

Determination of Optical Characteristics of Nanocoatings by Lorentz Transformation Approach

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Lorentz transformation formalism from special relativity theory was applied to light interference in films with thicknesses of a few hundreds and a few tens of nanometers. In this analogy between two theories, the speed of light is an imaginary number multiplied to the film refractive index. The interference in the layers of carbon nanotubes on a copper film was proposed to be described as a trajectory in (1+1) or (3+1) spacetimes. By using spacetimes of different dimensions, characteristic trajectories were possible to connect with experimental spectral ellipsometric data. The shape of a trajectory was determined by the real and imaginary parts of the film refractive index and therefore was sensitive to the presence of absorption bands in thin film dispersive curves. The coatings in this approach were characterized by a circular trajectory with singular points for absorption bands. If the optical conductivity of a film had a nonzero slope, the resulted trajectory in (1+1) spacetime formed a spiral. Both spectral ellipsometric and scanning tunneling microscopy experiments have been performed for a few tens of nanometers carbon nanotube thin film on a copper film with two hundred nanometer thickness. The copper film functioned as a Fabry-Perot interferometer with carbon nanotube layer as an interferometer's mirror. Multiray interference in the copper layer significantly improved the accuracy of the carbon nanotube film optical conductivity curve reconstruction. The thickness of the nanotube coating and its dispersive curves have been found by performing analysis of spectral ellipsometric curves by inverse gradient descent method. The approach based on classical optical experimental techniques allowed to determine parameters of a few tens nanometers layer though its thickness was one order of magnitude less than visible light wavelengths. Scanning tunneling microscopy measurements with high spatial resolution up to 1 nm registered nanotube bundles on the surface of the coating.

Keywords: Nanocoating, Lorentz transformation, Multiray interference, Spectral ellipsometry, Scanning tunneling microscopy.

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1. INTRODUCTION

Multilayer thin film coatings have been successfully studied and practically used for half a century. The coatings are being applied broadly in optical industry in manufactured polarizers, dichroic elements, laser mirrors, antireflection coatings, etc. Recently, a large attention was drawn to study the properties of ultrathin films on metallic surface in frames of their possible practical application, for example, as sensor elements. However, in order to estimate optical properties of films with tens nanometers thickness we have to apply robust and effective methods for their optical dispersion curve calculations.

Theoretical formalism for light interference in the layers was very well developed [1] and basically allows us to connect the coating parameters and their optical properties. The theoretical approach is based on the solution of Maxwell equations for plane waves with boundary conditions on the layer's borders. Each layer is characterized by a matrix and the resulting interference of the coating is calculated by multiplication of these matrices. Because of the complicity of the procedure, the optical coating parameters cannot be presented as an analytical expression. This absence of an analytical expression for interference makes some problems technically difficult. For example, in order to determine thin film parameters (refractive indices, thicknesses) from optical and ellipsometrical curves we have to apply

numerical optimization techniques which only may provide solutions with errors.

An alternatively different approach for thin film optics is based on its similarity with special relativity formalism. An alternative approach was developed in [2] with simplification of the known formula for Fabry-Perot resonator for dielectric layers with quarter and/or half-wave thickness. The authors modeled multiple interface reflections at the layer boundaries as with a simple addition rule. In [3], the reflection coefficient for a planar layered structure was proposed to be obtained applying Einstein's addition theorem for parallel velocities. In this work, the special relativity results were considered as a more general law of physics with further possible use for quantum theory. A comparison between special relativity and optics of planar media was introduced with possible application to the solutions of Schrödinger equation for reflected wave functions.

In [4], the authors described scattering matrixes of multilayer systems as elements of a group, which is isomorphic to a restricted Lorentz group. (3+1) vector, which is equivalent to the Stokes vector, was introduced. The field variables of the vector represent an invariant space-time interval. The formalism of multiray interference in planar films is similar to the one from special relativity theory and could be presented as a characteristic vector evolution in (1+1), (2+1) or (3+1) space.

In [5], the authors introduced Rindler space for thin

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films or in other words generalized the idea of Lorentz transformation by taking into account gravitational effects with included acceleration into the model. In terms of interference in thin films, this approach makes an analogy to specific multilayer films with particular values of refractive indexes and films thicknesses. The approach is based on light waves electrodynamics in the Rindler space. More exactly a connection between Rindler coordinates and Maxwell's equations solutions with specific attention to the wave's behavior at the Rindlers horizon was found.

Potentially, the special relativity formalism for planar films could be used for practical problems of spectral ellipsometry of films with tens nanometers thickness. It is clear, the inverse problem of the ellipsometry of the films with thickness of the order of magnitude less than light wavelength requires extra efforts to detailing very low phase shift in the coatings. Usually the numerical solutions for film dispersion curves are un-

$$\begin{vmatrix} a_{11} & a_{12} \\ a_{21} & a_{22} \end{vmatrix} = 1/2 \begin{vmatrix} 1 & 1/U_{N+1} \\ 1 & -1/U_{N+1} \end{vmatrix} \left(\prod_{i=1}^N \begin{vmatrix} \cos(\Delta_i) & \frac{i \times \sin(\Delta_i)}{U_i} \\ i \times \sin(\Delta_i) \times U_i & \cos(\Delta_i) \end{vmatrix} \right) \begin{vmatrix} 1 & 1 \\ U_0 & -U_0 \end{vmatrix} = 1/2 \begin{vmatrix} 1 & 1/U_{N+1} \\ 1 & -1/U_{N+1} \end{vmatrix} \times \begin{vmatrix} b_{11} & b_{12} \\ b_{21} & b_{22} \end{vmatrix} \times \begin{vmatrix} 1 & 1 \\ U_0 & -U_0 \end{vmatrix}. \quad (1)$$

Here, U_i are parameters equal to $n_i/\cos(\theta_i)$ for p-light polarization or $n_i\cos(\theta_i)$ for s-polarization. Subscript indices 0 and $N+1$ in Eq. (1) correspond to the substrate and air, respectively. θ_i and Δ_i are refraction angles and phase shift in the layers. $\Delta_i = 2\pi n_i d_i \cos(\theta_i)/\lambda$, where n_i is the refractive index and d_i is the thickness of i -layer.

N matrices from Eq. (1) can be transformed to special relativity theory equations. For (1+1) spacetime, Lorentz transformation [6] can be written applying matrices b from Eq. (1):

$$\begin{bmatrix} z' \\ t' \end{bmatrix} = \begin{bmatrix} \frac{1}{\sqrt{1-\beta^2}} & -\frac{\beta \times c}{\sqrt{1-\beta^2}} \\ -\frac{\beta/c}{\sqrt{1-\beta^2}} & \frac{1}{\sqrt{1-\beta^2}} \end{bmatrix} \times \begin{bmatrix} z \\ t \end{bmatrix}, \quad c = i/U, \quad (2)$$

$$\beta = v/c = \sqrt{1 - \cos^2 \Delta}$$

Here v is the speed and c is the speed of light.

Lorentz transformation is defined as a linear transformation of the space which preserves quadratic form which can be presented for (1+1) or (3+1) spacetime as:

$$\begin{aligned} z^2 - c^2 t^2 &= const, \\ x^2 + y^2 + z^2 - c^2 t^2 &= const \end{aligned} \quad (3)$$

Based on the second postulate of relativity, Eq. (3) introduces the space-time interval. The light cone which is a line for (1+1) spacetime for "classical" case transforms to a circle for thin films because speed of light is an imaginary number for non-absorbing media (Eq. (2)). If the absorption dispersion curve has sharp absorption lines, the characteristic curve of the thin films determined by Eqs. (2) should have singular points. In a hypothetic case of imaginary refractive index, Eq. (3) solution is a straight line instead of a circle.

Let us consider (3+1) spacetime instead of (1+1)

stable – they are too much dependent on spectral ellipsometry measurement errors. The objective of this article is to apply special relativity theory formalism for determining thin film parameters (optical conductivity, thickness) from experimental ellipsometric data.

The main goal of this work consisted in application of Lorentz transformation approach for thin films for spectral ellipsometry tasks. Particularly we are interested in overall practical solving inverse ellipsometry task, e.g. to determine optical constants of carbon nanotube nanocoating on a copper layer based on spectral ellipsometry and scanning tunneling microscopy measurements.

2. THEORETICAL APPROACH

Interference of light with wavelength which propagates through N layers can be described by multiplication of $N+2$ matrices [1]:

spacetime in Eqs. (1) and (2). Matrices (a) and (b) from Eq. (1) allow to connect electromagnetic fields at substrate and air boundaries of the coating:

$$\begin{aligned} \begin{vmatrix} \tilde{E}_{N+1}^+ \\ \tilde{E}_{N+1}^- \end{vmatrix} &= \begin{vmatrix} a_{11} & a_{12} \\ a_{21} & a_{22} \end{vmatrix} \times \begin{vmatrix} \tilde{E}_0^+ \\ \tilde{E}_0^- \end{vmatrix} \\ \begin{vmatrix} \tilde{E}_{N+1} \\ \tilde{H}_{N+1} \end{vmatrix} &= \begin{vmatrix} b_{11} & b_{12} \\ b_{21} & b_{22} \end{vmatrix} \times \begin{vmatrix} \tilde{E}_0 \\ \tilde{H}_0 \end{vmatrix}. \end{aligned} \quad (4)$$

Here, superscript sign \sim denotes tangential projections of the fields and superscript indices "+" and "-" are for light waves which propagate in air-to-substrate and in substrate-to-air directions, respectively. The matrix describes reflection and transmission in films and describes total electric and magnetic vectors on the films boundaries. Electric and magnetic vectors from Eq. (4) are connected by equations [1]:

$$\begin{aligned} \tilde{E}_{N+1}^+ &= (\tilde{E}_{N+1} + \tilde{H}_{N+1}/U_{N+1}) \times 0.5, \\ \tilde{E}_{N+1}^- &= (\tilde{E}_{N+1} - \tilde{H}_{N+1}/U_{N+1}) \times 0.5. \end{aligned} \quad (5)$$

For (3+1)-dimensional spacetime similarly to [4] approach, we can write a vector which contains simultaneously electric field components from Eq. (4). Characteristic vector for this case can be derived from Eqs. (1), (4), (5) as:

$$\begin{bmatrix} x \\ y \\ z \\ ct \end{bmatrix} = \begin{bmatrix} i \operatorname{Im} \left(2\sqrt{\tilde{E}_{N+1}^+ \tilde{E}_{N+1}^-} \right) \\ i \operatorname{Re} \left(2\sqrt{\tilde{E}_{N+1}^+ \tilde{E}_{N+1}^-} \right) \\ \tilde{E}_{N+1} \\ \tilde{H}_{N+1}/U_{N+1} \end{bmatrix} = \begin{bmatrix} i \operatorname{Im} \left(2\sqrt{\tilde{E}_{N+1}^+ \tilde{E}_{N+1}^-} \right) \\ i \operatorname{Re} \left(2\sqrt{\tilde{E}_{N+1}^+ \tilde{E}_{N+1}^-} \right) \\ \tilde{E}_{N+1}^+ + \tilde{E}_{N+1}^- \\ \tilde{E}_{N+1}^+ - \tilde{E}_{N+1}^- \end{bmatrix} \quad (6)$$

As we can see, quadratic form $x^2 + y^2 + z^2 - c^2 t^2$ for Eq. (6) vector components is invariant and satisfies

Eq. (3). Similarly to Eq. (6), relationship is presented in [4] but for quadratic forms of electric field. It allowed introducing Stokes parameters for multilayer thin film system similarly to Muller matrix and Jones vector formalism for polarized light. We used electric and mag-

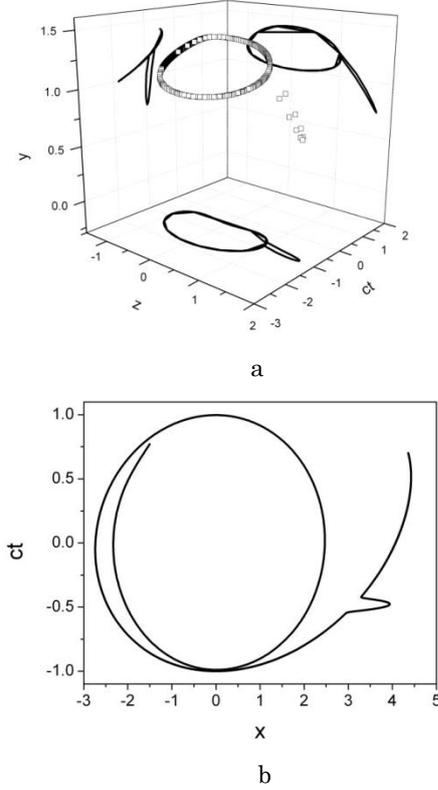


Fig. 1 – Calculated trajectories for 200 nanometers thickness films in a) (3+1) spacetime with refractive index $n + ik = 1.41 + i \cdot 0.02 \cdot \exp(-(\lambda - 335)^2 / 25)$, b) (1+1) spacetime with refractive index $n + ik = 1.41 + i \cdot (\lambda \cdot 0.01 + 0.02 \cdot \exp(-(\lambda - 335)^2 / 25))$

netic fields instead of intensities in Eq. (6) in order to formulate equations for (1+1) and (3+1) spacetimes with the same variables. Equation (6) simultaneously describes electric and magnetic vectors of incident planar wave as well as vectors of forward and backward moving light which completely portray multiray interference in the coating. The same variables in Eqs. (2) and (6) allow us to analyze simultaneously trajectories in (1+1) and (3+1) spacetime spaces. Introduction of equations for (3+1) space gives us an additional advantage of applying Lorentz transformation for experimentally measured ellipsometric parameters which are related to Eq. (6) vector components:

$$\begin{aligned} \cos \Delta &= \cos(\arctan(\frac{x_S}{y_S}) - \arctan(\frac{x_P}{y_P})) = \\ &= \frac{x_S x_P + y_S y_P}{\sqrt{(x_S)^2 + (y_S)^2} \sqrt{(x_P)^2 + (y_P)^2}} \\ \tan \psi &= \frac{(z_S - ct_S)(z_P + ct_P)}{(z_S + ct_S)(z_P - ct_P)}. \end{aligned} \quad (7)$$

Here, s and p indices describe s- and p-light polarization. As we can see from Eq. (7), ellipsometric parameters $\cos \Delta$ and $\tan \psi$ depend separately on pairs of (3+1) spacetime coordinates. A trajectory $\cos \Delta(\tan \psi)$ is a projection of the original four dimensional trajectory to a plane.

Thin film calculated trajectories in (1+1) and (3+1) spacetimes are presented in Fig. 1. As we can see, the trajectories have similar circular shape with sharp isolated singular points which are related to the spectral interval where the coating has absorption bands. If the absorption dispersion curve has a nonzero slope, the resulted characteristic trajectory is a spiral (see Fig. 2b), a curve with its curvature dispersion.

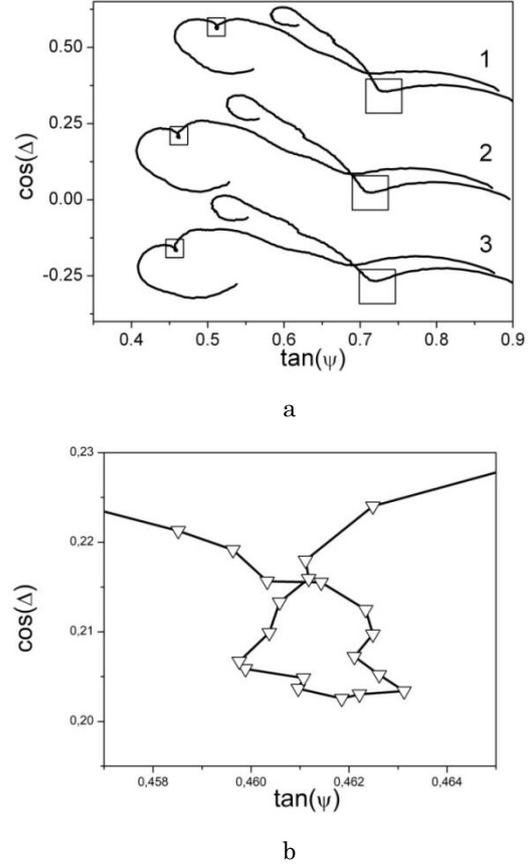


Fig. 2 – Experimental spectral ellipsometric curves. a) Singular points – acnodes (marked by large rectangles) for quartz covered by 200 nm copper film and loops (marked by small rectangles) for quartz covered by 200 nm copper film and a nanotubes film. Angles of incidence: 1 – 750, 2 – 700, 3 – 650. b) Magnified part of the curve 2 with a loop

3. EXPERIMENTAL

We carried out spectral ellipsometric measurements using Angstrom Advanced PHE 103 UV/VIS NIR Spectroscopic Ellipsometer which allowed us to perform measurements in 230-1100 nm spectral range. A microscope INTEGRA NT-MDT was used to conduct measurements in both atomic force microscopy (AFM) and scanning tunneling microscopy (STM) regimes. STM spatial resolution reached up to 1 nm and AFM – 40 nm. A sharp needle for STM measurements was fabricated from 0.5 mm Pt0.8Ir0.2 wire by mechanical cutting of its end. We performed our measurements in a regime

when STM setup supported a constant tunneling current through the needle, which was accomplished by tuning the sample position along the vertical direction. AFM measurements were performed in semicontact regime in 120 micron scan range. It allowed us to determine precisely the exact thickness of a deposited film by measuring the sample profile in the vicinity substrate/film substrate.

Copper films on quartz polished plates were thermally deposited in vacuum. The equipment was directly installed in the vacuum chamber which allowed us to control the copper temperature, deposition speed and substrate temperature. The substrates which have been used for the film deposition were carefully cleaned and polished quartz plates (the roughness was a few nm). The deposition time interval was chosen in order to reach a required film thickness, and the pressure in the vacuum chamber reached 10^{-5} Torr during the deposition process. 200 nm thick copper layer was deposited on a quartz plate. The thickness of the layer was precisely determined by performing AFM measurements with a linear XY stage having 120 microns scan range. Spectral ellipsometric measurements were made with a copper film sample and a copper film sample with deposited nanotubes. A carbon nanotube film was prepared similarly to the samples from [7]. In order to obtain large area of deposited from aqueous solutions thin film one can apply "doctor blading" technique [8].

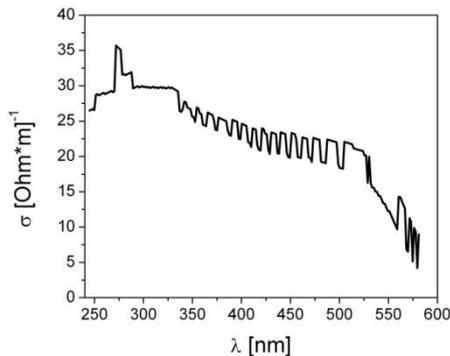


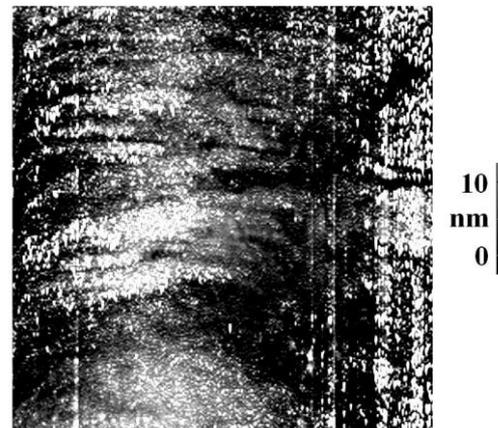
Fig. 3 – Reconstructed optical conductivity of carbon nanotubes of 45 nm thick film based on Fig. 2 measurement results

The results of the spectral ellipsometric measurements are presented in Fig. 2. We can see circular parts in the curves with singular points which mark the edge of an absorption band of copper film (marked by large rectangles) and copper film covered by the nanotube film (marked by small rectangles). The acnodes which are related to Cu absorption in 500 nm spectral region become less distinct if the copper layer is covered by thin nanotube film as well as circular parts of the curves for the samples with carbon nanotube film have larger diameter. The latter indicates more efficient multiray interference in Fabry-Perot interferometer due to an extra absorption carbon nanotube layer. The loops on the trajectories are related to nanotube film sharp absorption band in 280 nm spectral region. As a result, we have singular points on $\cos\Delta(\tan\psi)$ curve with sharp variations of the second order derivative there. Optical conductivity of the carbon nanotube was found by applying the inverse gradient descent method with the results which are presented in Fig. 3. A multi-variable

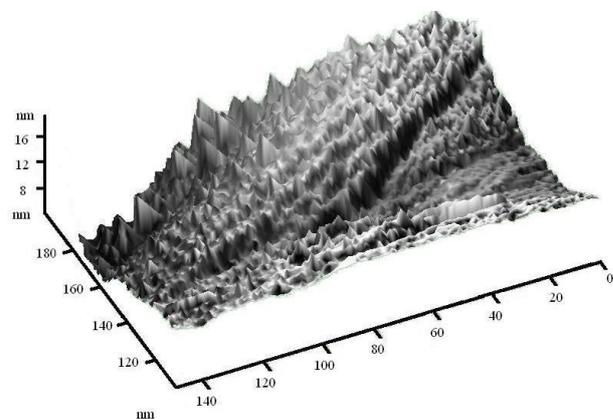
function to be optimized was taken with $\cos\Delta(\tan\psi)$ curve curvature as a part of the expression. The known

[9] relationship for function curvature $y'' / \sqrt{(1+(y')^2)^3}$

contains the first and second order derivatives, so the solution took into account both singular points on the curve and experimental curve shape. There are oscillations in Fig. 3 curve which seems to indicate the inverse gradient descent method calculation error. As we can see, the error margins are lower near 280 nm absorption band (which is related to the loops in Fig. 2 curves). The calculated thickness of the carbon nanotube film was found to be 45 ± 3 nm. The film thickness outside [42 nm; 48 nm] interval resulted in twice higher values of the oscillations margins.



A



b

Fig. 4 – Results of STM measurements of carbon nanotube film on a copper layer. Spatial resolution: a) 4 nm, b) 1 nm

Our attempts to deposit a few tens nanometers thickness carbon nanotube films on a transient substrate (quartz) and determine the optical film constants resulted in too huge calculation errors. Multiray interference was not very efficient for a sole a few tens nanometers thickness thin film because the light wavelength is more than one order of magnitude higher. Phase shifts in the film were much less than light wavelength and the film did not work as Fabry-Perot interferometer. Two thin films on a quartz substrate – thin

film (thin copper film in our case) and a nanoscale thickness thin film (carbon nanotube thin film) demonstrated noticeable interferometric patterns on dispersion ellipsometric curves. In this case, the nanoscale thickness thin film worked as a mirror on Fabry-Perot interferometer (200 nm thick copper layer). The thickness of the copper thin film was comparable with visible light wavelength and 45 nm carbon nanotube thin film effectively influenced the interferometer parameters.

The results of STM measurements are presented in Fig. 4. The surface of the sample is smooth (Fig. 4a) with average roughness of 5.2 nm. STM measurements with higher spatial resolution (Fig. 4b) reveal line-like patterns which resemble nanotube bundles on the surface. We tried to apply the inverse gradient descent method for the system quartz substrate/copper layer/ carbon nanotube layer/a few nanometers roughness layer in 50:50 hollows filling model. However, the extra roughness layer which took into account the sample surface topography resulted in practically no variations for the previously found optical conductivity dispersive curve (Fig. 3). It is known, the surface roughness can influence the optical properties of samples both significantly [10] or with small or even surprisingly negligible effect [11]. It means that different physical processes on the sample/air interface may be totally different, and as a result the surface roughness may or may not influence the reflected light parameters. In our case, multiray interference totally determined the ellipsometric parameters of the reflected light. A few nanometers roughness layer was too small comparing with two other film thicknesses (200 nm and 45 nm) from the point of view of interferometric processes in these two films.

4. CONCLUSIONS

Optical properties of thin films can be described by means of special relativity theory. The proposed theoretical formalism allowed us to relate in an apparent way the ellipsometric parameters of a thin film with the parameters of the relevant Lorentz transformation. We applied Lorentz transformation for light interference in absorbing coatings with a few tens and a few hundreds of na-

nometers thicknesses. A characteristic trajectory in two-dimensional or four-dimensional spaces was proposed to be used for a film representation and found to be circular shaped. A circle is a solution for space-time interval equation for thin films, and the circular trajectory was proposed to be taken as a characteristic curve for nonabsorbing thin film. Trajectories in a spacetime were shown to be connected with experimental spectral ellipsometric data by using Lorentz formalism in (3+1) spacetime. The characteristic curve shape robustly depends on the real and imaginary parts of the refractive index of the coating. Therefore, sharp absorption bands and a slope on the film's dispersion absorption curves result in transformation of the circle trajectory to a spiral with second derivative singularities which are visible as acnodes and loops on the experimental spectral ellipsometric curves.

The inverse gradient descent method taking the trajectory curvature as a function to be optimised can be used in order to find the films parameters. The approach allowed us to determine the optical conductivity and the thickness of a few tens of nanometers carbon nanotube film on a copper coating having thickness 200 nm. The copper film operated in this model as Fabry-Perot interferometer with carbon nanotube thin film as an interferometer mirror. Multiray interference in the interferometer significantly improved the accuracy of the carbon nanotube optical parameters reconstruction. Performed STM with 1 nm spatial resolution revealed nanotube bundles on the surface of the film allowing us to register the sample surface topography. The registered a few nanometers roughness of the upper nanotube layer did not influence significantly the calculated optical properties of the nanocoating.

In summary, lossless and absorbing thin films have different topological behavior of their characteristic trajectories. It plays a critical role for films' optical parameters reconstruction based on spectral ellipsometry data. Usually ultrathin films are not considerably noticeable by applying optical techniques because the films influence too low phase shift in the reflected visible light. Therefore the proposed approach may be critically important in spectral ellipsometry experiments on nanocoatings.

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Визначення оптичних характеристик нанопокриттів з використанням перетворення Лоренца

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Формалізм перетворення Лоренца зі спеціальної теорії відносності застосовувався до описання інтерференції світла в плівках товщиною в кілька сотень і декілька десятків нанометрів. У цій аналогії між двома теоріями швидкість світла є уявною одиницею, помноженою на показник заломлення плівки. Інтерференцію в шарах вуглецевих нанотрубок на мідній плівці було запропоновано описати, як траєкторію в (1+1) або (3+1) просторах. За допомогою просторів різних розмірностей ці траєкторії можна було поєднати з експериментальними спектральними еліпсометричними даними. Форма траєкторії визначається дійсними і уявними частинами показника заломлення плівки і тому чутлива до присутності смуг поглинання на дисперсійних кривих тонкої плівки. Покриття в цьому підході характеризується круговою траєкторією з особливими точками для смуг поглинання. Якщо оптична провідність плівки має ненульовий нахил, то отримана траєкторія в (1+1) просторі має форму спіралі. Для тонкої плівки вуглецевих нанотрубок з товщиною в декілька десятків нанометрів нанесеної на мідну плівку товщиною в двісті нанометрів були виконані виміри на спектральному еліпсометрі і на скануючому тунельному мікроскопі. Плівка міді функціонувала як інтерферометр Фабрі-Перо з шаром вуглецевих нанотрубок, які формували одно із дзеркал інтерферометра. Багатопротеневна інтерференція в плівці міді значно підвищила точність знаходження кривої оптичної провідності вуглецевих нанотрубок. Було знайдено товщину покриття та його дисперсійні криві, проводячи аналіз спектральних еліпсометричних кривих методом зворотного градієнтного спуску. Підхід, заснований на експериментальних методах класичної оптики, дозволив визначити параметри шару товщиною в декілька десятків нанометрів, хоча його товщина була на порядок меншою довжин хвиль видимого світла. Вимірювання на скануючому тунельному мікроскопі з високою просторовою роздільною здатністю до 1 нм зареєстрували джгути нанотрубок на поверхні зразка.

Ключові слова: Нанопокриття, Перетворення Лоренца, Багатопротеневна інтерференція, Спектральна еліпсометрія, Скануюча тунельна мікроскопія.