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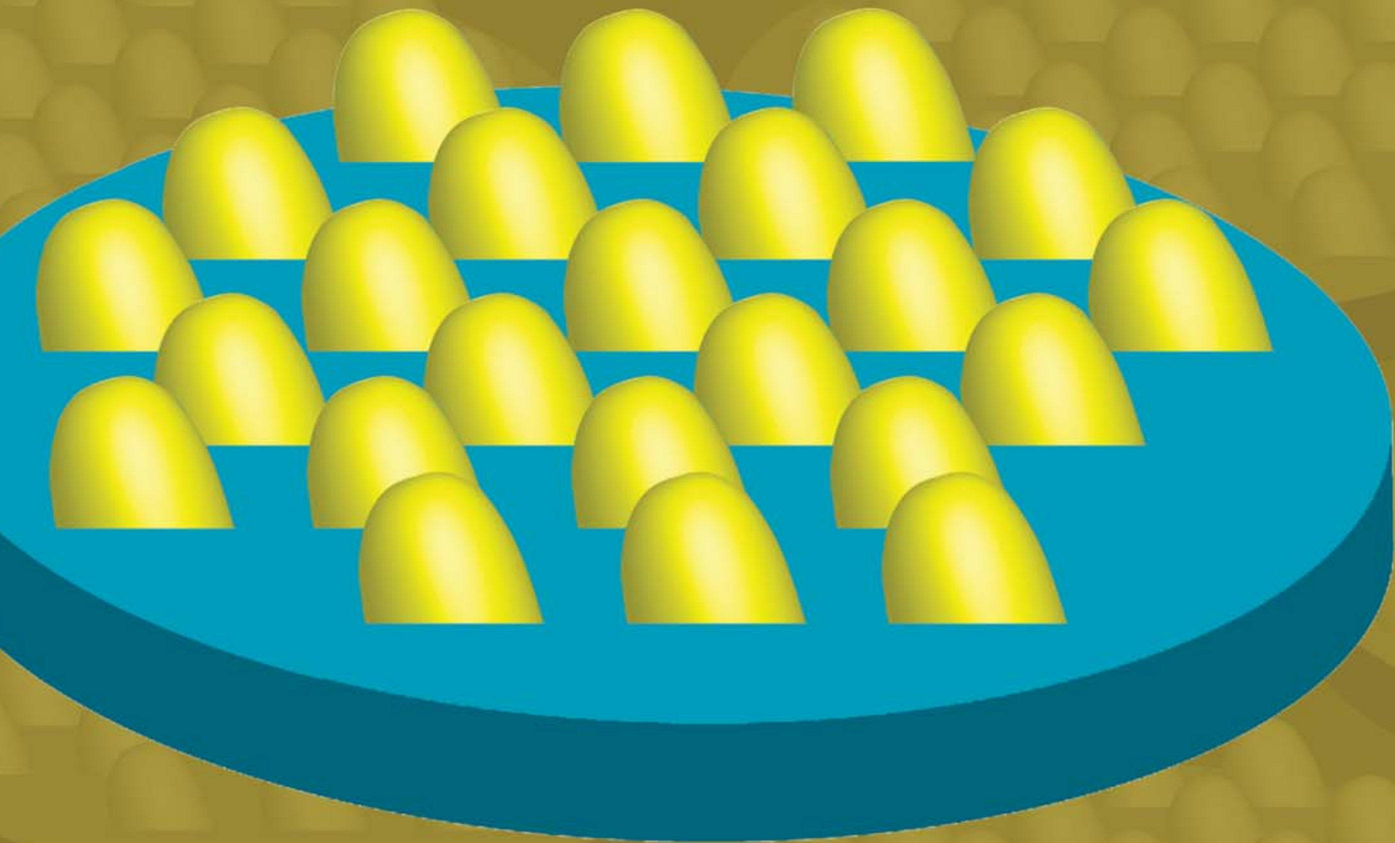
Low-Refractive-Index Films: A New Class of Optical Materials



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Low-Refractive-Index Films: A New Class of Optical Materials

J.-Q. Xi, Jong Kyu Kim, and E. F. Schubert

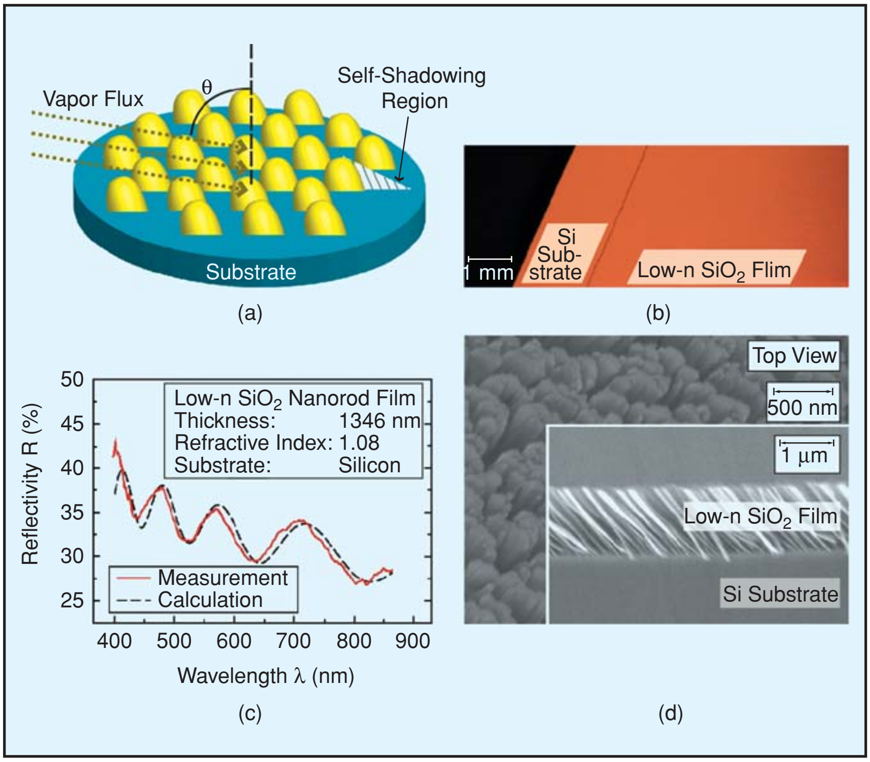

Fig. 1. (a), Schematic of low-n thin-film growth using oblique-angle deposition; (b), optical micrograph of a low-n SiO_2 nano-rod layer on Si substrate; (c), thin-film reflectivity versus wavelength of SiO_2 nano-rod film. Theoretical thin-film interference simulations reveal a refractive index of 1.08; (d), scanning electron micrograph of a low-n SiO_2 nano-rod layer.

Abstract

The refractive-index contrast is a key figure of merit for dielectric multilayer structures, optical resonators, and photonic crystals. This creates a strong demand for a new class of optical materials that have refractive indices much lower than those of conventional materials. Dielectric films consisting of an array of SiO_2 nano-rods grown by oblique-angle electron-beam deposition have unprecedented low refractive indices < 1.10. A single-pair distributed Bragg reflector employing a low-refractive-index (low-*n*) SiO_2 nano-rod layer has enhanced reflectivity, demonstrating the great potential of low-*n* films for applications in photonic devices and multilayer structures.

Introduction

In dielectric multilayer structures, optical resonators, and photonic crystals, multiple figures of merit strongly depend on the refractive-index contrast, i.e. the difference in refractive index between the high-index and low-index material. For distributed Bragg reflectors (DBRs), the reflectivity, spectral width of the high-reflectivity stop band, optical penetration depth, and the maximum angle for high reflectivity directly depend on the refractive index contrast. In optical micro-resonators, the effective cavity length, and thus the spontaneous emission enhancement, directly depends on the index contrast. In photonic crystals, the width of the photonic bandgap directly depends on the index contrast. However, a high refractive-index contrast is limited by the availability of materials with low refractive index. Here, we report on low-*n* thin films, a new class of optical materials with unprecedented low refractive index [1-4].

Gases are the materials with lowest refractive index (n_{air} @ 1.0). Although DBRs with air-gaps have been demonstrated, their fabrication requires under-etching and hence is slow and costly. Moreover, air gaps completely lack structural stability, making them unsuitable for the majority of applications. MgF_2, CaF_2, and SiO_2 with refractive indices n_{MgF2} = 1.39, n_{CaF2} = 1.44, n_{SiO2} = 1.46, respectively, are materials with refractive indices among the lowest available for conventional, dense optical coatings. Nanoporous SiO_2 thin film materials from sol-gel process have refractive indices lower than that of dense SiO_2, proper mechanical strength, and low light scattering. Recently, nanoporous SiO_2 with refractive index of 1.23 have been demonstrated for quarter-wave layers [2]. An even lower index (n = 1.10) was demonstrated for thicker layers [3]. However, it is difficult to control the thickness and the uniformity of films deposited by a spin-on sol-gel process. This motivates the development of processes capable of uniform deposition of films with a thickness of one quarter wavelength.

DEPARTMENT OF ELECTRICAL, COMPUTER, AND SYSTEMS ENGINEERING
DEPARTMENT OF PHYSICS, APPLIED PHYSICS, AND ASTRONOMY
RENSSELAER POLYTECHNIC INSTITUTE,
110 EIGHTH STREET, TROY, NY 12180

Low-*n* SiO_2 nano-rod layer

Very recently, optical films consisting of an array of SiO_2 nano-rods with a refractive index as low as n = 1.08 were demonstrated and shown to have viable optical properties thereby making them highly desirable for many applications [1]. The optical films are fabricated by oblique-angle deposition of SiO_2 using electron-beam evaporation. Oblique-angle deposition is a method to grow thin films with a porous microstructure, caused by the self-shadowing nature of the deposition process. Fig. 1(a) shows the principle of oblique-angle deposition. A random growth fluctuation on the substrate produces a shadow region that the incident vapor flux cannot reach, and a non-shadow region where incident flux deposits preferentially, thereby creating an array of oriented SiO_2 nano-rods.

The optical micrograph of a SiO_2 nano-rods film deposited on a Si substrate is shown in Fig. 1(b). The SiO_2 nano-rod film was grown by oblique-angle electron-beam deposition on a Si substrate with a vapor flux incident angle θ of 80°. The optical micrograph reveals a smooth specular surface with no indication of scattering. A scanning electron micrograph (SEM) of this film is shown in Fig. 1(d). The cross-sectional SEM clearly shows the tilted array of SiO_2 nano-rods. The gap between the SiO_2 nano-rods is $\leq$ 40 nm, i.e. much smaller than the wavelength of visible light, thereby strongly reduces Rayleigh and Mie Scattering. The growth direction of the nano-rods is about 40° with respect to the surface normal of the sample.

The experimental optical reflectivity of the low-n SiO2 nano-rod layer on Si substrate is shown in Fig. 1(c) as a function of wavelength. The reflectivity reveals periodic thin-film interference oscillations. Simulations of the reflectivity reveal that the oscillations are fully consistent with a refractive index of 1.08 and a thin-film thickness of 1.35 mm. These values were confirmed by both, ellipsometry measurements and thin-film thickness measurements using SEM shown in Fig. 1(d). Furthermore, the films were found to be fully transparent with an optical absorption below the detection limit. The refractive index of 1.08 is the lowest one ever reported for an optical coating. Additionally, due to the fact that such low-n material is deposited by evaporation, the thickness of the thin film can be smaller than quarter wavelength.

However, in multilayer structure fabrication with low-*n* SiO_2 nano-rods, a critical step in the deposition is a surface-sealing step in which a very thin SiO_2 nano-rod array layer is grown on top of the low-*n* film. The nano-rods orientation of the sealing film is near perpendicular to the orientation of the main film, thereby reducing the ability of a material deposited on top of the low-*n* film to enter the low-*n* film. This was found to occur in multilayer films without such sealant film. Fig. 2 shows fabrication steps for sealant layer on the top surface of the SiO_2 nano-rod layer. The deposition condition of the sealant layer is the same as that for SiO_2 nano-rod layer except that the vapor incidence angle is θ = - 45°.

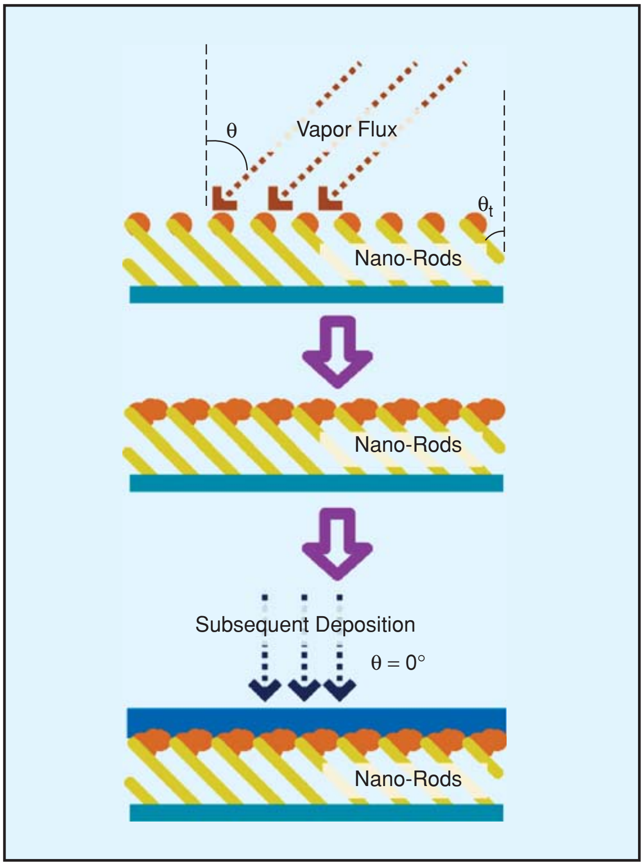

Fig. 2. Fabrication steps for sealant layer at the top surface of the low-n SiO_2 nano-rod layer.

Single-pair DBR with low-*n* SiO_2 nano-rod layer

A single-pair DBR using Si/SiO_2 nano-rod layer, fabricated on Si substrate, demonstrates the viability of the SiO_2 nano-rod films for use in multilayer optical components. At first, a low-*n* SiO_2 film was grown by oblique-angle electron-beam deposition on Si substrates with a vapor flux incident angle θ of 80°. Pure SiO_2 granules are used as evaporation source, and the deposition rate is well controlled at 5 Å/sec. A SiO_2 sealant layer

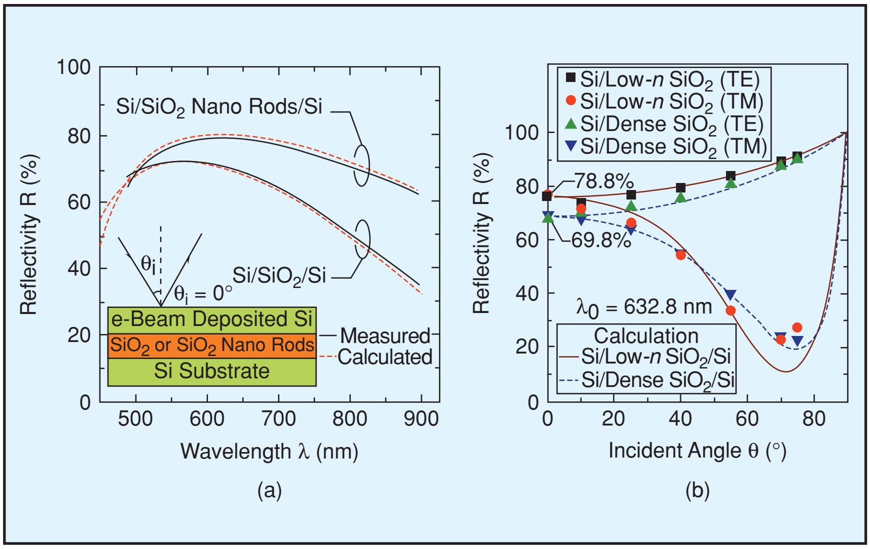

Fig. 3. (a), Reflectivity for both of the Si/SiO_2 nano-rod layer and Si/dense SiO_2 on Si substrate at normal incidence; (b), angular dependent reflectivity for both of the Si/SiO_2 nano-rod layer and Si/dense SiO_2 on Si substrate at wavelength λ_0 = 632.8 nm.

is formed on the top surface of the SiO_2 nano-rod layer. After the sealant layer deposition, a 41 nm-thick Si layer is deposited at normal incidence by electron-beam evaporation. For comparison, a single-pair DBR with Si/dense SiO_2 is deposited on a Si substrate by electron-beam evaporation using normal incidence evaporation. The thickness of dense SiO_2 is 107 nm to guarantee that the optical path length of the dense SiO_2 is the same as the one in the low-*n* SiO_2 layer.

The reflectivity at normal incidence of the DBRs is measured for visible and near infrared wavelengths. Both of the measured and calculated reflection spectra are plotted in Fig. 3(a). In the calculation, a refractive index of 1.08 and 1.46 is used for the low-*n* SiO_2 nano-rod layer and the dense SiO_2 layer, respectively. E-beam deposited Si has a complex refractive index 2.94 + 0.110 *i* at wavelength of 633 nm. Inspection of Fig. 3(a) reveals that the normal-incidence reflectivity is clearly enhanced for the DBR with SiO_2 nano-rod layer compared with the DBR using the dense SiO_2. The measured peak reflectivity of the Si/low-*n* SiO_2 nano-rod DBR is R = 78.9 %. The measured peak reflectivity of the Si/dense SiO_2 DBR is R = 72.0 %. The measured reflectivities match the calculated reflectivities very well. In addition to the normal-incidence results, the experimental and calculated angle-dependent reflectivities are also shown in Fig. 3(b) at wavelength λ_0 = 632.8 nm. The experimental results are in excellent agreement with the theoretical results, and the increase in reflectivity shows the huge potential of low-*n* thin films for optical coatings.

Conclusions

Dielectric films consisting of an array of SiO_2 nano-rods are shown to have extremely low values of the refractive index, significantly lower than that of conventional thin-film SiO_2. Films with a refractive index of 1.08 display excellent optical reflectance and transmittance properties. A single-pair Si/low-*n* SiO_2 distributed Bragg reflector is shown to have enhanced reflectivity. Low-*n* films may enable a new generation of dielectric multilayer structures, optical resonators, and photonic crystals with enhanced refractive-index contrast and superior optical properties.

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