Measurement of the complex refractive index of liquids and biological substances in the terahertz range at the NovoFEL facility

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***Abstract*—Terahertz ellipsometric measurement system has been commissioned with the Novosibirsk free electron laser being a source of monochromatic radiation. The highest precision of {ψ, Δ} ellipsometric parameters have been achieved for the THz range (0.3° for ψ, 0.01 for cos(Δ)). Complex refractive index of various liquids has been measured in the THz range using the ellipsometer with a silicon-prism internal reflection system. Since the characterization of biologically important highly absorbing substances is one of the important applications for the THz ellipsometry, the measurement technique has been optimized both theoretically and experimentally for water solutions. Precision of the measurement of the absolute values of the real and imaginary parts of the refractive index (n, k) equal to 0.01 is achieved experimentally.**

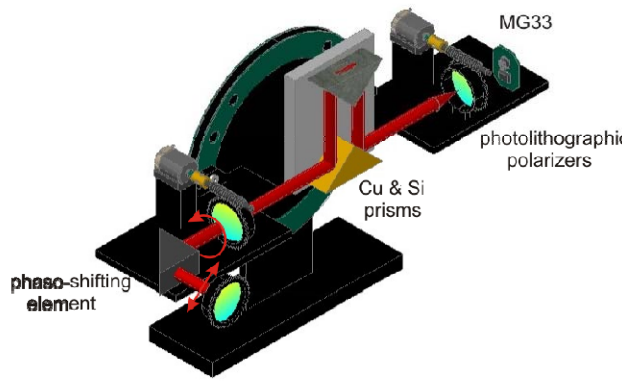

Fig. 1. Ellipsometric measurement system with an attenuated total reflection system.

I. Introduction

Study of liquid sample characteristics is very important for biological and medical applications. Nowadays there is no reliable equipment for measurement absolute values of the complex refractive index of liquids in the THz range. Ellipsometry is generally known as the most precise and sensitive technique for measurement of optical properties of substances. Most of biological substances contain water, which has a strong absorption over the whole THz range. For the purpose of study highly absorbing media, we have developed the ellipsometric system with a silicon-prism internal reflection system.

II. Experimental setup

Novosibirsk free electron laser (NovoFEL) is used as a source of monochromatic THz radiation [1]. Wavelength of NovoFEL can be smoothly tuned within the range 90 - 240 μm. All measurements were carried out at the wavelength of 130 μm. The measurements of the ellipsometric parameters Ψ and Δ are carried out using a static photometric algorithm in the multiangle analyzer mode [2]. A precision of polarizing element settings of about one arc minute was obtained in both setups. The rotation of the analyzer with a fixed position of the polarizer (P = ± 45 deg) was performed in a "start-stop" mode: rotation of the analyzer about a given angle, stop, and pick-up of a signal from the photodetector. At each analyzer position, 512 cycles (~ 5 s) were performed and the signal was averaged, for reduction of random errors. A pyroelectric detector MG-33 coupled to a lock-in amplifier was used for signal measurements. The ellipsometric parameters are retrieved from the Fourier components at the double frequency of the signal dependence on the azimuth position of the analyzer I(A). High-resistivity silicon was chosen as a clean, chemically inert material for prism with a high refractive index (3.418) [3]. Prism was made in the form of an isosceles triangle with the base angle of 45 degrees. Phase shift Δ for the normal incidence is equal to 84.6, which allows us to use the second identical prism as an input phase-shifting element.

ATR spectroscopy is based on the phenomenon of total internal reflection in a prism with a high-refractive index n_0 (e.g. a prism made of silicon or germanium). Reflection at an angle larger than a critical one causes an evanescent wave propagating along the surface of the prism. Placement of a sample absorbing the evanescent wave "breaks" the total internal reflection. The penetration depth depended on the radiation wavelength, the angle of incidence and the indexes of refraction for the ATR crystal and the medium being probed. In the THz region, the penetration depth in water is

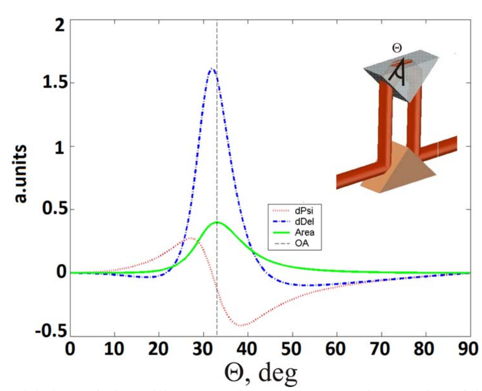

Fig. 2. Sensitivity of the ellipsometric system vs. the angle of incidence □. Shifts of the refractive and absorption indices by 0.01 each, relatively to initial values for water (n=2, k=0.49, see below), in the ψ parameter (red curve) and in the Δ parameter (blue curve). The green curve is the optimization for both parameters.

typically about 50 μm, which allowed us to explore the bulk properties of the sample. With the ATR technique applied to even highly absorbing objects, the reflection coefficient is not attenuated too much, and the reflected radiation magnitude remains measurable. ATR spectroscopy in the THz spectral range enables measuring substances with extinction coefficients from 10 to 10 000 cm^{-1}. It is worth noting that the effect of scattering in the ATR spectra is absent much lower that in visible region, which allows us to explore powder and nonhomogeneous samples.

In Fig. 2 we present the results of the optimization of the ellipsometric system with the internal reflection prism. The highest response in ψ and Δ to changing of optical properties of substances is achieved at the angle of incidence of 33 degrees. Thus, precision of the measurements of the ellipsometric parameters equal to 0.3° for ψ and 0.01 for cos(Δ) is achieved, that allow us to measure absolute values of the refractive index and the absorption coefficient for diluted water solutions with the precision of 0.01.

Optical constants were measured for a number of liquids (Fig. 3). For distilled water the complex refractive index was measured to be $N = 2.00 - i \cdot 0.49$. The refractive indices for decan and ethanol, which are the versatile solvents, was $N = 1.46 - i \cdot 0.03$ and $N = 1.49 - i \cdot 0.12$, respectively. Complex refractive index of dry blood and blood serum of healthy people and people with diseases varies from $N = 1.81 - i \cdot 0.31$ to $N = 1.88 - i \cdot 0.35$.

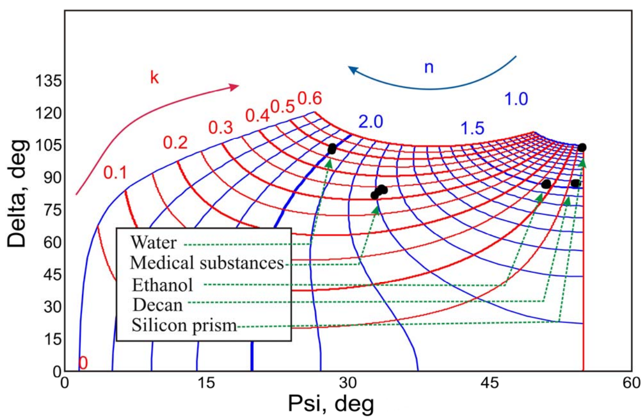

Fig. 2. Calculated nomograms for ψ and Δ. The coordinate system (red and blue lines) is formed with isolines of the refractive and absorption indices. Dark dots present experimental data for the silicon prism and for decan, ethanol, water and dry blood and blood serum situated on the top surface of the silicon prism.

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