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Citation	Journal Of Applied Physics, 2004, v. 95 n. 12, p. 8481-8483
Issued Date	2004
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JOURNAL OF APPLIED PHYSICS VOLUME 95, NUMBER 12 15 JUNE 2004

Profile of optical constants of SiO_2 thin films containing Si nanocrystals

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(Received 19 June 2003; accepted 21 March 2004)

For optoelectronic and photonic applications of Si nanocrystals (nc-Si) embedded in a SiO_2 matrix, the information of the depth profiles of the optical constants for the thin film system is necessary. In this work, an approach of the depth profiling for the thin film synthesized with ion implantation is developed. In this approach, the nc-Si depth distribution obtained from secondary ion mass spectroscopy measurement is modeled with the approximation of many sublayers, and for a given wavelength the optical constants of each sublayer are formulated with the nc-Si volume fraction in the sublayer and the nc-Si optical constants as variables based on the effective medium approximation. After the above procedures the nc-Si optical constants are obtained from the spectral ellipsometric fittings. Finally the optical constants of each sublayer are calculated, and thus the depth profiles of the optical constants for the SiO_2 thin film containing the nc-Si are obtained. [DOI: 10.1063/1.1739282]

Si nanocrystals (nc-Si) embedded in a SiO_2 matrix have recently attracted much attention because of their light-emitting ability.[1–6] A promising method to form such Si-based light-emitting structures is the implantation of Si ions into the robust SiO_2 thin films that provide good chemical and electrical passivation of the Si nanocrystals, and the synthesis process is fully compatible with the mainstream complementary metal–oxide–semiconductor (CMOS) process. Recently, optical gain has been reported in nc-Si synthesized by implantation of Si into SiO_2 and annealing,[7] leading to a hope of the fabrication of a silicon laser, and waveguiding effects have also been observed in the measurement of optical gain in a layer of nc-Si prepared by Si ion implantation into synthetic silica substrates.[8] For optoelectronic and photonic applications of nc-Si embedded in a SiO_2 matrix, the information of the profiles of optical constants of the material system is necessary. In this work, an approach of optical-constants profiling of the thin film system synthesized with ion implantation is developed.

A 550-nm-thick SiO_2 film, which was grown on *p*-type Si (100) substrates by wet oxidation of Si at 1000 °C, was implanted with a dose of 1×10^{17} atoms/cm^2 of Si^+ at 100 keV. Following the ion implantation, a thermal annealing was carried out at 1000 °C in nitrogen gas. It is observed that the nc-Si volume fraction in SiO_2 is slightly different for different annealing durations. The annealing duration is 100 min in this study. X-ray diffraction (XRD) measurement indicates the formation of Si nanocrystals with a mean size of about 4 nm embedded in the SiO_2 matrix. The depth distribution of the nc-Si volume fraction in the SiO_2 film was calculated based on secondary ion mass spectroscopy (SIMS) measurement, and it is shown in Fig. 1(a). To obtain the profiles of optical constants of the material system, spectroscopic ellipsometry measurements were carried out in the wavelength range of 400–1200 nm.

As can be seen in Fig. 1(a), nc-Si distributes from the surface to a depth of 250 nm, and there are few or no nanocrystals in the SiO_2 film beyond the depth of 250 nm. Therefore the SiO_2 film can be divided into two portions, i.e., the first portion ($0\leqslant$depth$\leqslant$250 nm) that contains nc-Si, and the second portion (depth$>$250 nm) that is basically a pure SiO_2 layer. For the first portion, as the nc-Si volume fraction (v) varies with the depth (x), the optical properties in this portion will also vary with x. To model the optical properties of the first portion ($0\leqslant x\leqslant 250\,\mathrm{nm}$), $v(x)$ is modeled with an m-sublayers approximation, i.e., this portion is divided into m sublayers with equal thickness d_0, which are labeled as sublayer 1, sublayer 2,..., sublayer m in the sequence starting from the surface. The average nc-Si volume fraction in each sublayer is denoted with v_i ($i=1,2,...,m$). Each sublayer has its own complex refractive index, $N_i=n_i+\mathbf{j}k_i$ ($i=1,2,...,m$), where n_i and k_i are the refractive index and extinction coefficient for the ith sublayer, respectively. Note that N_i is also a function of wavelength (λ). Therefore, in the elliposmetry analysis, the material system used in this study can be described with a ($m+3$)-phases model, i.e., air/sublayer 1/$\cdots$/sublayer m/pure SiO_2 layer (i.e., the second portion mentioned above)/Si substrate. Each phase is characterized by its complex refractive index, N_i ($i=0,1,...,m,m+1,m+2$). Note that $N_0=1$ for air, $N_{m+1}=N_{SiO_2}$ (i.e., SiO_2 refractive index) for the second portion, and $N_{m+2}=N_{Si}$ (i.e., Si complex refractive index) for the Si substrate. As N_0, N_{m+1}, and N_{m+2} are known, for a fixed incident angle the ellipsometric angles Ψ and Δ can be expressed as functions

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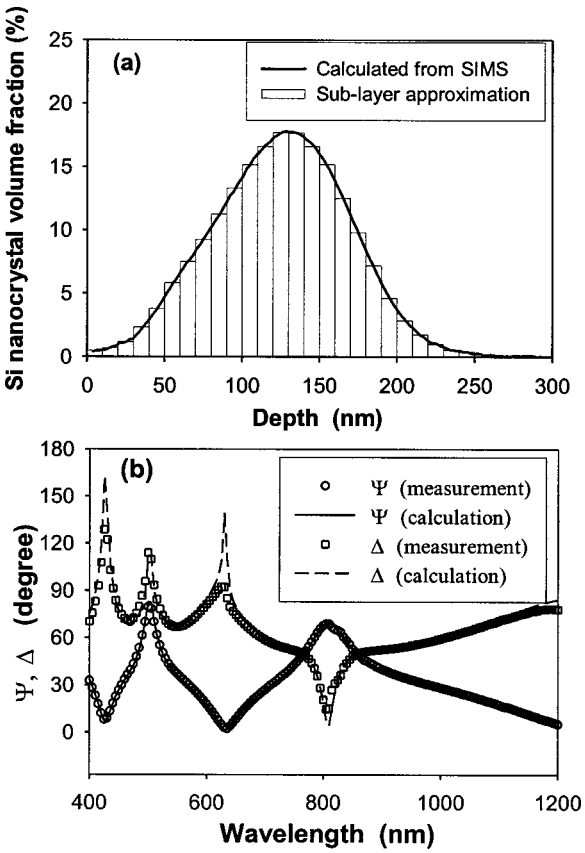

FIG. 1. (a) nc-Si depth distribution determined from SIMS measurement and the sublayer approximation with $m=25$; and (b) spectral ellipsometric fittings with $m=25$.

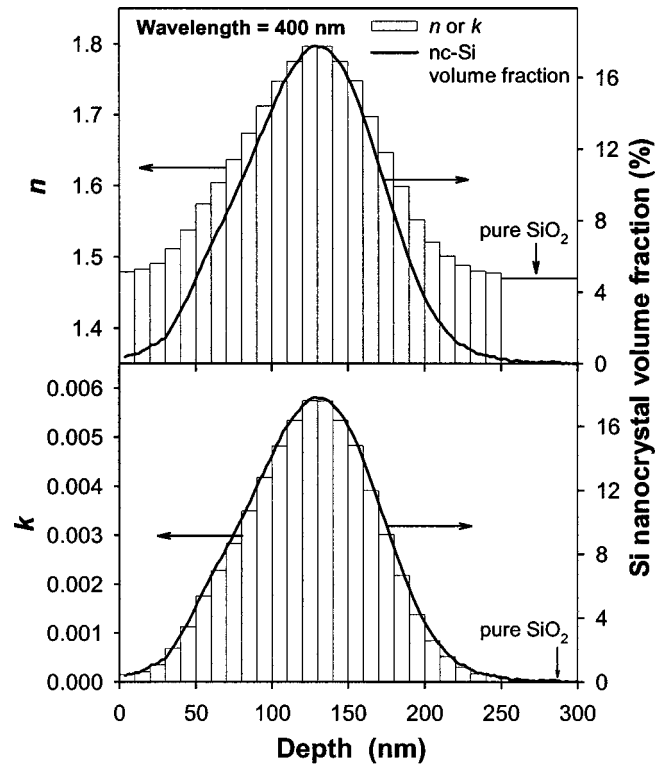

FIG. 2. Depth profiles of refractive index n and extinction coefficient k at the wavelength of 400 nm for the SiO_2 thin film containing nc-Si.

of the parameters n_1, $n_2,\ldots,n_m$, k_1, $k_2,\ldots,k_m$, and the wavelength λ,[9,10] i.e.,

$$\Psi=f_1(n_1,n_2,\ldots,n_m,k_1,k_2,\ldots,k_m,\lambda) \tag{1}$$

and

$$\Delta=f_2(n_1,n_2,\ldots,n_m,k_1,k_2,\ldots,k_m,\lambda) \tag{2}$$

where f_1 and f_2 are two complicated functions, which cannot be expressed as analytical formulas.

Each sublayer can be treated as a material system in which the SiO_2 is a host material while the nc-Si is an inclusion embedded in the host material. From the effective medium approximation,[9,11,12] the refractive index n_i and the extinction coefficient k_i for the ith sublayer ($i=1,2,\ldots,m$) can be obtained as follows:

$$n_i=\left[\frac{\sqrt{(A^2+B^2)(C^2+D^2)}+(AC+BD)}{2(C^2+D^2)}\right]^{1/2} n_{SiO_2}, \tag{3}$$

$$k_i=\left[\frac{\sqrt{(A^2+B^2)(C^2+D^2)}-(AC+BD)}{2(C^2+D^2)}\right]^{1/2} n_{SiO_2}, \tag{4}$$

where

$$A=(1+2v_i)(n_{\text{nc-Si}}^2-k_{\text{nc-Si}}^2)+2(1-v_i)n_{SiO_2}^2,$$

$$B=2(1+2v_i)n_{\text{nc-Si}}k_{\text{nc-Si}},$$

$$C=(1-v_i)(n_{\text{nc-Si}}^2-k_{\text{nc-Si}}^2)+(2+v_i)n_{SiO_2}^2,$$

$$D=2(1-v_i)n_{\text{nc-Si}}k_{\text{nc-Si}}.$$

In the above equations, $n_{\text{nc-Si}}$ and $k_{\text{nc-Si}}$ are the mean refractive index and extinction coefficient of the nc-Si (note that the influence of the nc-Si size distribution is considered in terms of the mean values), respectively, and n_{SiO_2} is the refractive index of SiO_2. Note that all the parameters are dependent of wavelength. For a given wavelength λ, as the volume fraction (v_i) and n_{SiO_2} are known, if the values of both $n_{\text{nc-Si}}$ and $k_{\text{nc-Si}}$ are given the calculation with Eqs. (1)–(4) can yield the values (Ψ_{cal} and Δ_{cal}) of the ellipsometric angles; and on the other hand, an ellipsometric measurement yields the experimental values (Ψ_{exp} and Δ_{exp}) of the ellipsometric angles. Therefore the optical constants ($n_{\text{nc-Si}}$ and $k_{\text{nc-Si}}$) of the Si nanocrystals at the wavelength can be found by comparing the calculated values (Ψ_{cal} and Δ_{cal}) with the experimental values (Ψ_{exp} and Δ_{exp}). In this study, we use a spectral fitting in the wavelength range from 400 to 1200 nm to determine the nc-Si optical constants at each wavelength. In the spectral fitting, the nc-Si optical constants at various wavelengths λ_1, $\lambda_2,\ldots,\lambda_M$ (M is the number of wavelengths) can be found by searching for one set of $n_{\text{nc-Si}}(\lambda_1)$, $n_{\text{nc-Si}}(\lambda_2),\ldots,n_{\text{nc-Si}}(\lambda_M)$, $k_{\text{nc-Si}}(\lambda_1)$, $k_{\text{nc-Si}}(\lambda_2),\ldots,k_{\text{nc-Si}}(\lambda_M)$ such that the error function below

$$F=\sum_{\lambda=\lambda_1}^{\lambda_M}[(\Psi_{exp}^{\lambda}-\Psi_{cal}^{\lambda})^2+(\Delta_{exp}^{\lambda}-\Delta_{cal}^{\lambda})^2] \tag{5}$$

is a minimum for the whole measured spectral range, where Ψ_{exp}^{λ} and Δ_{exp}^{λ} are the measured values while Ψ_{cal}^{λ} and Δ_{cal}^{λ} are the calculated values at the wavelength λ.

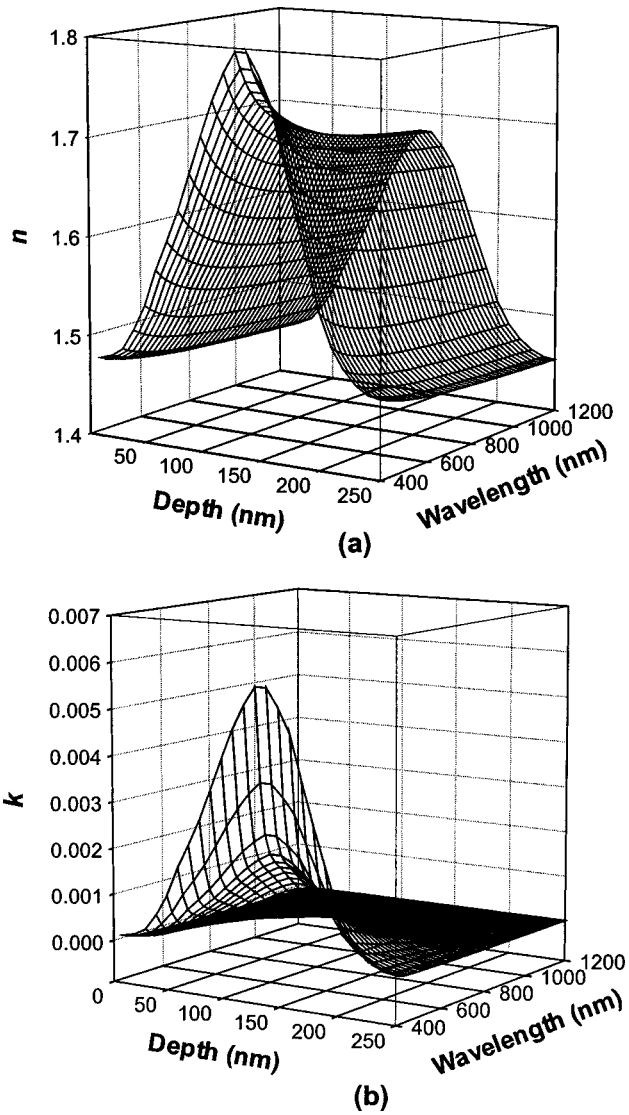

FIG. 3. Refractive index n and extinction coefficient k as functions of both the depth and wavelength for the SiO_2 thin film containing nc-Si.

By using the nc-Si optical constants obtained from the spectral fitting with $m=25$, the refractive index (n_i) and the extinction coefficient (k_i) of each sublayer ($i=1,2,...,m$) can be calculated with Eqs. (3) and (4), and thus the depth profiles of the optical constants (n and k) for the SiO_2 thin film containing the nc-Si are obtained. Figure 2 shows the depth profiles of the optical constants at the wavelength of 400 nm, and Fig. 3 shows the three-dimensional view of the refractive index and extinction coefficient as functions of both the depth and wavelength. As shown in Fig. 2 for the wavelength of 400 nm, there are large changes in the optical constants in the depth range of ~50–200 nm with the maximum changes (the maximum change in the refractive index is about 0.32) occurring in the depth of about 130 nm. In the region deeper than about 250 nm, the optical constants are basically the same as those of pure SiO_2. As shown in Fig. 3, the changes of the optical constants due to the Si nanocrystals depend on the wavelength with larger changes at shorter wavelengths, and the wavelength dependence of the change of the extinction coefficient is much stronger than that of the refractive index. For long wavelengths (near-infrared to infrared), the extinction coefficient is very small (and thus there is no strong light absorption) in the depth range of ~50–200 nm although the refractive index in this region is significantly larger than that of pure SiO_2. This provides the possibility of waveguiding in the SiO_2 thin film structures with appropriate depth profiles of Si nanocrystals.

In summary, we have developed a quantitative approach of optical-constants profiling of SiO_2 film containing nc-Si synthesized with the ion beam technique. In this approach, the nc-Si depth distribution obtained from SIMS measurement is modeled with the approximation of many sublayers, and for a given wavelength the optical constants of each sublayer are formulated with the nc-Si volume fraction in the sublayer and the nc-Si optical constants as variables based on the effective medium approximation. After the above procedures the nc-Si optical constants are obtained from the spectral ellipsometric fittings. Finally the optical constants of each sublayer are calculated, and thus the depth profiles of the optical constants for the SiO_2 thin film containing nc-Si are obtained.

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