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OPTICS

Ellipsometric Technique for Determining in Situ the Absorption Coefficient of Semiconducting Nanolayers

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Abstract—An algorithm that makes it possible to solve the inverse problem of ellipsometry aimed at determining the absorption coefficient on the basis of a single-zone ellipsometric experiment during the growth of thin semiconducting films is developed and implemented. The technique is based on analysis of the variation of ellipsometric parameters Ψ and Δ directly during the growth. The algorithm is tested in synthesis of $Si/SiO_2/Si(100)$ and $Hg_{1-x}Cd_xTe$ structures.

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The problem of control over the physical properties of new nanomaterials has become topical due to the rapid development of nanotechnologies in recent years [1]. In particular, thin-film heterostructures with alternating semiconducting and magnetic layers are of considerable interest for spintronics [2]. Ellipsometry is one of the effective methods for studying such structures, which ensures sensitivity up to a single monolayer and at the same time does not destroy the sample under investigation [3, 4]. The basic principle of ellipsometry is the measurement of the polarization state of a light beam reflected from any reflecting system. In this case, ellipsometric angles Ψ and Δ are measured, which are primarily functions of the optical constants of the growing layers that carry information on their composition, crystal structure, and other properties of the material [5–8]. The optical constants can be determined from the results of continuous measurement of ellipsometric parameters during the synthesis of a layer (so-called growth curve) [8–11]. However, the accuracy of their determination in this case is often limited due to the roughness of the surface, possible formation of surface layers (oxide films, possible contamination of the surface, etc.), or other imperfections of the optical model [11].

As a rule, single-zone ellipsometric techniques (i.e., those performed for the same azimuthal position of optical elements) are used in continuous in situ measurements. This is due to the fact that it is sometimes impossible to change the positions of optical elements of the instrument during the film synthesis because the characteristic growth rate is commensurate with the speed of rotation of optical elements. Single-zone measurements are known to lead to unavoidable systematic errors associated with various imperfections of optical elements [12]. For this reason, the use of single-zone dependences of ellipsometric parameters for exact solution of the inverse problem is not always justified and may lead to considerable errors (especially in long-term experiments). In this case, more reliable information is contained in the relative variations of ellipsometric parameters observed during the growth [13]. It is analysis of relative variations of parameters Ψ and Δ that forms the basis of the technique for determining the absorption coefficient proposed here, which is not so sensitive to the experimental errors mentioned above.

Let us consider a monolayer model consisting of a substrate (medium 2), an optically homogeneous layer (medium 1), and vacuum (medium 0). The ellipsometric parameters for such a model as functions of layer thickness d are expressed by the following relation [3]:

$$\rho = \tan\Psi e^{i\Delta} = \frac{r_{01p} + r_{12p}X}{1 + r_{01}r_{12p}X}\frac{1 + r_{01s}r_{12s}X}{r_{01s} + r_{12s}X}, \qquad (1)$$

where $r_{ijp,s}$ are the Fresnel coefficients for the corresponding interfaces and different polarizations (p and s),

$$X = \exp\left(-\frac{4\pi i}{\lambda}d\sqrt{N_1^2 - \sin^2\varphi}\right),$$

$N_1 = n_1 - ik_1$ is the complex refractive index of the growing layer, and φ and λ are the angle of incidence and the wavelength of light. During synthesis of semiconducting layers on a semiconducting substrate, the

optical contrast at interface 12 is much weaker than at interface 01; therefore, we can assume that $r_{12} \ll r_{01}$. In this case, Eq. (1) can be expanded into a series in the small parameter (confining analysis to the second-order terms inclusively) and to obtain the following expression for the growth curve:

$$\rho = \rho_1(1 + A_1X + A_2X^2) \quad (2a)$$

or, in a more convenient form,

$$\rho - \rho_1 = \rho_1A_1X + \rho_1A_2X^2, \quad (2b)$$

where ρ_1 is the complex ellipsometric parameter corresponding to an infinitely thick (opaque) layer ($d \longrightarrow \infty$), and $A_1 \gg A_2$ are the expansion coefficients depending on the optical constants of the layer and the substrate. The term in Eqs. (2), which is linear in X, described a logarithmic spiral [3] that rolls up to point ρ_1 in the complex ρ plane with quasi-period $d_0 = \dfrac{\lambda}{2\sqrt{N_1^2 - \sin^2\varphi}}$ (Fig. 1).

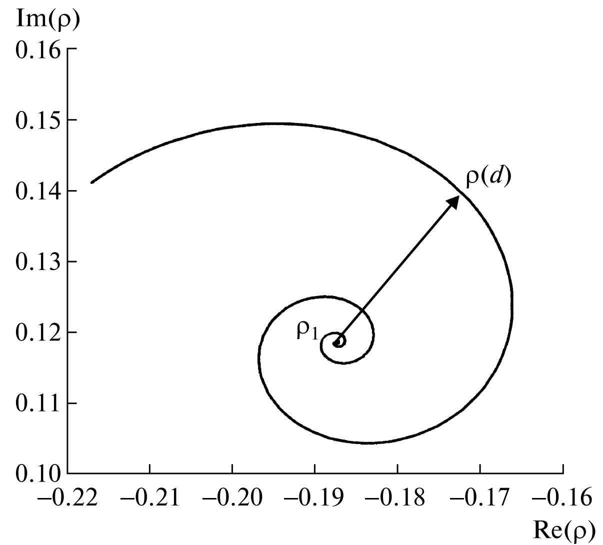

Fig. 1. Calculated growth curve for a system consisting of a HgCdTe ($x = 0.3$) layer on the CdTe substrate, depicted in the plane of the complex ellipsometric parameter ρ.

The radius vector describing the current position of ρ relative to ρ_1 exponentially decays with increasing d. The term proportional to X^2 gives a small correction to the exponential dependence of the radius vector, which is periodic with period $d_0/2$. Taking logarithms of the moduli of the left- and right-hand parts of expression (2b) and confining analysis to the linear approximation, we obtain

$$\ln|\rho - \rho_1| = \ln(\rho_1A_1) + \frac{4\pi}{\lambda}d\,\mathrm{Im}\sqrt{N_1^2 - \sin^2\varphi}. \quad (3)$$

For semiconductors, we have $k_1 \ll n_1$; therefore, we can disregard in the radicand the terms quadratic in k_1 as well as the term $\sin^2\varphi$, which is also much smaller than n_1^2. As a result, we obtain the dependence for the logarithm of the radius vector, which is linear in d:

$$\ln|\rho - \rho_1| = \ln(\rho_1A_1) - \frac{4\pi k_1}{\lambda}d. \quad (4)$$

The inclusion of the second order of smallness in expansions (2a) and (2b) supplements the above dependence (4) with a periodic term with period $d_0/2$, which should be averaged over the period. In the approximation considered here, the coefficient of the linear term depends only on k_1. Therefore, the value of k_1 can be determined experimentally from the rate of convolution of the growth curve if the variation of the layer thickness is known. The value of the thickness is unknown in most cases; for this reason, it is convenient to consider the dependence of $\ln|\rho - \rho_1|$ not on the thickness, but on the phase of factor X, which can easily be determined experimentally. In the simplified version, we can consider instead of the phase the number m of the spiral turn (interference period). Both these parameters (the phase and the turn number) are proportional to the thickness; therefore, the linear dependence is preserved. However, to determine k_1 in this case, we must know (at least approximately) the value of n_1 because it appears in the expression for X and d_0.

The method for determining the coefficient of absorption was tested experimentally for the synthesis of thin films of amorphous silicon Si(100) coated by a protecting SiO_2 layer [14]. The deposition of silicon films was carried out by thermal evaporation in ultra-high vacuum chamber [15] at room temperature. The measurements were taken with an ellipsometer [16] integrated into the vacuum deposition system. The probe radiation wavelength was $\lambda = 632.8$. Figure 2a shows the results of ellipsometry obtained during the growth of the Si layer. For convenience of subsequent calculations, the experimental data are represented not in the form of classical ellipsometric parameters Ψ and Δ, but as the real and imaginary parts of complex ellipsometric parameter ρ. In the figure, ρ_1, ρ_2, ρ_3, etc. are the extrema of function Re_$\rho(d)$ corresponding to different interference periods. To determine the absorption coefficient, we must construct the dependence of the logarithm of the radius vector on the interference period number $\ln|\rho - \rho_1| = f(m)$. In accordance with formula (3), this dependence is close to linear, but exhibits small oscillations (Fig. 2b) with period π, which are associated with the quadratic term in formula (2a). Another possible reason for the weak nonlinearity is systematic errors in single-zone measurements with period 2π. The linear approximation of the experimental curves over several interference periods makes it possible to eliminate both above factors. In accordance with relation (4), the slope of the resultant straight line is given by

$$\tan\alpha = \frac{4\pi k_1}{\lambda}md, \quad (5)$$

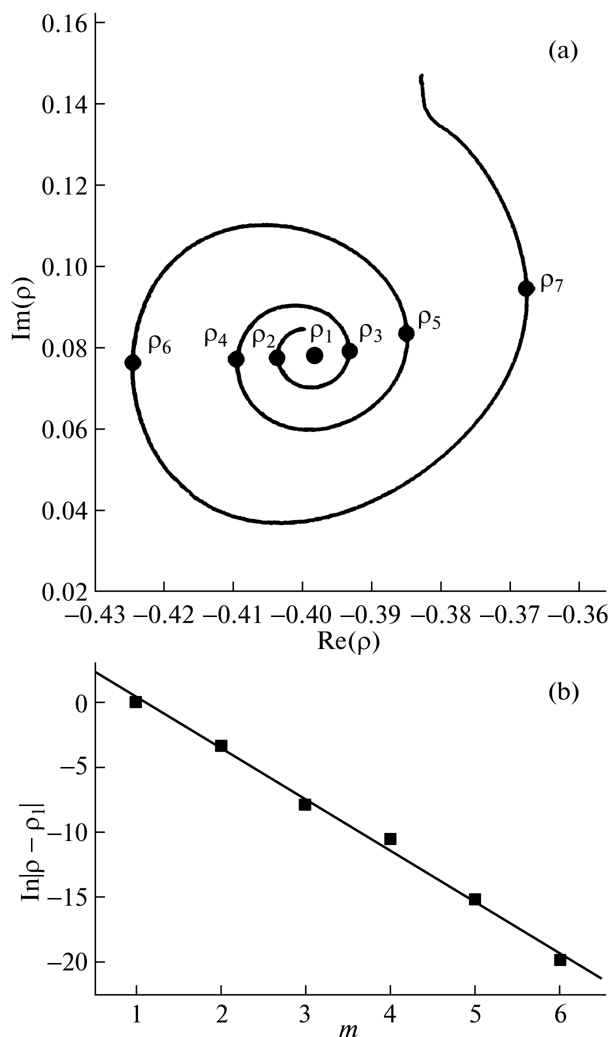

Fig. 2. Determination of the absorption coefficient for an amorphous silicon film.

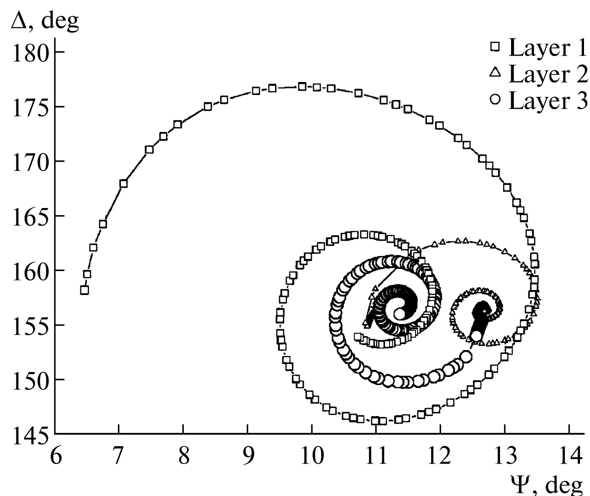

Fig. 3. Dependences of ellipsometric parameters measured according to single-zone technique during sequential growth of CMT layers with various contents ($x_1 = 0.535$, $x_2 = 0.395$, and $x_3 = 0.505$).

where m is the number of the interference period.

For the Si layer under investigation, the absorption coefficient k_1 is 0.48 ± 0.02; for $\lambda = 632.8$ nm, this corresponds to amorphous silicon [17].

The potentialities of the proposed technique were also demonstrated in determining the composition of films of $Cd_xHg_{1-x}Te$ (CMT) solid solutions grown by molecular-beam epitaxy. The absorption coefficient of CMT (and, hence, the rate of convolution of the growth curves) depends on content x. Figure 3 shows the ellipsometric curves measured during the growth of the films of various compositions.

Values of the content of CMT films determined by different methods

Layer number	Slope β	Content determined	
		from the rate of convolution of growth curve	from limiting point
1	0.1716	0.535	0.54
2	0.2015	0.395	0.375
3	0.1773	0.505	0.51

The films were grown successively one over another in a single technological process. Under these growth conditions, the technique under consideration can also be used because the rate of convolution of the growth curves depends only on the optical constants of the growing film (according to numerical simulation data) and is almost independent of the optical properties of the initial surface. The rate of convolution of complex ellipsometric parameter ρ to the limiting value ρ_1 was determined from these experimental curves. The corresponding dependences for amplitude $\ln|\rho - \rho_1|$ on phase $\phi = \arctan\left(\frac{\mathrm{Im}(\rho - \rho_1)}{\mathrm{Re}(\rho - \rho_1)}\right)$ are shown in Fig. 4.

After linear approximation of experimental curves over several interference periods, their slopes β were determined.

Comparing the experimentally determined slopes with the gauge curve (Fig. 5) obtained by calculating $\beta(x)$ taking into account the dependences of optical constants of CMT on the content and growth temperature measured earlier [17], we can determine the composition of the grown layers (see the table).

The table shows for comparison the values of content x obtained from the results of measurement of the limiting growth point ρ_1 to which the formulas for a semi-infinite medium are applicable [3]. Both methods give close results for content x. However, the limiting point method is more sensitive to the systematic error of measurements and to the presence of surface layers (in particular, to surface roughness).

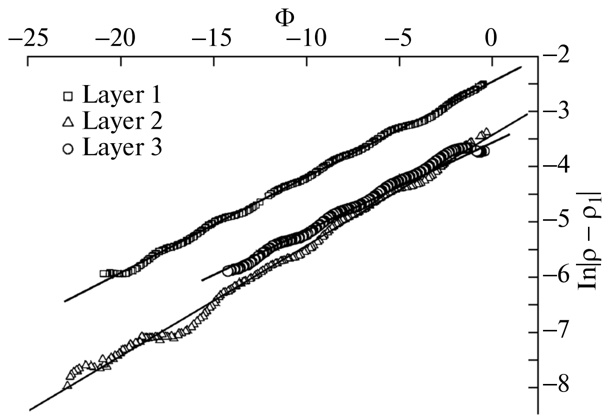

Fig. 4. Rate of convolution of the growth curves for CMT films with various contents ($x_1 = 0.535$, $x_2 = 0.395$, and $x_3 = 0.505$) and their approximation by the linear dependence.

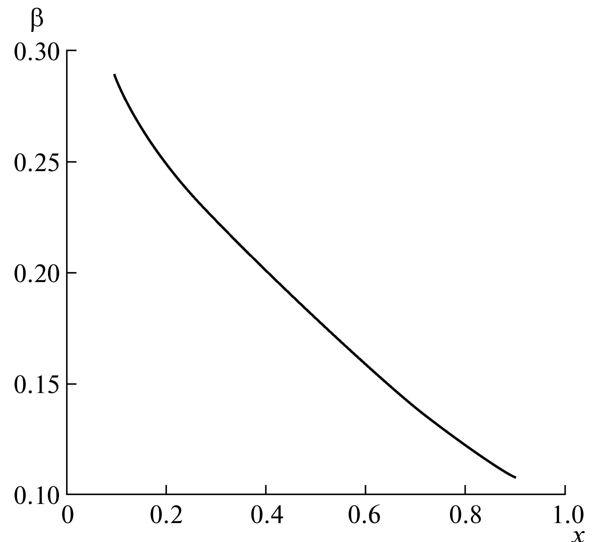

Fig. 5. Calculated dependence of slope β on the composition of CMT film.

Thus, we have developed a technique for estimating the absorption coefficient, which is based on relative variations of ellipsometric parameters Ψ and Δ (convolution rate of the growth curve). The criterion of applicability of this technique is a weak absorption of light ensured by absorption coefficient $k < 1$ (the value obtained for $\lambda = 632.8$ nm using a preliminary numerical experiment) and, hence, can be employed for determining the absorption coefficient for any material with $k < 1$.

It should be noted that slope β is determined only by the optical constants of the growing layer. It is almost independent of the presence of a transition or surface layer in the structure, which may considerably distort the shape of the growth curve, ruling out the determination of optical constants by fitting the data.

It is important that the technique developed here can be used for analyzing the growth of monolayer films as well as multilayer structures because the convolution rate of the growth curves depends only on the optical constants of the growing film and is almost independent of the optical properties of the initial surface.

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