A way to determine the permittivity of metallized surfaces at terahertz frequencies

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A method for determining the dielectric permittivity of metal surfaces at terahertz (THz) frequencies has been suggested and tested. The method implies excitation of surface plasmons (SPs) on the sample surface and measurements of the SPs propagation length and field penetration depth in air. The technique was tested on evaporated gold with Novosibirsk free-electron laser at the wavelength of 130 μm. The method suggested paves the way for refractometry of metallized surfaces at THz frequencies. © *2011 American Institute of Physics*. [doi:10.1063/1.3584130]

Dielectric permittivity at terahertz (THz) frequencies is of great importance for many practical applications of metals, namely, for calculations of the radiative balance of spaceships, remote radiometry, modeling special mirrors, protecting coverings, etc.[1]

However, no metal permittivity has been measured up to now as none of the applicable methods (absorption reflectometry, ellipsometry, Fourier spectroscopy, and others) is able to measure it in the far infrared (IR) range, due to the high reflectivity of metals. Handbooks of optical constants of metals (see, for instance, Ref. 2) present data restricted by the long wavelength limit of 25 μm (400 cm^{-1}).

A few decades ago a powerful method for optical study of a conducting material surface was developed. The method is based on generation of surface plasmons (SPs), the complex of an evanescent *p*-polarized electromagnetic wave and a wave of free electron density propagating along a conducting surface, by probing radiation.[3]

SPs of the visible and middle IR ranges are widely used in surface science, spectroscopy, and sensing.[4,5] In the THz range, however, the understanding of the SP phenomenon, let alone its use, is far from being complete. The cause is not only the recent lack of well-developed sources and detectors of THz radiation but the specific properties of THz SPs. These peculiarities are as follows: (1) SP refractive index exceeds that of light in air only by hundredths or even thousandths of a percent; (2) SP field extends into air over 100λ (here λ is the radiation wavelength in free space); (3) as small as hundredths to thousandths of the total SP field energy is transported in the metal, which results in small absorption of SPs and vast propagation length, reaching tens of centimeters or even meters.[6]

The most acute problem in developing THz SP techniques is efficient excitation of SPs. To transform bulk radiation into SPs one has to match their phase velocities and the tangential components of the wave vectors. In far IR range, the attenuated total reflection method does not work as the introduction of a prism into the SP field makes the incidence angle of optimum SPs excitation smaller than the critical one. That is why the only opportunity to excite THz SPs by bulk radiation is the diffraction of the latter on an object placed within the SP field in air.

The prime experiments on THz SPs excitation on gold were performed about 30 years ago with a silicon prism as a coupling element and a "whisky" laser generating at λ =118 μm.[7,8] Several astonishing facts were established: (1) the THz SP propagation length *L* was found to be one to two orders smaller as compared with the value of *L* calculated using the Drude model; (2) the efficiency of the SPs excitation turned to be as low as hundredths of a percent if not less; (3) SPs excitation was accompanied by production of strong idle beams of diffraction origin propagating along the surface, their fields thus superimposed on the SP field. Later on, similar experiments were done with gratings[9] and screen edges[10] used as coupling elements.

The problem of tiny transformation efficiency took a lot of effort to bridge it over. First it was suggested to cover the metal surface with a dielectric layer, which resulted in redistribution of the SP field from air into the metal and its adhesion to the surface.[11] Such SPs with compressed field may be excited with efficiency reaching tens of percent, but their propagation length collapses to millimeters, thus making almost all SP measuring techniques practically useless at THz frequencies.

A decade ago a revolution seemed to take place in the THz plasmonics. It was suggested to use the time-domain spectroscopy (TDS) approach and photoconductive antennas to excite and detect THz SPs.[12,13] It looked very attractive to span the whole THz band in one-procedure measurements determining both the SP amplitude and phase spectra. In their pioneer TDS experiments on SPs, Saxler *et al.*[14] made an attempt to establish the SP amplitude spectrum. Unfortunately, TDS measurements are not able to bring information on SP phase spectrum due to unpredictable phase shift on the input and output elements as well as to the 2π ambiguity in phase shift for the spectrum components.

As for the idle beams produced on the input element, a number of useful tricks to get rid of them were elaborated: starting from disposing in and out elements on conjugated facets of the sample (Ref. 8) to placing absorbing screens over the SP track (Refs. 10 and 13). Recently it was suggested to form a geodesic prism on the SP way to deflect SPs from the plane of incidence and thus separate overlapped

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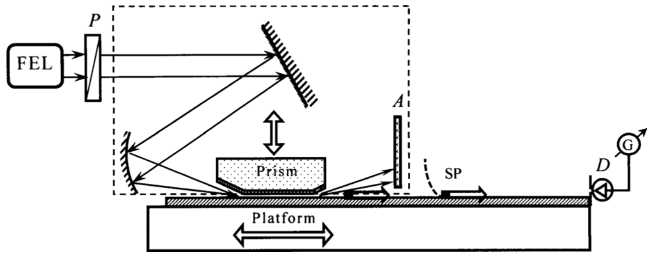

FIG. 1. Scheme of the attachment for measuring the THz SP propagation length L and the field decay δ in air.

surface and bulk waves running in the same direction.[15] Nowadays it may be stated that all the basic techniques and elements required for the THz SP metrology have been developed and the time for putting it into practice has come.

Here we discuss the possibility for determining the optical constants (or dielectric function) of real gold surfaces at THz frequencies by measuring the SP propagation length L and the SP field penetration depth (decay length) δ in air.

It is well known that solving the dispersion equation for SPs in a given guiding structure, one can determine two its parameters provided the complex refractive index $\kappa=\kappa' + j\cdot\kappa''$ (here j is the imaginary unit) of the surface wave is measured. Specifically, in case of a two-layer structure composed of a plane metal surface and adjacent air, the equation having the following form:

$$\kappa = \sqrt{\frac{\varepsilon_1 \cdot \varepsilon_2}{\varepsilon_1 + \varepsilon_2}} \quad (1)$$

(where $\varepsilon_1=\varepsilon_1'+j\varepsilon_1''$ and $\varepsilon_2=\varepsilon_2'+j\varepsilon_2''$ are the dielectric functions of the metal and air, accordingly) may be resolved relative to ε_1' and ε_1''.

So, the only problem in the THz SP refractometry of real conducting surfaces is the determination of κ' and κ''. A number of interferometric and noninterferometric techniques for determining both parts of κ have been developed lately.[16] All of them are based on the particular features of THz SPs, specifically the ability of such SPs to propagate over macroscopic distances and their field to penetrate deep into air. The technique used in the experiments performed is briefly described below.

To determine κ'' we measured the value of L related to κ'' by $L=(2k_o\times\kappa'')^{-1}$, where $k_o=2\pi/\lambda$, using the acknowledged "two-prism method," implying measurements of the SP intensity at two points or more along the track.[3]

To determine κ' we realized the noninterferometric technique[16] based on two facts: (1) at THz frequencies, the magnitude of SP decay length δ can be measured with a photodetector array or a single moveable detector; (2) the value of δ depends on κ as

$$\delta=[k_o\cdot \mathrm{Re}(\sqrt{\kappa^2-\varepsilon_2})]^{-1}. \quad (2)$$

With regard to the relation between L and κ'', one can solve Eq. (2) relative to κ' and obtain the following formula:

$$\kappa' = \frac{k_o^2\delta^2\kappa''\varepsilon_2''}{2\cdot[1+k_o^2\delta^2\cdot(\kappa'')^2]} + \sqrt{\frac{1}{1+k_o^2\delta^2\cdot(\kappa'')^2} \times \left\{\frac{1}{k_o^2\delta^2} + (\kappa'')^2 + \varepsilon_2' - \frac{k_o^2\delta^2\cdot(\varepsilon_2'')^2}{4\cdot[1+k_o^2\delta^2\cdot(\kappa'')^2]}\right\}}. \quad (3)$$

Figure 1 sketches the device we used in the experiments. The collimated monochromatic THz radiation produced by the free-electron laser (FEL) passes through rotatable wire polarizer P and on reflection from the plane and cylindrical mirrors is focused at the entry mouth of the element transforming bulk radiation into SPs. The element was elaborated by Grischkowsky and co-workers[17] and is characterized by high transformation efficiency. It is a bar (prism) with two axial cuts of the lower facet. Both the cuts and the facet are covered with a nontransparent gold film. Brought close to the sample (a shaded box) the metallized base of the prism and the sample surface form a waveguide able to lead modes of the TM type. These modes (the single-mode regime—preferable) diffract at the exit mouth of the element and are partially converted into SPs. The diffracted bulk waves are blocked by absorber A while the SPs slip under it. Detector D, fixed at the edge of the sample and provided with an aperture, indicates the arrival of the SPs. To change the distance run by the SPs the sample is installed on a computer-controlled, horizontally moving platform. Detector D is able to move in the plane of incidence in order to measure the SP field distribution in air.

The sample was a firm aluminum plate (40 mm wide and 250 mm long) with optically polished top surface covered with a thermally evaporated 1 μm thick gold film. The input element was made of glass in the form of prism with the right-angle box cross-section, the 40×40 mm^2 top base and the 20×40 mm^2 bottom base; the cut angles of the bottom base were 20° and 8° for the entry and exit mouths, accordingly. A wadding sheet disposed at 35 mm from the prism and raised 3 mm over the sample was used as absorber A. The intensity of the SP field was measured by a Golay cell with optical sensitivity of 10^5 V/W.

The experiments were performed at the Novosibirsk FEL (NovoFEL).[18] The measurements were performed in the air atmosphere with the absorption coefficient $\alpha=2k_ok_2=0.173$ m^{-1} (here k_2 is the absorption index of air) at $\lambda=130$ μm. To make sure that we dealt, namely, with THz SP the following tests were used:

(1) Polarization change from p to s type led to disappearance of the signal produced by the detector;
(2) Measured decay of the wave field intensity both along the surface and the normal to it proved to be exponential;
(3) An approximate permanence of the photosignal as a standard paper strip put on the surface and crossing the surface wave track at the right angle was moved along the track.[19]

Figures 2 and 3 present results of the measurements. The vertical axes correspond to normalized signals U/U_{max} produced by the detector while the distance a (run by the SPs) and the span z from the sample surface refer to the x coordinates, respectively. The exponential character of the dependences is obvious, though some interference oscillations could be distinguished in both graphs if one connects the experimental points without the approximations. It means that absorber A failed to block all the diffracted bulk waves and that some extra measures should be undertaken to clear the diffractional noise from the SP field.

According to the measured dependences of the SP field intensity on a and z, it decreases by a factor of $1/e$ at the distance $L=38.4$ mm along the SP track and at the distance

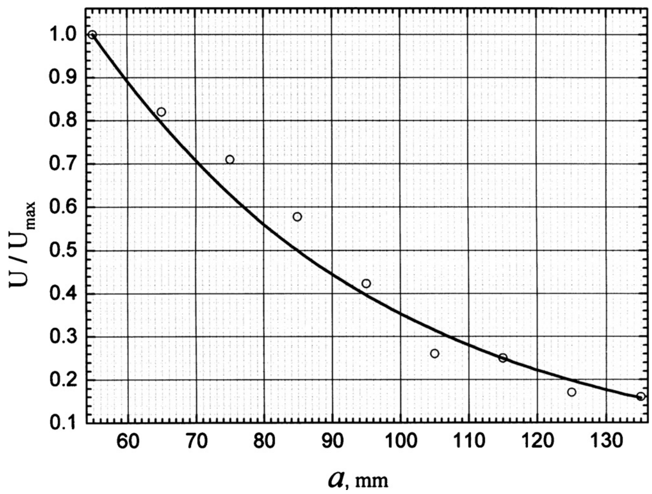

FIG. 2. Measured dependence of the normalized SP field intensity on the distance *a* run by the wave along the gold–air interface.

δ=1.3 mm along the normal to it. Substituting the magnitude of L in $L=(2k_o\times\kappa'')^{-1}$, we calculated $\kappa''=2.7\times10^{-4}$. After that we put the values of κ'' and δ in Eq. (3) and computed the corresponding value of $\kappa'=1.000\,657$. Taking into account the value for α in air and the fact that the refractive index of air n_2 at THz frequencies equals 1.000 272 6,[20] we found the air permittivity $\varepsilon_2=\varepsilon_2'+j\varepsilon_2''=(n_2+jk_2)^2=1.000\,545+j\times0.000\,003\,6$. Finally, substituting the values of κ', κ'', and ε_2 in Eq. (1) and solving it relative to ε_1, we determined the permittivity of the evaporated gold surface $\varepsilon_1=-877+j\times610$.

To check the validity of the procedure for ε_1 estimation, we compared values of the relation δ/L composed of the experimental results and those calculated using the approximate formulae for δ and L given in Ref. 21. The first value equals 0.034 and the second—0.025, that is 25% mismatch.

Note that accounting for the absorption in air does not influence the results very much. For example, if we assume α=0 in the foregoing calculations, then $\varepsilon_1=-875+j\times615$. Inaccuracy in measurements of L has a small effect on the result of calculations too: 1% deviation in L involves 0.5% changes both in ε_1' and ε_1''. But inaccuracy in δ measurements is much more significant: 1% deviation in δ brings about 0.5% change in ε_1' and nearly 2% in ε_1''.

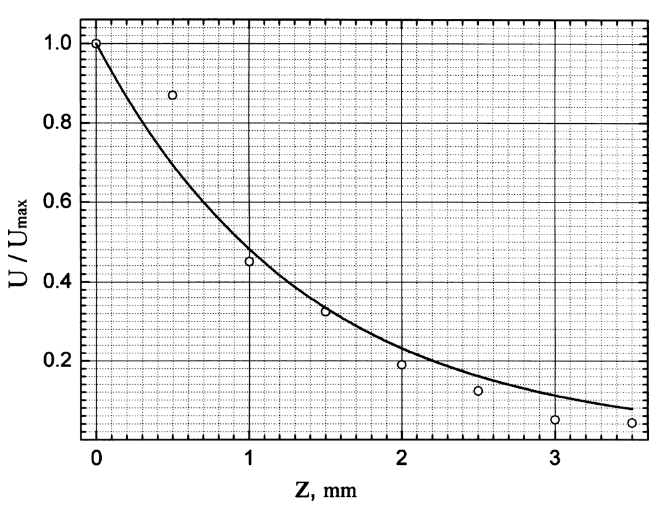

FIG. 3. Profile of the normalized SP field intensity along the *z* axes, normal to the gold–air interface, measured at the distance of 13.5 cm from the input element.

The result obtained for ε_1 is very far from that calculated by the Drude model for metal permittivity: with the damping frequency ν_τ=215 cm^{-1} and the plasma frequency ν_p=728 00 cm^{-1} for gold[22] we get $\varepsilon_1'=-101\,641$ and $\varepsilon_1''=284\,090$ at $\lambda=130$ μm. The measurable quantities L and δ for SPs on a gold sample with such ε_1 equal 126 cm and 4.8 mm, respectively. The difference in the experimentally determined and theoretical values for ε_1' and ε_1'' is striking at first sight, but in our opinion it reflects the difference in the optical properties of surface and bulk regions of gold, let alone that it was not crystal but thermally evaporated in our case.

In conclusion, we would like to underline that the method suggested is only a way to determine the permittivity of metallized surfaces at THz frequencies. Particular devices implementing the method still have to be developed, built, and tested.

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