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Generation of surface electromagnetic waves in terahertz spectral range by free-electron laser radiation and their refractive index determination

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Abstract

First experiments for observation of surface electromagnetic waves (SEW) in the terahertz spectral range generated on dense aluminum films covering the optical quality glass plates are presented in this paper. Coherent radiation of the new free-electron laser covering the frequency range from 30 to $100\,cm^{-1}$ was used. The interference technique employing SEW propagation in the part of one shoulder of the asymmetric interferometer was applied. From the interference pattern the real part of SEW's effective refractive index $æ'$ was determined for the two laser emission wavelengths: at $\lambda = 150\,\mu m - æ' = 1 + 5 \times 10^{-5}$, at $\lambda = 110\,\mu m - æ' = 1 + 8 \times 10^{-4}$. High sensitivity of the interference patterns to overlayers made of Ge and Si with thickness of 100 nm was demonstrated as well.

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1. Introduction

Up to now investigations of *surface plasmon-polaritons* (SPP) [1,2] were mainly performed in relatively small spectral regions adjacent to more

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intensive lines of generation of CO [3], CO_2 [4] lasers as well as methanol vapor lasers [5]. The last decade the spectroscopists have got a unique opportunity to study SPP dispersion in broad areas of optical frequencies ranging from plasma ($\sim$50000 cm^{-1}) to terahertz (30–1000 cm^{-1}) frequencies with the appearance of *free-electron lasers* (FEL) [6].

It was demonstrated in papers [6–9], devoted to spectroscopy and optics of SPP, existing in the form of propagating *surface electromagnetic waves* (SEW), that combination of bulk and surface waves has good perspectives for two methods of characterization of metal surfaces and thin films on them. The first one, known as the two prisms method [6], gives the information on SEW losses related with imaginary part ($\text{æ}''$) of SEW's complex refractive index æ, the second one—the interferometric method—gives the information on real part ($\text{æ}'$) of æ. Both methods used together give possibility to reconstruct full refractive index $\text{æ} = \text{æ}' + j\text{æ}''$ spectrum in very broad frequency range, while other well-known methods such as reflectometry and infrared (IR) spectro-ellipsometry are unable to do it [10,11].

The new FEL recently built in Korea Atomic Energy Research Institute (KAERI) with technical support of Budker Institute of Nuclear Physics (Novosibirsk, Russia) generates radiation in the longest far IR spectral range from 30 to 100 cm^{-1} [12]. The main purpose of the experiments we have done at KAERI was to test the applicability of SEW interferometric technique in this range for determination of complex refractive index of SEW on smooth metallic surfaces.

2. Brief theory and experimental setup description

SEW can exist on an interface of two media only under the condition that real parts of their dielectric constants ε_1 and ε_2 have opposite signs. Than SEW dispersion equation is [1,2]:

$$\text{æ} = \sqrt{\frac{\varepsilon_1 \varepsilon_2}{\varepsilon_1 + \varepsilon_2}} \tag{1}$$

$\text{æ} = \text{æ}' + j\text{æ}''$ (j–imaginary unit)—the effective complex refractive index of SEW.

It is seen from Eq. (1), that having measured $\text{æ}'$ and $\text{æ}''$, we can proceed to complex dielectric function $\varepsilon = \varepsilon_2' + j\varepsilon_2''$ of the metal. Our method is the only one up to now promising to do this. From other side if we could find ε_2' and ε_2'' independently, for example from the Handbook [11], we could check our data for $\text{æ}'$ and $\text{æ}''$. But the data for optical constants in this newest Handbook are given for 11 metals only for wavelengths shorter than 31 μm, while for longer wavelengths are given not very reliable Drude extrapolation curves (aluminum (Al), for example).

As the value of $\text{æ}'$ is larger than the refractive index of the environment $n_1 = \sqrt{\varepsilon_1}$ with dielectric constant $\varepsilon_1 > 0$, a special coupling device is needed to excite SEW by a plane electromagnetic wave. A metallic screen placed over the sample's surface at the distance $d \sim 10\lambda$ (λ—wavelength) can be used for SEW excitation [6,9].

The scheme of the experimental setup is presented in Fig. 1. FEL's radiation is directed through a lens **1** onto the aperture **2** where it diffracts and partially transforms into bulk cylindrical waves propagating above the specimen's surface (glass substrate–**3**, metallic film–**4**) and another part of the incident radiation is converted into SEW propagating just along the sample's surface. While SEW runs the distance a to the specimen's edge it gains phase shift $\Delta\varphi = k_o \text{æ}' a$ (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 meet on the plane of IR detector **5** scanning, separated from the specimen's edge by distance b, they do form an interference picture **6** (interferogram), containing information about characteristics of SEW, namely about its phase velocity and attenuation.

According to Refs. [8,9], the condition for the interference maximum at a point z is the following:

$$\text{æ}' a + \sqrt{b^2 + (z - z_0)^2} - \sqrt{(b + a)^2 + (z - z_0)^2} = (m + \theta)\lambda \tag{2}$$

where z_0 corresponds to the zero optical paths difference between two beams: one of them is the SEW containing as a part, and the second is a pure bulk wave; $m = 1, 2, 3 \ldots$ is the integer corresponding to the order's number of maximum, θ is the

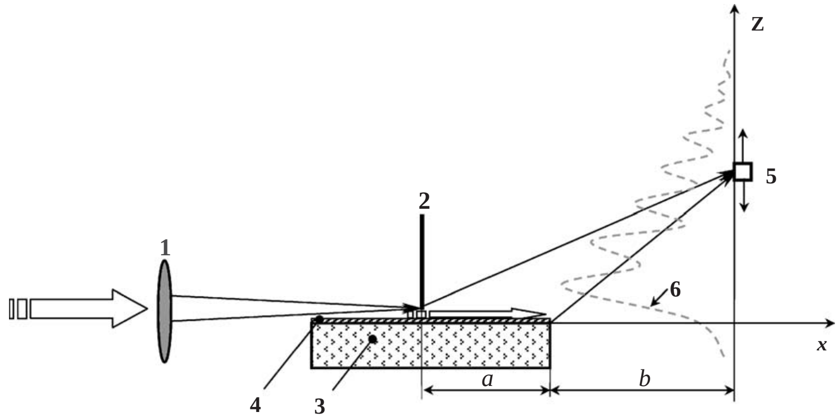

Fig. 1. The experimental setup scheme for performing SEW interferometry of solids' surfaces in terahertz region: **1**—polyethylene lens, **2**—razor blade (aperture), **3**—glass substrate, **4**—metallic film, **5**—scanned detector, **6**—space distribution intensity of interferogram.

additional phase shift undergone on the edge as SEW transforms into a bulk wave.

The value of z_0 can be determined by the subtraction of two equations of type (2), composed for two maxima having known phase differences in the same interferogram:

$$\sqrt{b^2 + z_m^2} - \sqrt{(b+a)^2 + z_m^2} - \sqrt{b^2 + z_{m+1}^2} + \sqrt{(b+a)^2 + z_{m+1}^2} + \lambda = 0 \qquad (3)$$

where $z_m = z - z_0$ and z is the coordinate of mth maximum. For better precision z_0 should be obtained for several combinations of couples of maxima inside the same interferogram and its averaged value should be calculated.

As the value θ does not depend on the distance a we can get rid of it and determine $æ'$ by the subtraction of two equations of type (2), composed for different a. In case we are dealing with the same interference order m of two interferograms obtained at the same b and different distances a_1 and a_2 the subtraction brings the following result:

$$æ' = \frac{\sqrt{b^2 + (z_{2m} - z_{02})^2} - \sqrt{b^2 + (z_{1m} - z_{01})^2} + \sqrt{(b+a_1)^2 + (z_{1m} - z_{01})^2} - \sqrt{(b+a_2)^2 + (z_{2m} - z_{02})^2}}{a_1 - a_2} \qquad (4)$$

where z_{01} and z_{02} are the values of z_0 obtained from the interferograms using Eq. (3) at a_1 and a_2, respectively; z_{1m} and z_{2m} are the coordinates of any mth maxima of the interferograms registered at a_1 and a_2, correspondingly.

3. Experimental results and discussion

The experiments were performed at two frequencies of the tunable FEL's radiation with wavelengths $\lambda = 110$ and $150\,\mu m$. Note that the radiation arrived at the input of the experimental setup in the form of macropulses with duration of $3\,\mu s$ (10 W, at $\lambda = 110\,\mu m$) and repetition period of 1 s, while registered radiation intensity (normalized on independently measured beam intensity) was averaged over eight sequential pulses, detected and memorized by a digital oscilloscope.

We studied the specimens made in different metals, but here we present the most reliable data only for Al. The nontransparent mirrors with metallic layers on the optically polished glass substrates (with dimensions $30 \times 150 \times 5\,mm^3$) were made by thermal evaporation in vacuum. A sample was placed as a part of SEW interferometer on a special attachment with possibility to change and measure the distance a with accuracy of 0.1 mm. An ordinary razor blade was used as a diffraction element (aperture) for SEW excitation. The sharp edge of the blade was over the surface at the distance 2 mm in all experiments described. The FEL radiation was focused at the blade's edge by a polyethylene lens with the focal length of 15 cm. As a detector the Ga-doped germanium photoresistor, cooled by liquid helium and provided with 0.2 mm horizontal diaphragm on the entrance window of cryostat was used. The signal-to-noise ratio was on the level of 100. Distance b between the detector and the sample remained constant (200 mm) in all experiments.

The interferograms registered in the experiments with $\lambda = 150\,\mu m$ and different distances a are presented in Fig. 2. Using the formula (3) we obtained $z_{01} = 80.548$ mm at $a_1 = 120$ mm and $z_{02} = 77.398$ mm at $a_2 = 80$ mm. Calculations by formula (4) brought us the value of $æ' = 1.00005$. As for the measurements and analogous calculations for $\lambda = 110\,\mu m$ we got the following results: $z_{01} = 90.887$ mm at $a_1 = 80$ mm and $z_{02} = 74.226$ mm at $a_2 = 20$ mm, meanwhile the value of $æ' = 1.0008$. The accuracy of $æ'$ determination at $\lambda = 150\,\mu m$ is higher due to smaller spectral width of the emission line of FEL and more accurate interference pattern view in contrast with the emission line at $\lambda = 110\,\mu m$.

As soon as we have $æ'$ value, the next step to the dielectric constant ε_2 is to find $æ''$ of $æ = æ' + jæ''$. From Eq. (1) in this case we could find $\varepsilon_2 = \varepsilon_2' + j\varepsilon_2''$. The difficulty for the approach to this goal is in the estimation of $æ''$ value from the same interferogram. It is possible to do it by the procedure described in Ref. [13], but the error will be too big. So we restricted our ambitions just by the $æ'$ determination, giving the value of the SEW phase speed $\vartheta = C/æ'$ (where C is the speed of light in free space). For correct determination of losses ($æ''$) we are planning a new experiment in future, which provides the possibility for direct determination of $æ''$ by the two prisms method [6].

To test SEW interferometer sensitivity to thin films on the metal surface we registered interferograms for $\lambda = 150\,\mu m$ and $a = 120$ mm for two Al specimens having silicon (Si) and germanium (Ge) layers on both of them with thickness 100 nm (Fig. 3). The interferograms' displacement in

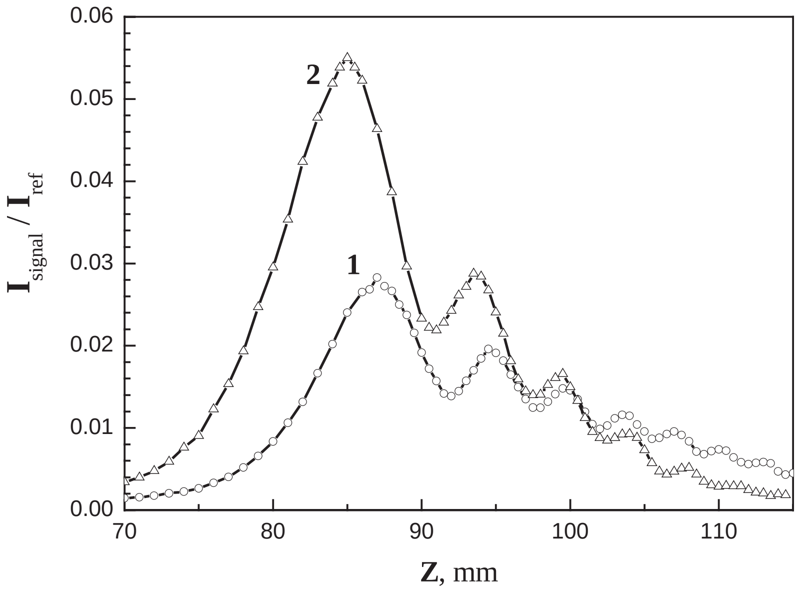

Fig. 2. Interferograms at $\lambda = 150\,\mu m$ at distance a of SEW propagation along aluminum specimen surface: **1**—$a = 120$ mm, **2**—$a = 80$ mm.

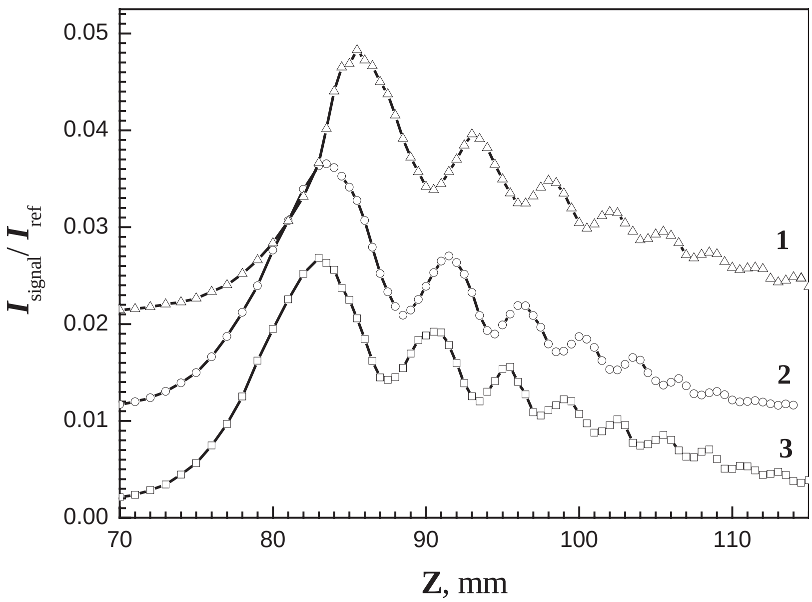

Fig. 3. Interferograms at $\lambda = 150\,\mu m$ for three samples: **1**—clean Al film, covered with silicon—**2**; and germanium (both 100 nm layers)—**3**. The interferograms of samples with Si and Ge films are shifted up on 0.01 and 0.02 units correspondingly. In all experiments, $a = 120\,mm, b = 200\,mm$.

comparison with clean Al surface to smaller values of z takes place due to the increase of $æ'$, which is proportional to the refractive index of the overlayer's material. Both interferograms, registered for Al surface with an overlayer, are identical, which means that films' refractive indexes are practically the same. Similar effect was observed in the 10 μm range for both Ge and Si films $n \cong 3$ [14], which was explained by conditions of films evaporation. One can see that even in the very far infrared SEW are sensitive to the presence of thin films on the guiding metallic surface.

4. Conclusion

For the first time in the terahertz frequency range, the interferometric studies of SEW on surface of thick Al films were made; the real part of SEW refractive index ($æ'$) was determined and the sensitivity of interference patterns of SEW to overlayers of Ge and Si was demonstrated. The further improvement of the experimental setup, data acquisition system and software are promising the high accuracy of the optical constants determination in this terahertz range for metals, metal oxides, dielectric films and crystals.

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