# Aluminum optical constants in far infrared determined from surface electromagnetic waves characteristics

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## ABSTRACT

Aluminum dielectric permittivity has been determined for free-electron laser radiation of 110 micrometers by measuring surface electromagnetic waves (SEW) refractive index and propagation length. Both SEW's characteristics were measured using a newly developed double-beam surface-plasmon interferometer. The measurements were performed for clean aluminum surface and in the presence of a transparent germanium over layer.

**Keywords:** optical constants, far infrared, surface electromagnetic waves, surface plasmons, terahertz radiation, aluminum, free-electron lasers

## 1. INTRODUCTION

Optical constants (indexes of refraction *n* and absorption *k*) are fundamental constants of substance. They are of great importance for many practical applications of metals, namely for calculations of spaceships' radiative balance, for remote radiometry, for modeling special mirrors and protecting covers etc. The need of data for far infrared is provoked by increasing application of the terahertz (THz) waves (from 400 to 30 cm$^{-1}$) in communication problems as well[1].

However metals' optical constants frequency dependences in the referred spectral range have not been measured up to now because none of the practiced optical spectral methods (such as transmission or reflection intensity measurements, ellipsometry, Fourier-spectroscopy and others) is able to determine the constants due to the high reflectance ability of metals in far infrared. In handbooks on optical constants (see for example[2]) data only for twelve metals are presented and furthermore it is restricted by the long wavelength boundary of 25 μm (400 cm$^{-1}$). Data on other metals are not available in this handbook at all.

A few decades ago a new powerful method for conducting materials surface optical study was developed. The method is based on generation of *surface electromagnetic waves* (SEW) by the probing radiation[3]. Since that time SEW were widely used in surface science of metals as well as for their refractometry, bringing good results in the visible and middle infrared spectral ranges[4,5].

But up to now the SEW-method was not practiced in far infrared, as at these frequencies the phase velocity of SEW differs from the speed of light in free space very slightly (the excess of SEW's refractive index æ′ over unity Δæ′= æ′-1 is about 10$^{-6}$), while SEW's propagation length *L* reaches 10 meters. Due to these peculiarities accuracy of optical constants determination in far infrared is very low and the SEW-method is as useless for refractometry of metals at THz frequencies as any other optical method.

To change the situation we modernized the known interferometric technique enabling one to establish SEW phase velocity in the middle infrared [6]. The new technique employs the sample's surface coverage by a thin transparent layer that increases Δæ′ and decreases *L*. Thus we hope to increase accuracy of both Δæ′ and *L* measurements and make it possible to determine the metal's dielectric permittivity and optical constants.

The developed technique was successfully tested on aluminum samples covered by a germanium film at the frequency of 91 cm$^{-1}$. We hope that this technique can be applied to many metals in the whole far infrared spectral range.

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## 2. METHODOLOGY

Surface plasmon-polaritons, that is SEW in the case of metal samples, can exist on an interface of two media only under the condition that real parts of their dielectric constants $\varepsilon_1$ and $\varepsilon_2$ have opposite signs. SEW are characterized by complex effective refractive index æ=æ′+$j$·æ″ (where $j$ – imaginary unit)[3]:

$$æ = \sqrt{\frac{\varepsilon_1 \cdot \varepsilon_2}{\varepsilon_1 + \varepsilon_2}}\ , \qquad (1)$$

determining the wave's phase velocity $\vartheta$=C/æ′ (where C – is the speed of light in the medium with $\varepsilon_2$) and the SEW's propagation length $L$~1/æ″.

If we know the real part æ′ of SEW's effective refractive index along with its imaginary part æ″, than it is possible to estimate the metal's complex dielectric permittivity $\varepsilon_1=\varepsilon'+j\cdot\varepsilon''$ and its optical constants $n$ and $k$, related to $\varepsilon_1$ as $\varepsilon'=n^2-k^2$ and $\varepsilon''=2nk$.

### 2.1 Determination of the real part of SEW's refractive index

To determine æ′ we used the SEW-interferometry method[6]. The method can be realized on a modified Michelson interferometer where radiation in one of the shoulders passes a part of its path in the form of SEW. Being converted into SEW the probing radiation accumulates information about the sample's surface. The information is encoded in the interference picture formed on the interferometer's screen. The method was tested in the middle infrared (~1000 $cm^{-1}$) on metals[7] as well as at THz frequencies on dielectric crystals[8]. But under these conditions the phase shift acquired by SEW at a distance of several centimeters (average SEW's propagation length on the mentioned materials and frequencies) amounts up to $10^{-3}$ radians, while on metal samples at THz frequencies the shift gained by SEW at a distance of 10 cm (size of the specimen) is estimated to be $10^{-5}\div10^{-6}$ radians only.

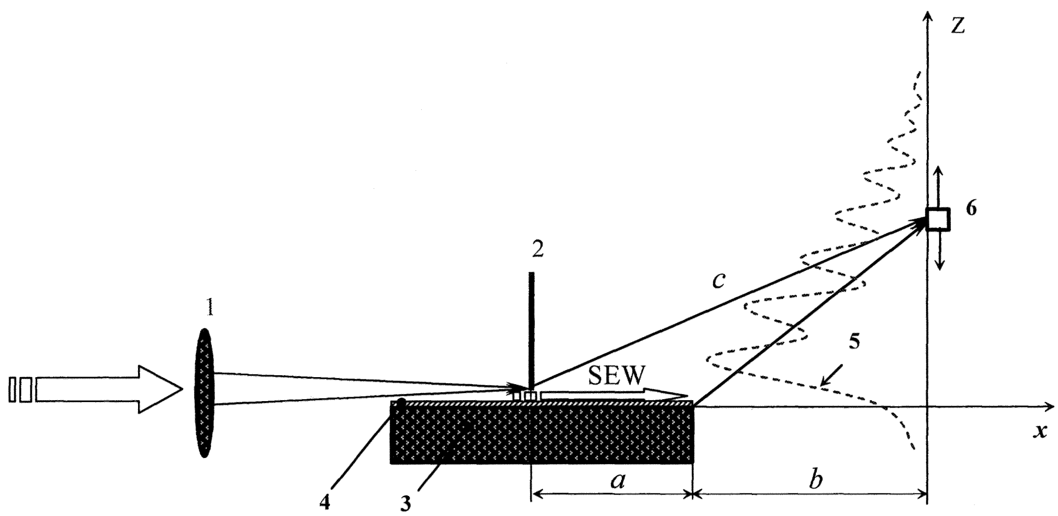


Figure 1: The scheme of the experimental set-up implementing the SEW-interferometry method.

The scheme of the experimental set-up is presented in Fig.1. Laser radiation of wavelength $\lambda$ is focused by the lens **1** onto the aperture **2** where it diffracts and is split into two parts, one of which is formed by a bunch of dispersing bulk waves propagating in the surrounding medium at various angles relatively to the surface of the specimen (substrate – **3**, metallic film – **4**) and the other part of the incident radiation is converted into SEW propagating just along the specimen's surface. While SEW runs the distance $a$ to the specimen's edge it gains phase shift $\Delta\varphi = k_o\cdot æ'\cdot a$ (where $k_o=2\pi/\lambda$) and transforms at the edge into a bulk wave, carrying information about the surface. As these two bulk waves (the first one produced at the aperture **2** and the second – at the specimen's edge) meet an interference picture **5** (interferogram), containing information about characteristics of SEW, namely about its phase velocity, is formed. The interferogram is registered by a photo detector **6** scanned in the vertical direction at a distance $b$ from the specimen's edge.

Analytical procedure for determining the value of æ′ from two interferograms obtained at different distances $a$ run by SEW is described in our paper[9]. Unfortunately to determine the value of Δæ′= æ′-$n_2$ (where $n_2$ – refractive index of the medium bordering the metal) by the SEW-interferometry method the with accuracy of $10^{-4}$ or less one has to localize the interference pattern maxima positions with precision of about 10 μm, that can hardy be realized under laboratory conditions.

The situation changes for the best if we cover the metal surface with a transparent layer. This way to influence SEW characteristics is used both in visible[3] and middle infrared spectral ranges[10]. It is known that a thin-layer coverage of the metal sample decreases both the SEW's propagation length $L$ and phase velocity, that is increases the corresponding values of æ′ and Δæ′. So we followed the way to increase æ′ and Δæ′.

We elaborated a new procedure for obtaining quantitative information about æ′ from the interferograms got for clean metal surface and for the surface covered with a layer. Here are the main points and the calculation formula of the procedure.

Suppose we have registered two interferograms (prior and after formation of the layer) for the same distances $a$ and $b$ (see fig.1). The latter interference pattern is displaced to smaller values of $z$ by distance $\Delta z$ as compared with the former one. That means that the $m$-th maximum found at the coordinate $z$ shits to the coordinate ($z$-$\Delta z$) as the layer appears. This displacement takes place due the increase of æ′. Let $c_1$ and $c_2$ be the distances covered in air by the dispersing bulk waves produced at the aperture 2 prior and after formation of the layer, accordingly. Furthermore we have all foundations to anticipate that in the case of clean metal's surface placed in air æ′≈ $n_2$=$n_{air}$. Than the conditions for obtaining interference maxima of the $m$-th order at the screen's points with coordinates $z$ and ($z$-$\Delta z$) prior and after formation of the layer are the following:

$$\begin{cases} c_1 \cdot n_{air} - \left(a + \sqrt{b^2 + z^2}\right) \cdot n_{air} \approx m \cdot \lambda, & \text{(for bare metal's surface)} \quad (2) \\ c_2 \cdot n_{air} - \left(a \cdot æ' + \sqrt{b^2 + (z - \Delta z)^2}\right) \cdot n_{air} = m \cdot \lambda, & \text{(for covered metal's surface)} \quad (3) \end{cases}$$

where $c_1=\sqrt{(a+b)^2 + z^2}$ and $c_2=\sqrt{(a+b)^2 + (z - \Delta z)^2}$ .

Solving the system of equations (2) and (3) relatively æ′, we get the expression for calculating the real part of the SEW's effective refractive index in the case when the metal is covered with a layer:

$$æ' \approx n_{air} \cdot \left[1 + \left(\sqrt{b^2 + z^2} - \sqrt{b^2 + (z - \Delta z)^2} - \sqrt{(a+b)^2 + z^2} + \sqrt{(a+b)^2 + (z - \Delta z)^2}\right) \times \frac{1}{a}\right]. \quad (4)$$

**2.2 Determination of the image part of SEW's refractive index**

To establish the value of æ″ in the middle infrared they usually measure the SEW's propagation length $L$ as these SEW characteristics are related by the expression: $L=\lambda/(2\pi \cdot æ'')$. The most convenient way to determine $L$ is the two-prism method, when they measure intensity of the SEW's field $I_1$ and $I_2$ as the surface wave runs distances $x_1$ and $x_2$, correspondingly[3]. Assuming that $x_1>x_2$, the value of $L$ can be calculated by the formula:

$$L = \frac{x_1 - x_2}{ln(I_1/I_2)}. \quad (5)$$

As for THz frequencies we were not sure that the two-prism method will be appropriate. These doubts were based on the results of computer simulations, which had been done using the Drude model for the complex dielectric permittivity of some metals (aluminum, copper, gold etc). The calculations indicated that SEW's propagation length should amount up several meters and even more. If so than one can hardly measure $L$ with accuracy of at least 20%, having at his disposal a specimen with dimension of 10÷15 cm (the common size of substrates used for evaporation of uniform metal films).

To use the same experimental set-up for determining æ′ and æ″ at THz frequencies we simplified the two-prism method by removing the output prism and using the specimen's edge instead of it. In this case the surface wave is converted into a bulk wave due to diffraction at the edge, while the distance *a* (run by SEW) can be changed by moving the aperture 2 (see Fig.1) over the specimen's surface in the direction of the radiation propagation.

**2.3 Calculation of the metal's optical constants using determined SEW's characteristics**

Having determined both parts of the SEW's complex effective refractive index æ we can estimate the metal's complex dielectric permittivity $\varepsilon_1$ and its optical constants *n* and *k* using SEW's dispersion equation for the corresponding wave guiding structure.

In case of bare metal's surface one has to solve the equation (1) relatively $\varepsilon_1$ substituting æ′ and æ″ (experimentally found by the above described procedures) in it.

In case of the metal's surface covered with a layer of thickness *d* made of a material with refractive index $n_l$ and surrounded by a medium with dielectric constant $\varepsilon_2$, one has to solve relatively $\varepsilon_1$ the SEW's dispersion equation for a three-layers structure[3]:

$$\left(\varepsilon_1 \cdot \sqrt{(æ')^2 - \varepsilon_l} + \varepsilon_l \cdot \sqrt{(æ')^2 - \varepsilon_1}\right) \times \left(\varepsilon_l \cdot \sqrt{(æ')^2 - \varepsilon_2} + \varepsilon_2 \cdot \sqrt{(æ')^2 - \varepsilon_l}\right) + \left(\varepsilon_1 \cdot \sqrt{(æ')^2 - \varepsilon_l} - \varepsilon_l \cdot \sqrt{(æ')^2 - \varepsilon_1}\right) \times \left(\varepsilon_l \cdot \sqrt{(æ')^2 - \varepsilon_2} - \varepsilon_2 \cdot \sqrt{(æ')^2 - \varepsilon_l}\right) \times exp\left(-2k_o \cdot d \cdot \sqrt{(æ')^2 - \varepsilon_l}\right) = 0 , \quad (6)$$

where $\varepsilon_l = n_l^2$ - is dielectric permittivity of the material the layer is made of.

Finally one may calculate the optical constants (indexes of refraction *n* and absorption *k*) of the metal substituting the found values of ε′ and ε″ into the following formulae[2]:

$$n = \frac{\varepsilon''}{\sqrt{2} \cdot \sqrt{-\varepsilon' + \sqrt{(\varepsilon')^2 + (\varepsilon'')^2}}} , \quad k = \sqrt{-\frac{\varepsilon'}{2} + \frac{1}{2} \cdot \sqrt{(\varepsilon')^2 + (\varepsilon'')^2}} . \quad (7)$$

## 3. RESULTS OF THE EXPERIMENTS

The experiments were performed at the Korea Atomic Energy Research Institute (KAERI) where they recently built with technical support of the Budker Institute of Nuclear Physics (Novosibirsk, Russia) the new FEL generating THz radiation in the spectral range from 60 to 100 $cm^{-1}$ [11].

**3.1 Description of the specimens and the set-up equipment**

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 aluminum films (thickness more than 100 nm) made by thermal evaporation on optically polished glass substrates (with dimensions 30×150×5 mm) in vacuum. A specimen was mounted as an essential part of the SEW-interferometer (described in section 2.1) onto a specially designed attachment enabling us to measure and control the distance *a* with precision of 0.1 mm. An ordinary razor blade was used as a diffraction element for SEW excitation (the «aperture» method of excitation). The sharp edge of the blade was placed at a distance of 2 mm over the specimen's surface and remained constant and the same in all the experiments performed. FEL's radiation with λ=110 μm and 10 mm in diameter of its cross section was focused on the slit formed in this way by a polyethylene lens with the focal length of 15 cm. The main optical axis of the lens was inclined relatively to the horizon by approximately 10° 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.

**3.2 SEW's phase velocity measurements**

To establish the real part æ′ of the SEW's refractive index, that determines SEW's phase velocity, we used the newly developed double-beam surface-plasmon interferometer described in section 2.1. Two types of experiments were performed: for clean specimen surface and the surface containing the coverage layer made of germanium with thickness of 100 nm.

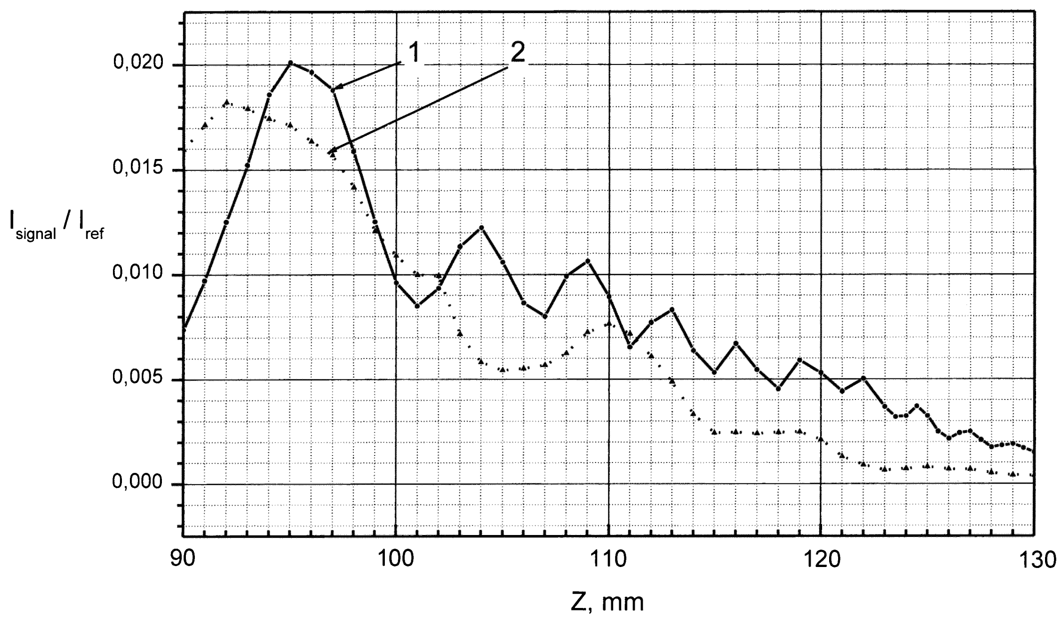


Figure 2: Interferograms registered for the aluminum specimen at various distances *a* covered by SEW on the specimen's surface: curve 1 – *a*=80 mm, 2 – *a*= 20 mm.

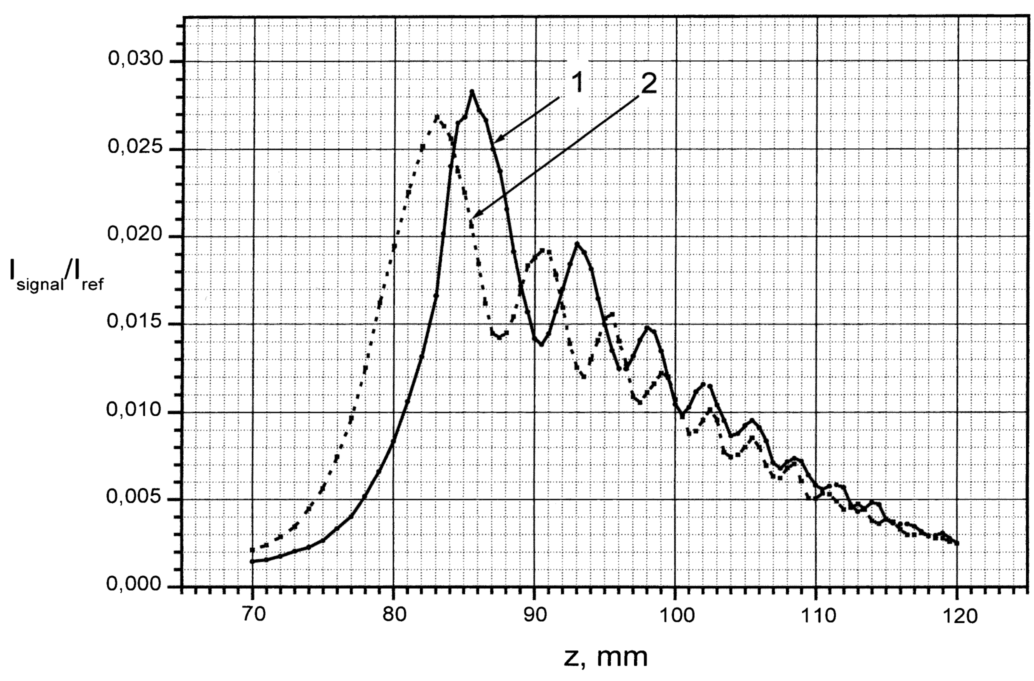


Figure 3: Interferograms obtained for clean aluminum surface (curve 1) and for the surface covered with 10 nm thick germanium layer (curve 2). Distance run by the SEW is *a*=120 mm.

As indicated above æ′ determination in case of clean metal surface suggests registration and further execution of at least two interferograms obtained for different distances $a$ run by the SEW[9]. So we registered interference pictures at the same distance $b$ between the detector and the sample (200 *mm*) and two different distances $a$ of 80 and 20 mm (Fig.2). Following the execution procedure described in [9] we found that æ′ value was just the same as the refractive index of air ($n_{air}$=1.00027) within the error interval. But according to the formula (1) æ′ should be larger than $n_{air}$. To our mind this experimental "blindness" means that the precision of the interference maxima positions determination was not sufficient. We moved the detector along the axis $z$ with the step not less than 0.5 mm, while maxima coordinates in the case of bare metal surface should be determined with accuracy of not worse than 10 μm in order to estimate the value of æ′ up to the forth figure after decimal point.

That is why we came to the idea that it is necessary to increase æ′ artificially by covering the metal's surface with a transparent layer. Under these conditions (when value of æ′ is considerably larger than $n_{air}$) the detector's displacement step of 0.5 mm permits us to determine æ′ with less relative error.

So we covered one of the aluminum specimens with the germanium layer of thickness 100 nm and registered interferograms (presented in Fig. 3) this time for the bare and covered specimens at the distances $a$=120 mm and $b$=200 mm. The interferograms have the same number of maxima but the curves are shifted relatively one another by the easily measurable distance $\Delta z$ = 2.5 mm. Having measured the coordinates $z$ of the interferogram 1 (corresponding to $d$=0) maxima counted from the specimen's surface coordinate $z_o$=80.0 mm, and using values of $\Delta z$, $a$ and $b$ we calculated æ′ by formula (4). Now the result was reasonable, namely æ′=1.0004.

**3.3 SEW's attenuation measurements**

SEW's propagation distance measurements were done with the same two specimens (bare and covered by 100 nm germanium layer) using the simplified two-prism method described in section 2.2. Take notice that in these series of experiments the distance $b$ changed while the specimen was moving towards the detector to change the distance $a$. To take into account this change of $b$ we recalculated the electric signal meter readings on the assumption that the bulk wave (produced by the SEW at the specimen's edge) intensity was inversely proportional to the distance covered by the wave in air.

Results of the experiments are presented in Fig.4. One can see that SEW's intensity on the bare aluminum surface (curve 1) decreases by about 50% at the distance of 10 cm that corresponds to the SEW's propagation length $L\approx15$ cm and æ″≈$1.16\cdot10^{-4}$. The germanium layer (curve 2) brings down $L$ up to about 10 cm, that corresponds to æ″≈$1.75\cdot10^{-4}$.

These values of $L$ are unexpectedly small. They are inconsistent with the modeling results for $L$ obtained in case we use the Drude model for dielectric permittivity $\varepsilon_1$ of aluminum [2]. The calculations brought $L$ values of about 7 meters (in the case of clean aluminum surface), while the experiments gave $L\approx15$ cm.

We are not the first to discover this discrepancy. A number of investigators, who used water vapor and methanol lasers, came across with this fact and could not give some reasonable explanation to it [12-15]. The same is true for us at the moment. More thorough and sophisticated investigations of the phenomenon shall be done to explain it.

**3.4 Calculation of aluminum optical constants using the experimental data**

We used the experimental data (æ′=1.0004 and æ″≈$1.75\cdot10^{-4}$) got the specimen covered with the germanium layer and presented in sections 3.2 and 3.3 for calculating dielectric permittivity $\varepsilon_1$ and optical constants ($n$, $k$) of the evaporated aluminum using the dispersion equation (6) for SEW in a three-layer structure. The calculations brought the following results: $\varepsilon'\approx$-4000 and $\varepsilon''\approx$6500, which corresponds to $n\approx43$ and $k\approx76$.

These values of $\varepsilon'$ and $\varepsilon''$ are very far from the values ($\varepsilon'\approx$-32000 and $\varepsilon''$=230000) calculated using the Drude model (using the model we took the values of the plasma and collision frequencies $\nu_p$=11900 cm$^{-1}$ and $\nu_\tau$=660 cm$^{-1}$ inherent for crystal aluminum[2]).

Certainly these discrepancies have many in common in with those ones for the SEW's propagation length $L$. Nevertheless we shall express our opinion on possible reasons of $\varepsilon'$ and $\varepsilon''$ misestimating. The discrepancies may be the result of: 1) difference in values of $\nu_p$ and $\nu_\tau$ for crystal and evaporated aluminum; 2) presence of an oxide layer on the metal's surface; 3) ignoring the intensity distribution of the bulk waves produced due to diffraction at the aperture and the specimen's edge; 4) radiation losses due to the roughness of the metal's surface etc.

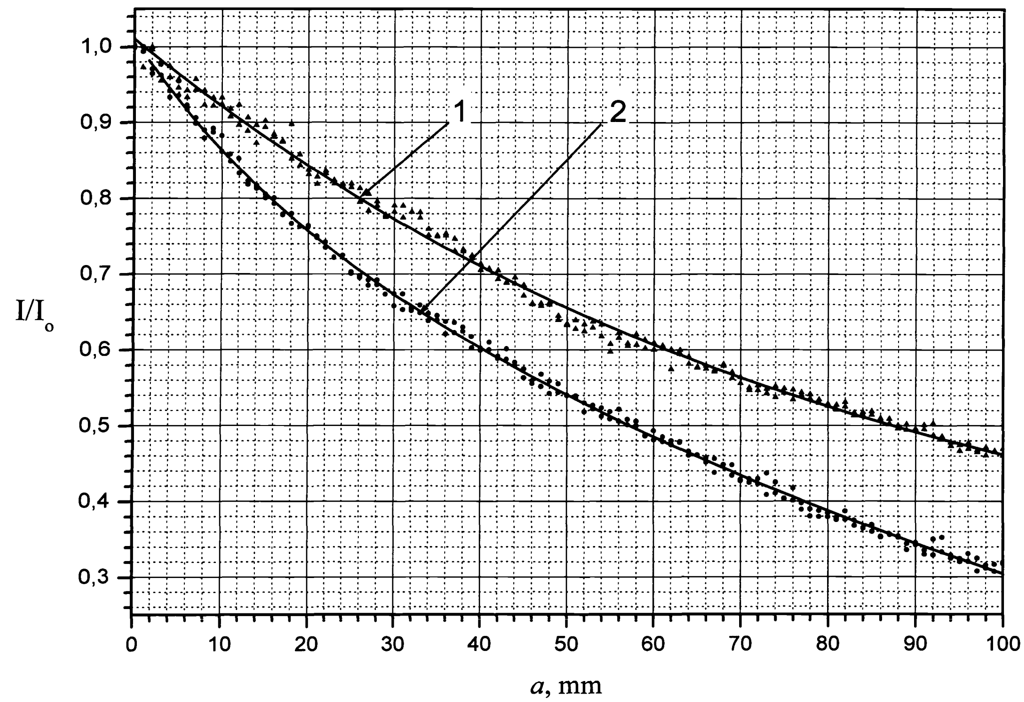


Figure 4: Relative SEW's intensity $I/I_0$ visa distance *a* run by the SEW for clean aluminum surface (curve 1) and the surface covered with 100 nm germanium layer (curve 2).

## 4. CONCLUSIONS

Aluminum complex dielectric permittivity $\varepsilon_l$ for terahertz radiation of 110 micrometers using measured surface electromagnetic waves (SEW) characteristics has been determined for the first time. The results do not correlate with the values of the real and image parts of $\varepsilon_l$ calculated using the Drude model. This does not mean that the experiment's results are false or the Drude model is not true for the far infrared. We suppose that the found value of $\varepsilon_l$ is an effective one and inherent for the evaporated aluminum not for the metal's crystal form.

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

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