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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 $\kappa$ 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 $\kappa$ in one measuring procedure. Devices implementing the method may be either 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 ПЛЕНК С ПОМОЩЬЮ ИНТЕРФЕРЕНЦИИ
ПОВЕРХНОСТНОГО ПЛАЗМОНА С ОБЪЕМНОЙ ВОЛНОЙ

Разработан новый метод терагерцовой дисперсионной спектроскопии тонких пленок, использующий поверхностные плазмы. Метод основывается на сильной зависимости комплексного показателя преломления $\kappa$ 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 $\kappa$ 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 плазмы, 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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index) 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  ) away from it. This is the main reason why SP face and decreases exponentially with moving away. This results not only in rising transformation distribution of the SP field from air into the metal. Therefore SP are widely used for the terahertz (THz) region, SP characteristics are very similar to those of a plane wave in air (with a refractive index ) or tunable monochromatic radiation source [1].

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  and decrease of  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  and  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  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 , 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  is larger than the BW refractive index  in air 6.

As a result of the Joule loses, the SP intensity decreases exponentially with the absorption factor  (here  is the 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.

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illuminate the detector’s sensitive element with the intensity \( I \) described by the expression:
\[
I(x) = I_1 + I_o \cdot \exp(-\alpha \cdot x) + 
+ 2 \cdot \sqrt{I_1 \cdot I_o} \cdot \exp(-\alpha \cdot x) \cdot \cos(\Delta \phi),
\]
(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 \( \Lambda \) of the registered interference pattern (interferogram) is constant and linked to \( \kappa' \) by the evident formula:
\[
\kappa' = n + \lambda / \Lambda.
\]
(2)

The SP absorption index \( \kappa'' \) can be calculated by putting the intensity values measured in two different interferogram maxima in the following formula\(^1\):
\[
\kappa'' = 2 \cdot \ln \left( \frac{\sqrt{I_{m1}} - \sqrt{I_1}}{\sqrt{I_{m2}} - \sqrt{I_1}} \right) / \left[ k_o \cdot (x_2 - x_1) \right],
\]
(3)

\(^1\) Formula derivation is presented in the Appendix.

On putting the found values of \( \kappa' \) and \( \kappa'' \) 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 \( 5 \lambda \) to \( 20 \lambda \).

To illustrate the technique let us consider the following example. Suppose we have to determine the dielectric permittivity \( \varepsilon_{Ge} \) of a 0.7 \( \mu \)m thick germanium (Ge) layer, deposited on an aluminum (Al) substrate at \( \lambda = 100 \mu \)m 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.
From the graph presented it follows that: 1) \( \Lambda=10.675 \text{ cm} \), which according to (2) corresponds to \( \kappa' = 1.00121 \); 2) the resulting intensities in the first \( I_{m1} \) and the second \( I_{m2} \) maxima, reached at the distances \( x_1=10.565 \text{ cm} \) and \( x_2=21.240 \text{ cm} \), 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 \( \kappa'' = 6.3 \times 10^{-5} \).

At the final stage of the execution procedure the SP dispersion equation for a three-layer structure is solved relatively to \( \varepsilon_{Ge} \). Thus in the example considered we obtain that \( \varepsilon_{Ge} \) permittivity at \( \lambda=100 \mu \text{m} \) equals to \( \varepsilon_{Ge} = 16+i \cdot 0.008 \).

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

References


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:

\[
I_{m1} = \sqrt{I_1 \cdot I_2} + 2I_1 \cdot I_2
\]

and

\[
I_{m2} = I_1 + I_2 + 2\sqrt{I_1 \cdot I_2},
\]

here \( I_1 \) – is intensity of the bulk wave, \( I_2 \) and \( I_2 – \) 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: \( \alpha \cdot \Delta x = \ln(I_{21} / I_{22}) \).

Substituting the expressions for \( I_{21}, I_{22} \) and \( \alpha \) in the last equation we get the required formula (3).