Study on optical anisotropy properties of SiO_2 films with different thermal annealing temperatures

Huasong Liu (刘华松)[1*], Yugang Jiang (姜玉刚)[1,2], Lishuan Wang (王利栓)[1], Chenghui Jiang (姜承慧)[1], Yiqin Ji (季一勤)[1,3], and Deying Chen (陈德应)[3]

[1] *Tianjin Key Laboratory of Optical Thin Film, Tianjin Jinhang Institute of Technical Physics, Tianjin 300192, China*

[2] *Key Laboratory of Advanced Micro-structure Materials, Ministry of Education, Department of Physics, Tongji University, Shanghai 200092, China*

[3] *National Key Laboratory of Science and Technology on Tunable laser, Institute of Optical-electronics, Harbin Institute of Technology, Harbin 150080, China*

[*] *Corresponding author: liuhuasong@gmail.com*

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SiO_2 films are deposited on Si substrates by an ion beam sputtering technique and continuously annealed in a quartz culture dish in air at various annealing temperature ranging from 20 to 750 °C with a step of 100 °C for a fixed time of 24 h. The effects of thermal treatment on optical anisotropy properties of SiO_2 films are investigated by spectroscopic ellipsometry. When the annealing temperature is 550 °C, the optical anisotropy properties of SiO_2 film is minimum. The obtained results indicate that the optical anisotropy properties of SiO_2 films can be improved by a proper thermal annealing process.

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SiO_2 is a very important low refractive index material and can be used in combination with high refractive index material such as Ta_2O_5 to manufacture the coatings with ultra-low optical loss. Because of its stability, amorphous nature (no grain boundaries), high density, adjustable refractive index, and low particulate contamination, SiO_2 has many typical applications, such as high-reflection coatings, antireflection coatings, all-dielectric mirrors, beam-dividers, band-pass filters, and polarizers[1−4]. Several different deposition methods have been tried to get high quality SiO_2 films including ion beam assisted deposition (IBAD)[5], molecular beam epitaxy (MBE)[4], radio frequency magnetron sputtering[6], plasma enhanced chemical vapor deposition (PECVD)[7], and reactive pulsed laser deposition (RPLD)[8]. The refractive index of SiO_2 films can be matched easily by adjusting the deposition conditions and the partial pressure of oxygen.

In order to design high reflective and antireflective coatings with low optical loss more accurately, it is necessary to precisely measure and calculate optical constants of high and low refractive index materials. The determination of the optical constants of thin films is of great importance for the design of any optical and optoelectronic device. The methods used for the determination of optical constants are usually classified as single-wavelength methods[9−11] and multi-wavelength methods[12,13]. In the multi-wavelength methods, the optical constants are strongly related to one another and to the wavelength, and this dependency can be represented by certain optical dispersion equations. As a result, the latter methods are relatively insensitive to random measurement errors and lead to better results.

Thermal annealing is an effective method to improve optical properties of SiO_2 films. After annealing, the transmittance and rigidities of SiO_2 films had been great improved, and the obtained best annealing temperature is 600 °C[6]. The dielectric properties of SiO_2 films can be improved and a blue shift of the infrared absorption spectral peak in a Fourier transform infrared attenuated total reflection spectrum was observed with thermal annealing[14].

In this letter, SiO_2 films are deposited on Si (110) substrates by an ion beam sputtering technology. Spectroscopic ellipsometry is mainly used to measure optical constants of the SiO_2 films. Cauchy and Uniaxial layer model are used as the fitting model to study the effect of thermal annealing on refractive index and optical anisotropy properties. The effects of different thermal annealing temperature on optical anisotropy properties of SiO_2 films are investigated.

The Si (110) is chosen as the materials of substrates and the substrates are 100 mm in diameter and 0.3 mm in thickness. SiO_2 target with the purity of more than 99.995% and the diameter of 14 inch is used as thin films forming material. Due to ion beam sputtering being one of the best deposition methods for achieving the low absorption and low scattering coating, so we choose ion beam sputtering as the deposition methods in our experiment. The designed physical thicknesses of SiO_2 films were all about 860 nm. When SiO_2 films have been prepared, we will both measure ellipsometric spectra data before and after thermal treatment for SiO_2 films.

Air-annealing experiments were carried out in a high temperature cabinet (PHH101S, ESPEC, Japan) with temperature control precision of ±1 °C. In order to study the effects of annealing temperature on optical properties of SiO_2 films, sample 1#, 2#, 3#, 4#, 5#, 6#, 7#, and 8# were continuously annealed in a quartz culture dish in air at various annealing temperature ranging from 20 to 750 °C with a step of 100 °C for a fixed time of 24 h.

The spectroscopic ellipsometry is in general more accurate in determining optical constant because of the sensitivity of the phase difference for small variations in the optical thickness. In order to study the effects of annealing time and temperature on optical properties of $\mathrm{SiO_2}$ films with amorphous structure, we have recorded the ellipsometric parameters ψ and Δ of these samples at different annealing times and temperatures.

WVASE32 software was used to fit the measured Psi (ψ) and Delta (Δ) data. In the fitted process, the optical model consisted of the Si substrate, the effective medium approximation (EMA) with 50% Si and 50% film bulk, the Cauchy layer, simple graded index material and a top surface roughness layer with 50% voids and 50% film bulk was used to fit the thickness and refractive index of $\mathrm{SiO_2}$ films with the measured $\varPsi$ and Δ experimental data. The best numbers of sublayers is proved to be 51, so 51 sublayers were all chosen in the calculation of optical constants of $\mathrm{SiO_2}$ films in our experiments.

Over part of the spectra for many non-absorbing materials, the refractive index n can be represented by a slowly varying function of wavelength. The refractive index dispersion can be modeled by Cauchy formula[9,15,16]

$$n = A_n + \frac{B_n}{\lambda^2} + \frac{C_n}{\lambda^4}, \qquad (1)$$

where n is the refractive index, λ is the wavelength in μm, A_n describes the long-wavelength asymptotic index value, and B_n and C_n are the dispersion terms that add upward slope to the index curve as wavelengths become shorter (B_n and $C_n \geqslant 0$). Equation (1) can describe dispersion for a material that is essentially non-absorbing over the measured wavelength range. Equation (1) can provide a means to curve-fit dispersion. However, the Cauchy term is most effective when curve fitting normal dispersion (n decreasing with wavelength). The Cauchy dispersion relation was design to model the normal dispersion seen in transparent materials.

In order to obtain more accurate optical anisotropy properties of $\mathrm{SiO_2}$ films with different annealing temperatures, we further chose Uniaxial layer model as the fitting model after finishing the fit of thickness and refractive index. In the fitting process, a model consisting of a Si substrate, the Cauchy layer, dNz layer, Nz(cauchy)/100%(dNz) layer, and Uniaxial layer are employed. The Cauchy layer, dNz layer, and Nz(cauchy)/100%(dNz) layer don't need to fit the thickness and they are all zero thickness. The entire film thickness is represented by Uniaxial layer and the dummy layers are used only to define a set of optical constants.

Spectroscopic ellipsometry measurements were made with a two-channel spectroscopic polarization modulation ellipsometer in the spectral range from 0.6 to 5.2 eV with an interval of 0.04 eV at an angle of incidence 65° by Woollam variable angle spectroscopic ellipsometry. Due to obtaining more accurate optical constants in the visible range, so the wavelength range was chosen from 400 to 800 nm in the fitting process. The experimental and model fit of $\varPsi$ and Δ data before and after annealing are shown in Fig. 1. Compared with the measured ellipsometric spectra of $\mathrm{SiO_2}$ films before annealing, the corresponding spectra extend to blue shift after thermal annealing. It can be seen that the fit data agree very well with the experimental data.

Using the calculated methods as above, the measured ellipsometric data with different annealing temperatures are all fitted to study the effect of thermal annealing on optical anisotropy properties of $\mathrm{SiO_2}$ films. Various calculated refractive indexes from z direction to xy plane with different annealing temperatures are shown in Fig. 2. As the annealing temperature of $\mathrm{SiO_2}$ films is increased, the various quantity of refractive index from z direction to xy plane of $\mathrm{SiO_2}$ films become diminution, when the annealing temperature reaches 550 °C, the various quantity of refractive index from z direction to xy plane reaches the least, and then the various quantity of refractive index become large as the annealing temperature continues to increase. The least corresponding variation quantity is all below 0.0005 in the wavelength range from 400 to 800 nm. The obtained results illustrate that optical anisotropy properties of $\mathrm{SiO_2}$ films can be improved.

The obtained typical refractive index curves for xy plane and z direction of $\mathrm{SiO_2}$ films after coating and after thermal annealing with 550 °C temperature are shown in Fig. 3. It can be seen that the various quantity of refractive indexes of xy plane are different from the values of z direction after coating. Before annealing, the difference of refractive indexes between xy plane and z direction are large and the variety is about 0.002. After thermal annealing with 550 °C temperature, the calculated refractive indexes between xy plane and z direction are almost the same.

The refractive indexes of $\mathrm{SiO_2}$ films with different annealing temperatures were calculated from the measured $\varPsi$ and Δ data. The various quantities of refractive indexes with different annealing temperatures at 633 nm are shown in Fig. 4. As the annealing

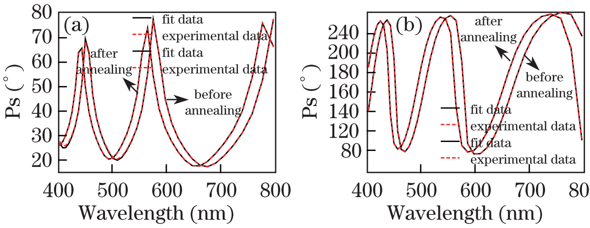

Fig. 1. (Color online) Experimental and model fit of (a) $\varPsi$ and (b) Δ data before and after annealing.

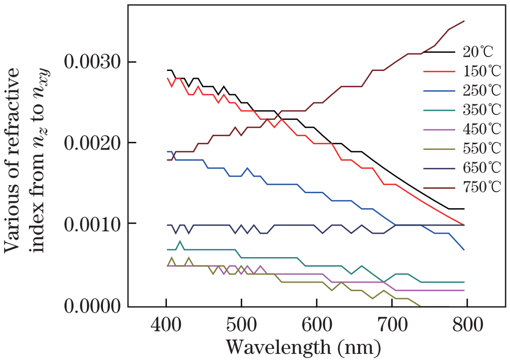

Fig. 2. (Color online) Various refractive indexes from z direction to xy plane with different annealing temperatures.

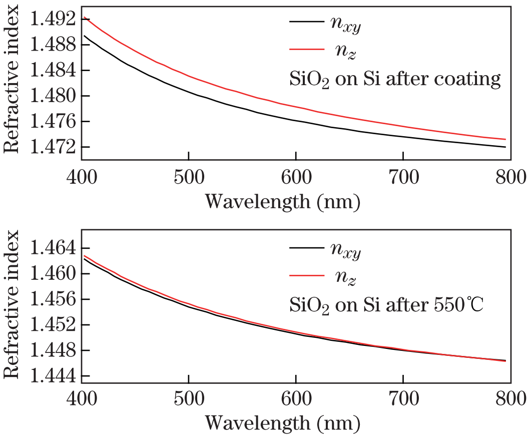

Fig. 3. (Color online) Refractive index curves for xy plane and z direction of SiO$_2$ films after coating and after thermal annealing.

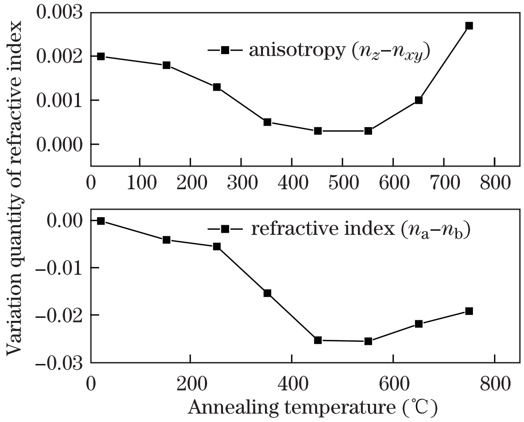

Fig. 4. Various quantity of anisotropy and refractive indexes from z direction to xy plane after annealing, from after annealing to before annealing at 633 nm.

temperature of SiO$_2$ films is increased, the refractive index of SiO$_2$ films become diminution, when the annealing temperature is 550 °C, the various quantity of refractive index reaches the largest, and then the various quantity of refractive index become diminution as the annealing temperature continues to increase. The various quantities of refractive indexes from z direction to xy plane after annealing at 633 nm are also shown in Fig. 4. It can be seen that the tendency of various quantity of refractive index are the same between optical anisotropy properties and refractive index.

In conclusion, SiO$_2$ films on Si (110) substrate are prepared by an ion beam sputtering technology, and thermal annealing effects on optical anisotropy properties of SiO$_2$ thin films are investigated. Spectroscopic ellipsometry is used to measure Ψ and Δ data. Cauchy and Uniaxial layer model are used as the fitting model to study the effect of thermal annealing on refractive index and optical anisotropy properties. As the annealing temperature of SiO$_2$ films is increased, the various quantity of refractive index from z direction to xy plane of SiO$_2$ films become diminution, when the annealing temperature reaches 550 °C, the various quantity of refractive index from z direction to xy plane reaches the least, and then the various quantity of refractive index becomes large as the annealing temperature continues to increase. The tendency of various quantity of refractive index is the same between optical anisotropy properties and refractive index. The obtained results indicate that optical anisotropy properties of SiO$_2$ films can be improved with appropriate thermal annealing.

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