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TERAHERTZ DISPERSIVE SPECTROSCOPY FOR THIN-FILM STUDY VIA SURFACE-PLASMON – BULK WAVE INTERFERENCE [*]

A new technique for terahertz (THz) dispersive spectroscopy of thin films employing surface plasmons (SP) has been developed. The technique is based on the SP’s complex refractive index κ strong dependence on the transition layer optical constants and employs interference in parallel beams of bulk and surface waves. It is remarkable for its accuracy and enables investigators to determine both parts of κ in one measuring procedure. Devices implementing the method may be whether of static or dynamic character; the latter requires measuring time equal at least to one pulse duration.

Keywords: surface plasmons, terahertz radiation, far infrared, dispersive spectroscopy, thin films, interference, free-electron laser.

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ТЕРАГЕРЦОВАЯ ДИСПЕРСИОННАЯ СПЕКТРОСКОПИЯ ДЛЯ ИССЛЕДОВАНИЯ ТОНКИХ ПЛЕНОК С ПОМОЩЬЮ ИНТЕРФЕРЕНЦИИ ПОВЕРХНОСТНОГО ПЛАЗМОНА С ОБЪЕМНОЙ ВОЛНОЙ

Разработан новый метод терагерцовой дисперсионной спектроскопии тонких пленок, использующий поверхностные плазмоны. Метод основывается на сильной зависимости комплексного показателя преломления κ поверхностного плазмона от оптических констант переходного слоя и использует интерференцию в параллельных пучках объемной и поверхностной волн. Метод является очень точным и позволяет определять обе части κ за одно измерение. Устройства, использующие данный метод, могут быть статическими и динамическими, причем последние требуют времени измерения, по крайней мере, равное одному импульсу повторения.

Ключевые слова: поверхностные плазмоны, терагерцовое излучение, дальнее инфракрасный диапазон, дисперсионная спектроскопия, тонкие пленки, интерференция, лазер на свободных электронах.

Introduction

Dispersive spectroscopy (DS), sometimes called “dispersion” or “dielectric” spectroscopy, establishes the dielectric properties of a medium as a function of frequency, in other words it determines frequency dependences of optical constants’ (refractive n and absorption k

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indexes) as a result of amplitude-phase measurements employing a wide-band continuous or tunable monochromatic radiation source [1].

A few decades ago a new powerful method for optical study of conducting surfaces was developed [2]. The method is based on generation of surface plasmons (SP), a kind of surface electromagnetic waves, by probing radiation. SP field has its maximum at the sample's surface and decreases exponentially with moving away from it. This is the main reason why SP characteristics (propagation length L, phase velocity V_{ph}, and air penetration depth δ) are very sensitive to optical properties of the surface and its transition layer. Having determined the SP complex refractive index $\kappa = \kappa' + i\cdot\kappa''$, employing the measured $L = (2k_o\cdot\kappa'')^{-1}$ and $V_{ph} = C/\kappa'$ (here $k_o = 2\pi/\lambda$, λ - wavelength and C – speed of the radiation in free space), one can calculate two unknown parameters of a film at the surface or optical constants of the metal substrate. Therefore SP are widely used in studying surface of metals as well as for their refractometry, bringing good results in the visible and middle infrared (IR) spectral ranges.

As for the far IR spectral range, specifically for the terahertz (THz) region, SP characteristics are very similar to those of a plane wave in air (with a refractive index n): the difference $(\kappa'-n) < 10^{-4}$, L amounting to meters, and δ reaching centimeters [3]. Due to these peculiarities, the process of SP excitation at a clear metal surface is rather embarrassing, which results in very low transformation efficiency (from hundreds to thousandths of a percent) of bulk radiation in SP. On the other hand, the SP features at THz frequencies stipulate unacceptably low measurement accuracy both for L and V_{ph}.

The situation reverses if we cover the sample surface with a layer that results in redistribution of the SP field from air into the metal. This results not only in rising transformation efficiency, but in increase of κ' and decrease of L as well as makes their measurement accuracies tolerable and the SP spectroscopy technique in a whole efficient at THz frequencies.

In this paper we discuss effective technique for determining κ' and κ'' via THz SP and bulk wave (BW) interference. Mastering this technique is important for the following reasons: 1) there is no other optical methods of investigating films with thickness $d << \lambda$ at THz frequencies; 2) reflectometry and ellipsometry practically cannot be used for spectroscopic study of conducting surfaces and their transition layers due to the very high reflectivity of metals in the far IR.

Surface-plasmon – bulk wave interferometer

The principle idea of interferometric SP spectroscopy described in [4] can be realized with a modified Michelson interferometer where monochromatic radiation in one of the shoulders passes part of its path in the form of SP accumulating information about the sample surface. The information is contained in the interference picture formed by two bulk waves: the reference one and the wave produced by the SP due to diffraction at the sample's edge. However, accuracy of this method was found to be insufficient as the beams interfere at a large angle, making the period of the pattern comparable with the wavelength.

We have developed a simpler and more effective scheme of THz SP dispersive spectrometer, involving interference in parallel instead of converging beams: one of which is the SP beam itself, while the other is a bulk radiation produced on the matching element when transforming the incident light into the SP.

The interferometer (Fig.1) functions as follows. By mirrors 2 and 3, radiation of source 1 is directed towards the edge of screen 5, spaced from the specimen 4 plane surface by a distance h, controllable in the limits from 5λ to 20λ. Due to diffraction, the radiation is partially transformed to SP and BWs, propagating at various angles from the surface. Among this set of BWs there is a wave with a wave vector parallel to the surface and field overlapping with the SP field. The BW and SP run along the surface with different phase velocities since κ' is larger than the BW refractive index n in air 6.

As a result of the Joule loses, the SP intensity decreases exponentially with the absorption factor $\alpha = k_o\cdot\kappa''$ (here $k_o = 2\pi/\lambda$). Having covered the same distance x, the BW and SP meet detector 7 (on a moveable platform 8) and acquire phase incursion differing in value by the magnitude $\Delta\varphi = k_o x\cdot(\kappa'-n)$. Being coherent, the BW and SP interfere with each other and

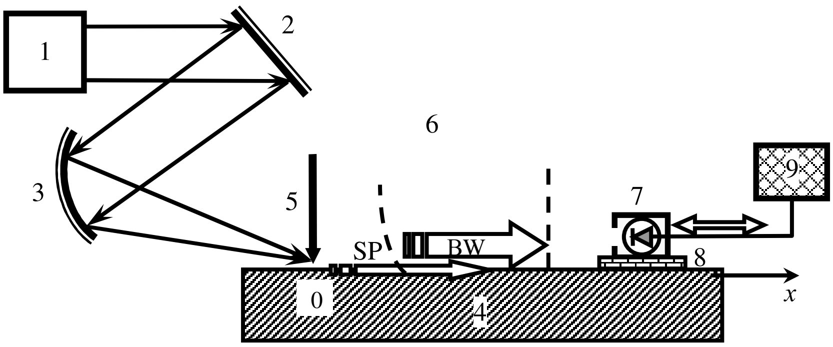

Fig. 1. Scheme of the SP interferometer employing interaction of surface and bulk beams

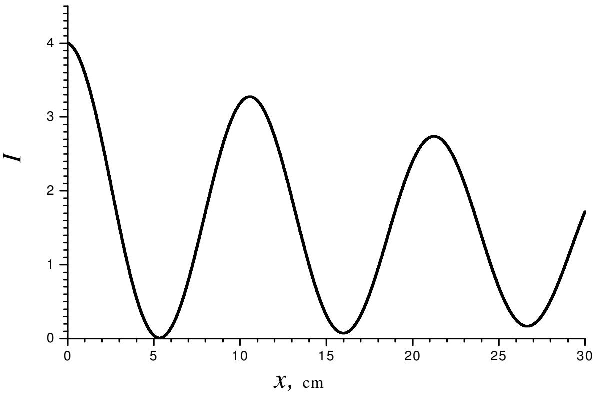

Fig. 2. Interferogram for the structure "*Al* –*Ge* layer 0.7 μm thick - air" at λ=100 μm

illuminate the detector's sensitive element with the intensity I described by the expression:

$$I(x) = I_1 + I_o \cdot exp(-\alpha \cdot x) +$$
$$+2 \cdot \sqrt{I_1 \cdot I_2 \cdot \exp(-\alpha \cdot x)} \cdot \cos(\Delta\phi), \qquad (1)$$

here I_1 is the BW intensity, independent on the distance x; I_o is the SP intensity right under screen 5 when x=0.

The period Λ of the registered interference pattern (interferogram) is constant and linked to κ′ by the evident formula:

$$\kappa' = n + \lambda/\Lambda. \qquad (2)$$

The SP absorption index κ″ can be calculated by putting the intensity values measured in two different interferogram maxima in the following formula[1]:

$$\kappa'' = 2 \cdot \ln\left(\frac{\sqrt{I_{m1}} - \sqrt{I_1}}{\sqrt{I_{m2}} - \sqrt{I_1}} \right) \Big/ \left[k_o \cdot (x_2 - x_1) \right], \qquad (3)$$

here x_1 and x_2 are the coordinates of corresponding maxima, $x_2 > x_1$.

On putting the found values of κ′ and κ″ in the SP dispersion equation for a three-layer structure [2], unit 9 computes two parameters of the structure: either both the thickness and refractive index of the transition layer or the complex dielectric permittivity of its material. Note that the contrast of the interferogram can be controlled by changing the distance h from the screen edge to the specimen surface in the limits from 5λ to 20λ.

To illustrate the technique let us consider the following example. Suppose we have to determine the dielectric permittivity ε_{Ge} of a 0.7 μm thick germanium (*Ge*) layer, deposited on an aluminum (*Al*) substrate at λ=100 μm using the method. Assume that the screen's position ensures $I_1 = I_o$, i.e. the intensity of the BW propagating parallel to the surface equals the SP intensity under the screen. The surrounding medium is air (n=1.00027). The calculated dependence $I(x)$ for the interferogram in this case is depicted in Fig.2. In the calculations we used the Drude model for *Al* dielectric permittivity.

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[1] Formula derivation is presented in the Appendix.

From the graph presented it follows that: 1) Λ=10.675 cm, which according to (2) corresponds to κ'=1.00121; 2) the resulting intensities in the first I_{m1} and the second I_{m2} maxima, reached at the distances x_1=10.565 cm and x_2=21.240 cm, are equal to 3.275 and 2.739, accordingly. Putting the values of I_{m1}, I_{m2}, x_1 and x_2 into (3), we get $\kappa''=6.3\cdot10^{-5}$.

At the final stage of the execution procedure the SP dispersion equation for a three-layer structure is solved relatively to ε_{Ge}. Thus in the example considered we obtain that *Ge* permittivity at λ=100 μm equals to ε_{Ge}= 16+$i\cdot$0.008.

Having done similar measurements and calculations for other λ one can determine ε_{Ge} in the whole THz range. Note that there is still no other optical method able to determine thin layer spectra at THz frequencies.

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Appendix

Suppose we have measured intensity I_{m1} and I_{m2} in two maxima of the interference pattern corresponding to distances x_1 and x_2 run by the SP. With regard to the fact that for maxima $\Delta\varphi = 2\pi b$ (here b – is an integer) these intensities in accordance with formula (3) may be described as follows:

$$I_{m1} = \sqrt{I_1 \cdot I_{21}} + 2\, I_1 \cdot I_{21}$$

and

$$I_{m2} = I_1 + I_{22} + 2\sqrt{I_1 \cdot I_{22}}\,,$$

here I_1 – is intensity of the bulk wave, I_{21} and I_{22} – are intensities of the SP at coordinates x_1 and x_2.

Solving these equations relatively I_{21} and I_{22} we get:

$$I_{21} = \left(\sqrt{I_{m1}} - \sqrt{I_1}\right)^2$$

and

$$I_{22} = \left(\sqrt{I_{m2}} - \sqrt{I_1}\right)^2.$$

In view of the exponential SP field decay we can express I_{22} through I_{21} on assumption that $x_1<x_2$: $I_{22} = I_{21}\cdot exp(-\alpha\cdot\Delta x)$, here $\alpha = k_o\cdot\kappa''$ – is the SP's absorption coefficient, $\Delta x = x_2 - x_1$. Wherefrom it follows that: $\alpha\cdot\Delta x = ln\left(I_{21}/I_{22}\right)$. Substituting the expressions for I_{21}, I_{22} and α in the last equation we get the required formula (3).

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