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Very thin germanium films: optical and structural properties

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Very thin and ultrathin optically isotropic layers are key components for many applications in X-ray optics, microelectronics and optical storage of the information. Here we present an application of a recently developed spectrophotometric method for determination of optical constants (refractive index, n, extinction coefficient, k, and physical thickness, d) of very thin films to thermally deposited germanium (Ge) films with d between 10 and 25 nm. The method is based on limited development of the Abelès characteristic matrix elements. (n, k, d) are obtained by analytical solution of the system $(1+R_f)/T_f$, $(1-R_f)/T_f$ and $(1-R'_f)/T_f$, where (T_f) is the film transmittance, (R_f) is the front side and (R'_f) backside reflectance. For comparison to the so-obtained (n, k, d), Veritable Angle Spectroscopic Ellipsometry is used as an independent technique. The ellipsometric angles are fitted, using a generalized oscillation layer. An acceptable relative difference between (n, k, d), obtained by both methods, is achieved.

Keywords: Ge thin films, Optical constants

INTRODUCTION

The rapid expansion of contemporary nanotechnology stimulates the development of methods for the synthesis, preparation and characterization of very thin films. Optical methods for thin film characterization have the advantage that they are nondestructive, fast and effective. Several recently developed spectrophotometric [1, 2] and ellipsometric [3] methods for the estimation of the optical constants (refractive index n and extinction coefficient k) and the thickness d of nanolayers can be found in the literature. However, they have their own limitations.

Here, we report the application of a recently developed method for the determination of the complex refractive index $\tilde{n} = n - ik$ and the thickness to thermal deposited Ge films with d between 10 and 30 nm. In this method, the evaluation of n, k, and d is made by the use of spectrophotometric data of the thin film transmittance T_f, front side reflectance R_f and backside reflectance R'_f. Due to the nanothickness of the films we derived analytical expressions for R_f, R'_f and T_f by expansion of the Abelès characteristic matrix elements [4] to the 4-th order in terms of $\tilde{n}d/\lambda$. The exact analytical approach is used to estimate n, k and d. Thus, the problems related to multiple solutions or the lack of any solutions is overcome.

EXPERIMENTAL

The Ge films were deposited by thermal evaporation with a deposition rate of 0.3-0.4 nm/s at a base pressure of $< 10^{-4}$ Pa in the vacuum chamber of a LAB 500 evaporator (Leybold Optics GmbH). The intended thickness of the films was in the range from 10 nm to 25 nm. The deposition rate and the thickness of the film were controlled by a standard oscillating quartz sensor. 2 mm thick selected white float glass substrates (Präzisions Glas & Optic GmbH) were used. Prior to thin film deposition the substrates were cleaned in a H_2SO_4:H_2O_2 = 1:1 solution at 120°C for 10 min, subsequently rinsed in de-ionized water and spin-dried.

The optical transmission and reflection of the films were measured with a Cary 5E (Varian Co.) spectrophotometer at normal incidence in the range from 450 to 700 nm with an accuracy of 0.2% and 0.5%, respectively.

A spectroscopic ellipsometer M44 (J. A. Woollam Co. Inc.) was employed for the measurement of the ellipsometric angles (ψ and Δ)

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of the films. These ellipsometric parameters were derived for two angles of incidence (50° and 60°) within the spectral region from 450 - 700 nm.

Transmission Electron Microscopy (TEM) was carried out, using a Philips CM300 equipped with Super TWIN objective lenses.

RESULTS AND DISCUSSION

Determination of (n, k, d) of Ge films, by the proposed method

Two Ge films were studied. They were measured spectrophotometrically, and after proper correction for the finite thickness of the substrate (multiple reflections within it) and its optical performance (small spectral absorption) [5], the exact analytical approach was applied to estimate n, k and d [6]. First, by the help of the system $(1+R_f)/T_f$, $(1-R_f)/T_f$ and $(1-R'_f)/T_f$ we obtained $n(\lambda)$, $k(\lambda)$ and $d(\lambda)$. The physical thickness is not wavelength dependant and that is why we need an estimate of its average value. Then with so obtained n, k and d and the exact matrix elements we calculate T_f, R_f and R'_f. The differences $\Delta T = T_{cal} - T_f$ and $\Delta R = R_{cal} - R_f$, where T_{cal} and R_{cal} are the transmittance and reflectance of the film, calculated with the obtained n, k and d, using exact Abelès matrix elements [4]; T_f and R_f denote the experimental data, manipulated in order to take into account the finite thickness of the substrate. These corrections of measured values are decisive for the choice of the film thickness. In this case the estimation of the average thickness is done in the spectral range 450-500 nm, where ΔR changes its sign. The following thicknesses were calculated: d = 16 nm and 24 nm. With these values we recalculate n and k of the both films. The spectral dependences of n and k are plotted in Fig. 1. The calculated values of n and k for both films are sufficiently close to each other.

As a measure of the accuracy of the proposed method we use the differences ΔT and ΔR. Their spectral dependences are presented in Fig. 2. For the film with d = 16 nm the differences ΔT are ≅ 0.4% and ΔR ≅ 0.1% for whole spectral region under investigation, which are close to the maximum uncertainties of the Cary 5E spectrophotometer we use for the spectral measurements.

Additionally, variable angle spectroscopic ellipsometry (VASE) measurements have been carried out with Ge films. ψ and Δ are fitted using a generalized oscillation layer. It was supposed that both films have one and the same n and k, but different thicknesses (d_{VASE}). The film thicknesses (d^1_{VASE} и d^2_{VASE}) and the oscillator parameters: amplitude *Amp* [eV^2], central energy *En* [eV] and broadening energy *Br* [eV] were fitted. The following results are obtained: d^1_{VASE} = *16.7* nm, d^2_{VASE} = 23.5 nm; *Amp* =18.7, *En* = 3.7 Ev; *Br* = 5.4 eV with a *MSE* = 1.17. The dispersions of n_{VASE} and k_{VASE} are plotted in Fig. 1 and Fig. 2, respectively.

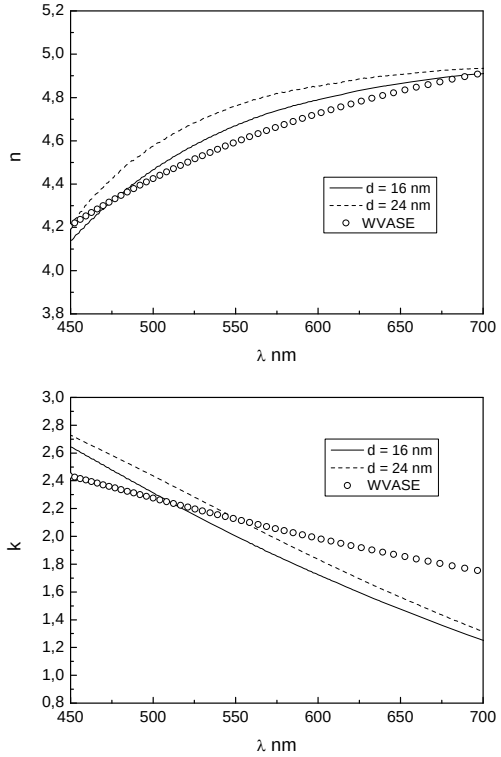


Fig. 1. Dispersion of n and k for the Ge films: ---n and k for d = 24 nm; — n and k for d = 16 nm; ○○○ n and k VASE results for the thicker film.

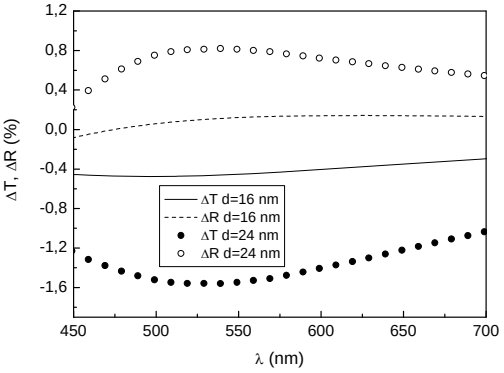


Fig. 2. Dispersion of ΔT and ΔR for Ge films: — ΔT for d = 16 nm, --- ΔR for d = 16 nm, ● ΔT for d = 24 nm and -○- ΔR for d = 24 nm.

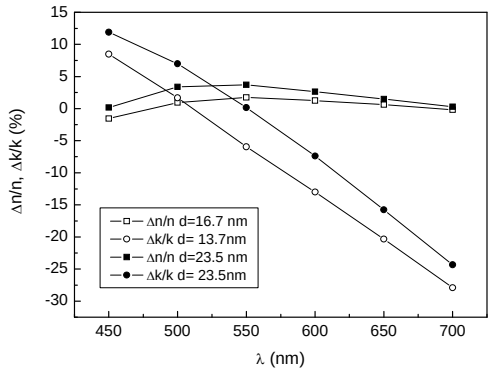
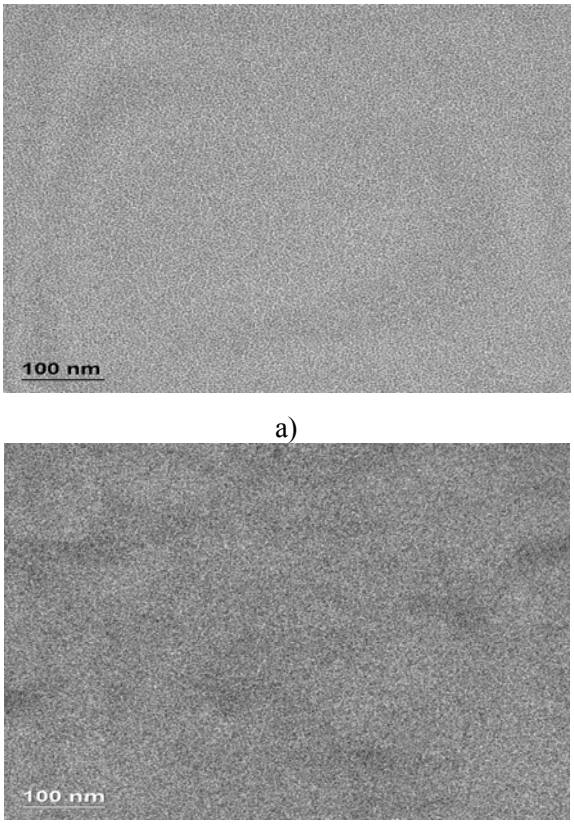


Fig. 3. Dispersion of Δn/n and Δk/k for the Ge films: -■- Δn/n (d = 23.5 nm), -□- Δn/n (d = 16.7 nm), -O- Δk/k (d = 16.7 nm) and -●- Δk/k (d = 23.5 nm).



a)

b)

Fig. 4. TEM micrograph of Ge film with d = 16 nm (a) and d = 24 nm (b).

We define the relative difference between n and k on the one hand and n_{VASE} and k_{VASE} on the other hand as: $\Delta n/n = (n_{vase} - n)/n_{vase}$ and $\Delta k/k = (k_{vase} - k)/k_{vase}$.

In Fig. 3 the spectral dependences of $\Delta n/n$ and $\Delta k/k$ for the both film are shown. Acceptable values for $\Delta n/n$ and $\Delta k/k$ are obtained. For instance, the maximum value of $\Delta n/n$ for the thinner is 1.7% and 3.7% for the thicker film at λ = 550 nm. The values of $\Delta k/k$ curve of the thinner film reach 25 % at 700 nm. We must have in mind that the generalized oscillator model, which is used in the VASE evaluations, is not at all proven to be close to the physical reality of evaporated very thin Ge films. Our approach is more realistic, flexible and free from pre-assumed models.

TEM characterisation

TEM was applied to investigate the morphology of the both thin films. The micrographs of the thinner and thicker film are shown in Fig.4a and Fig. 4b, respectively.

It can be seen that the films have fine granular structure and the grain dimensions of the both films are quite similar. Thus, TEM results are in a good agreement with the results from the optical characterization, which show independence of n and k from d in the thickness range 15 – 25 nm.

CONCLUSION

We have applied a simple and effective method for the determination of *n*, *k* and *d* of Ge thin films in VIS. The thin film optical parameters are evaluated in three steps. First, *n*, *k* and *d* are estimated for each wavelength of the spectral measurements. Then, from the obtained data, a single value of the physical thickness is evaluated within a specific spectral region. Finally, *n* and *k* are re-calculated in order to minimize the thin film optical response, this time with the estimated "scalar" value of the physical thickness. This approach is very functional and practical, because there is no need for *n* and *k* dispersion models (Cauchy, Drude, Selmeier, etc.). It does not use numeric minimization techniques, i.e. derivative methods, which must be used in the case of very thin films optics with extreme precaution. a

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МНОГО ТЪНКИ ГЕРМАНИЕВО СЛОЕВЕ: ОПТИЧЕСКИ И СТРУКТУРНИ СВОЙСТВА

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(Резюме)

Тънките и свръх тънките слоеве са основни градивни елементи на много приложения в рентгеновата оптика, микроелектрониката и оптичния запис на информация. В настоящата работа представяме приложението на развит от нас спектрофотометричен метод за определяне на оптичните константи (показател на пречупване, n, коефициент на поглъщане, k, и физична дебелина, d на много тънки слоеве към термично отложени германиеви слоеве с d между 10 и 25 nm. Методът се основава на ограничено развитие на елементите на характеристичната матрица на слоевете. (n, k, d) се пресмятат чрез аналитично решаване на системата уравнения $(1+R_f)/T_f$, $(1-R_f)/T_f$ и $(1-R'_f)/T_f$, където (T_f) е пропускането на слоя, (R_f) е отражението от страната на слоя и (R'_f) е отражението от страната на подложката. Като допълнителна методика, за сравнение на така пресметнатите (n, k, d), е използвана спектрална елипсометрия с променлив ъгъл като елипсометричните ъгли на измерените слоевете са фитнати чрез обобщен осцилаторен модел. Получена е приемлива разлика между (n, k, d), прсметнати по двата метода.