propagation losses at the frequency of 90 cm^{-1} have been estimated for aluminum and copper evaporated specimens. The experimental results do not correlate with the calculated ones. To our mind this does not mean that the experiment's results are false or Drude model is not true for the THz range. We suppose that the discrepancies between the experiments and the calculations follow predominantly from different physical properties of the evaporated and crystal forms of the metals.

The method for decreasing both SP's phase velocity and propagation length and employing the metal's surface coverage with a transparent layer has been tested at THz frequencies. The method enables scientists to use SP in far-infrared opto-electronic devices as well as to perform SP-spectroscopy of super thin transition layers on metal surfaces in the THz range.

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