Very low-refractive-index optical thin films consisting of an array of SiO_2 nanorods

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The refractive-index contrast in dielectric multilayer structures, optical resonators, and photonic crystals is an important figure of merit that creates a strong demand for high-quality thin films with a low refractive index. A SiO_2 nanorod layer with low refractive index of $n=1.08$, to our knowledge the lowest ever reported in thin-film materials, is grown by oblique-angle electron-beam deposition of SiO_2. A single-pair distributed Bragg reflector employing a SiO_2 nanorod layer is demonstrated to have enhanced reflectivity, showing the great potential of low-refractive-index films for applications in photonic structures and devices. © 2006 Optical Society of America

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In distributed Bragg reflectors,[1] the refractive-index contrast, which is the difference in refractive index between the two constituent materials, is directly related to the reflectivity, the spectral width of the stop band, and the penetration depth. In optical microresonators,[2] the effective cavity length, and hence the enhancement of spontaneous emission, is directly dependent on the index contrast. In photonic crystals,[3] the photonic bandgap width is directly related to the index contrast. In semiconductor optoelectronics, a dielectric material with a very low refractive index is the key component of the optical parts in the devices.[4,5] The need for such a component motivates the development of new airlike optical materials with a refractive index close to 1.0.

Although multilayer structures with air gaps have been demonstrated,[1] air gaps completely lack structural stability, making them unsuitable for the majority of applications. MgF_2, CaF_2, and SiO_2 are materials with refractive indices among the lowest available for conventional, dense optical coatings. However, their refractive indices are much higher than the index of air, 1.0. Nanoporous SiO_2 thin-film materials made from solgel processes[6,7] have low refractive indices,[4,5] good mechanical strength, and low scattering coefficients. However, it is difficult to precisely control film thickness and the uniformity of spin-coating processes used for solgel materials.

Oblique-angle deposition[8] is a technology by which to grow porous, sculptured thin films[9] as a result of the self-shadowing effect during the deposition process.[10,11] Figure 1(a) shows the deposition principle of oblique-angle deposition. A random growth fluctuation in the substrate produces a shadow region that the subsequent incident vapor flux cannot reach. Also produced is a high edge where the incident flux is deposited preferentially, thereby creating an array of oriented rods.

In this Letter, a low-refractive-index (low-n) optical thin film consisting of randomly distributed SiO_2 nanorods grown by oblique-angle electron-beam (e-beam) deposition is presented. It is shown that the refractive index of the SiO_2 nanorod layer is $n=1.08$, to our knowledge the lowest ever reported in thin-

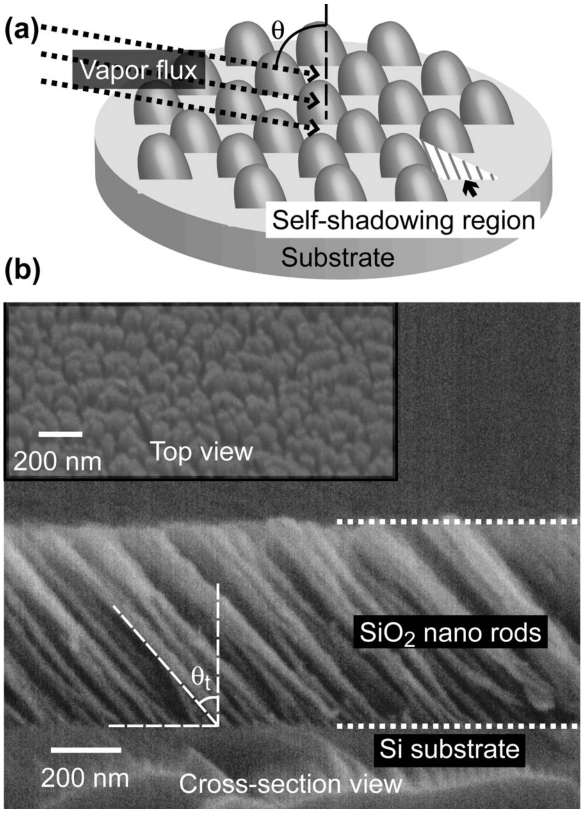

Fig. 1. (a) Schematic of oblique-angle deposition. (b) Top and cross-sectional views of a SEM for a SiO_2 nanorod layer.

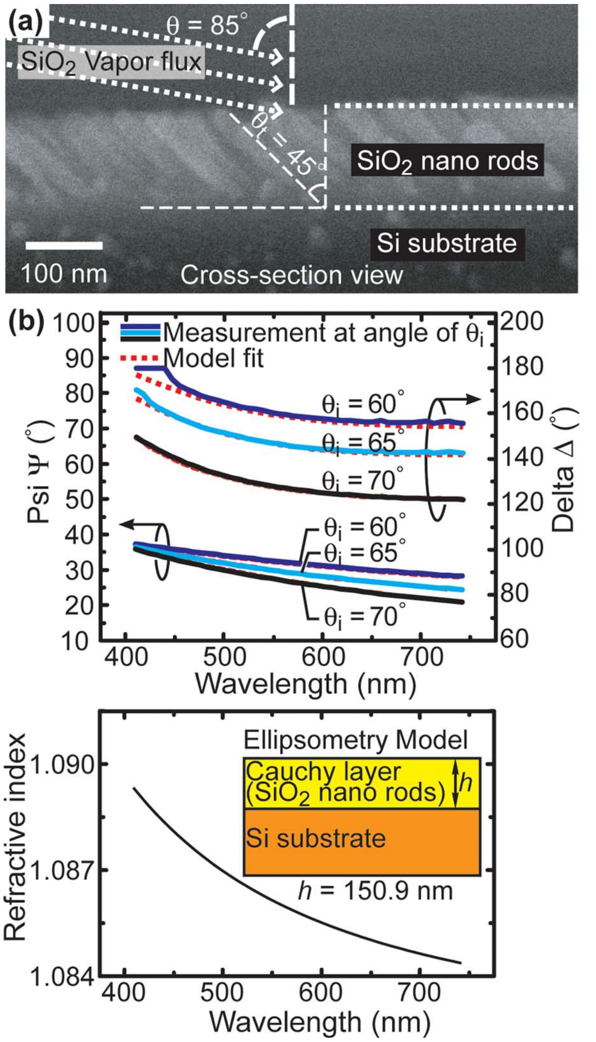

Fig. 2. (Color online) (a) Cross-sectional view of a SEM of a SiO_2 nanorod film. (b) Ellipsometry raw data, Ψ and Δ, and the model fit of the Cauchy layer on the Si substrate versus wavelength. The refractive index and thickness of the Cauchy layer are fitted. The mean square error of the fit is 5.3.

film materials. In addition, a single-pair dielectric reflector employing a SiO_2 nanorod layer is demonstrated to have much higher reflectivity than one employing dense SiO_2 because of the higher refractive-index contrast of the former.

Figure 1(b) shows a scanning electron micrograph (SEM) of the SiO_2 nanorod layer grown by oblique-angle e-beam deposition on a Si substrate. The oblique incident angle of the vapor flux, θ, is 85°. The evaporation source material is pure SiO_2 granules. During deposition, the chamber pressure is 2×10^{-6} Torr, and the deposition rate is 0.5 nm/s. The SiO_2 nanorods are uniformly distributed with a tilt angle of $\theta_t=45°$. The gap between SiO_2 nanorods is less than 30 nm, much smaller than the wavelength of visible light and hence sufficiently small to minimize optical scattering. Furthermore, since the film is deposited by evaporation, the controllability of the film thickness is excellent and suitable for deposition of quarter-wavelength optical films.

A thin SiO_2 nanorod film ($h=150$ nm) is grown on a Si substrate for ellipsometry measurement. Figure 2(a) shows a cross-sectional SEM of the SiO_2 nanorod layer. The sample is measured at multiple beam incident angles, 60°, 65°, and 70°. The raw ellipsometry data, Ψ and Δ, and the model fit versus wavelength are shown in Fig. 2(b). The ellipsometry model assumes a Cauchy-type layer on a Si substrate. The refractive index versus wavelength is also shown in Fig. 2(b). Within the visible spectrum, the refractive index of the SiO_2 nanorod layer is extremely low, $n=1.08$. The thickness of the SiO_2 nanorod layer, determined from the ellipsometry measurement, is $h=150.9$ nm, confirming the thickness obtained by scanning electron micrography [Fig. 2(a)]. Oblique-angle deposited films have been reported to be anisotropic.[12] However, no pronounced anisotropy is found in our thin SiO_2 nanorod film, possibly due to its high porosity and small thickness. Assuming an approximately linear dependence of refractive index on porosity, the very low refractive index of 1.08 indicates a porosity of 80.5%.

A single-pair dielectric reflector with a Si/SiO_2 nanorod layer is fabricated to demonstrate the viability of the SiO_2 nanorod films for use in multilayer optical components. Figure 3(a) shows the fabrication

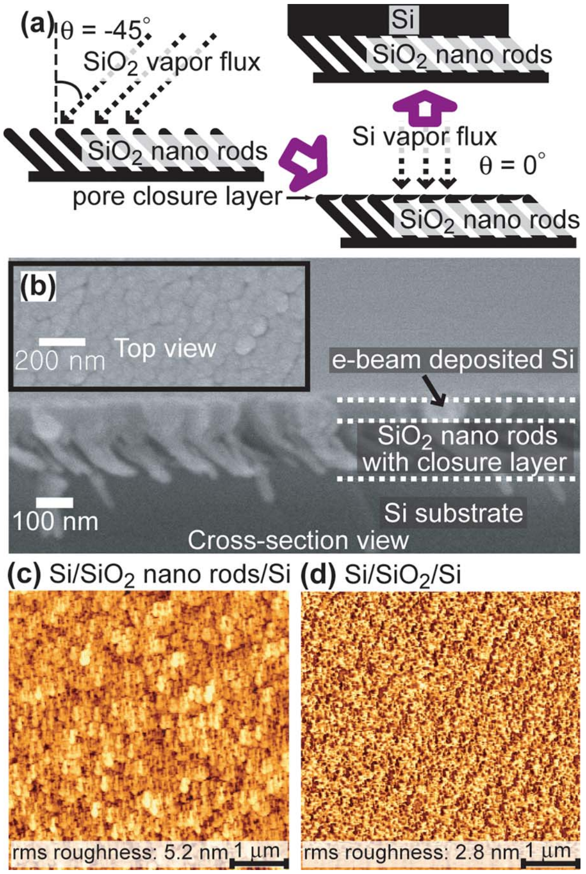

Fig. 3. (Color online) (a) Fabrication steps for closing the openings at the top surface of the SiO_2 nanorod layer. (b) SEM of a single-pair dielectric reflector incorporating a SiO_2 nanorod layer. (c) AFM of a single-pair dielectric reflector incorporating a SiO_2 nanorod layer $(5\ \mu\text{m}\times 5\ \mu\text{m})$. (d) AFM of a single-pair dielectric reflector, Si/dense SiO_2 $(5\ \mu\text{m}\times 5\ \mu\text{m})$.

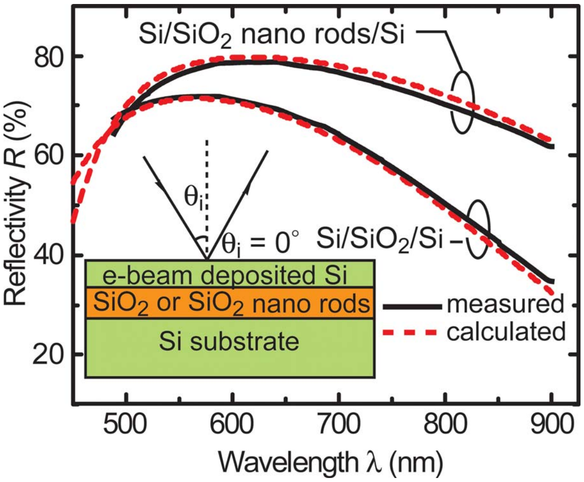

Fig. 4. (Color online) Reflectivity of a single-pair dielectric reflector employing a SiO_2 nanorod layer and a reflector employing dense SiO_2 at normal incidence. The solid curves are measured reflectivity, and the dashed curves are calculated reflectivity.

steps of the reflector. To avoid the filling of subsequent deposits into the openings between SiO_2 nanorods, a very thin (20 nm) pore-closure layer is formed on the top surface of the SiO_2 nanorod layer. The deposition condition of the pore-closure layer is the same as that of the SiO_2 nanorod layer, except that the vapor incidence angle is $\theta = -45°$, which changes the nanorods' growth direction.[13] After that, a 41 nm thick Si layer is deposited at normal incidence by e-beam evaporation. Figure 3(b) is a SEM of the Si/SiO_2 nanorod reflector. The cross-sectional view clearly shows that the e-beam-deposited Si is located on the pore-closure layer on the SiO_2 nanorod layer, forming a sharp interface between them. An atomic-force micrograph (AFM) of the surface of the reflector, shown in Fig. 3(c), has a rms surface roughness of 5.2 nm.

For comparison, a single-pair dielectric reflector with Si/dense SiO_2 is deposited on a Si substrate by e-beam evaporation using normal-incidence evaporation. The thickness of the dense SiO_2 is 107 nm to guarantee that the optical path length of the dense SiO_2 is the same as that of the SiO_2 nanorod layer. The thickness of the Si is 41 nm. An AFM of the Si/dense SiO_2 reflector, shown in Fig. 3(d), has a rms surface roughness of 2.8 nm.

The reflectivity at normal incidence of the reflectors is measured for the visible and near-infrared wavelengths. Both the measured and the calculated reflection spectra are plotted in Fig. 4. In the calculation, refractive indices of 1.08 and 1.46 are used for the SiO_2 nanorod layer and the dense SiO_2 layer, respectively. The e-beam-deposited Si has a refractive index of $2.94 + 0.110i$ at a wavelength of 633 nm. Inspection of Fig. 4 reveals that the normal-incidence reflectivity is clearly enhanced for the single-pair dielectric reflector with the SiO_2 nanorod layer compared with the one using dense SiO_2. The measured peak reflectivity of the Si/SiO_2 nanorod reflector is $R = 78.9\%$. The measured peak reflectivity of the Si/dense SiO_2 reflector is $R = 72.0\%$. The measurement data match the calculation data well. The increase in reflectivity also shows the huge potential advantage of low-n thin films for optical coatings.

In conclusion, optical thin films consisting of randomly distributed SiO_2 nanorods have been shown to have a very low refractive index of $n = 1.08$. The low-n thin films are grown by oblique-angle e-beam deposition of SiO_2. A single-pair dielectric reflector consisting of this low-n SiO_2 nanorod layer is demonstrated to have higher reflectivity than the reflector with dense SiO_2, which is attributed to the higher refractive-index contrast of the former. The low-n thin film is expected to have a wide range of applications and to be well suited for improved multilayer reflectors, optical resonators, and photonic crystals.

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