Anti-reflection coating at 550 nm fabricated by atomic layer deposition

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Trimethylamine (TMA), $TiCl_4$, and water are applied as the precursors to deposit Al_2O_3 and TiO_2. With different substrate temperatures, the optical properties and surface morphologies of the two oxides TiO_2 and SiO_2 are studied, respectively. With substrate temperature of 120 °C, amorphous TiO_2 can be obtained, and the surface roughness (RMS) is only 0.928 nm. Applying Al_2O_3 and TiO_2 deposited in 120 °C as low and high refractive index materials, anti-reflection (AR) coating at single point (550 nm) is designed. Furthermore, with the calibrated growth rates, this AR coating is fabricated, and its ultimate reflectance for the AR coating at 550 nm is less than 0.2%, which can meet the requirement for most applications.

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Atomic layer deposition (ALD), also called as atomic layer epitaxy, is based on the surface controlled reaction. During coating process, two or more kinds of chemical vaporous or gaseous precursors enter into the chamber. These precursors react sequentially on the reactive sites of substrate surface, producing a solid thin film, and the by-product is purged by the inert gas simultaneously[1,2]. Because the film growth in ALD has very high packing density, excellent adhesion, and can realize unconditionally conformal deposition, ALD has been widely adopted to fabricate complementary metal oxide semiconductor (CMOS), dynamic random access memory (DRAM), and other semiconductor devices in recent decades[3−6]. At present, the application scope of ALD is expanded into optics areas. For example, E. I. du Pont de Nemours and Company introduces ALD to fabricate the barrier layer of organic light-emitting diodes (OLEDs), and improve its service life significantly. Corning Inc. applies the ALD deposited Al_2O_3 as the packaging layer in micromirror device (DMD).

Al_2O_3 and TiO_2 are the common high and low refractive materials, and are extensively used in optical multilayer coating fabrication. In ALD, the developments of the two materials are also relatively mature, and their growths can readily be controlled. Compared with electron-beam evaporation, Al_2O_3 and TiO_2 deposited by ALD are more promising, because they have very high packing density, accurate stoichiometry, and less temperature sensitivity[7−9]. In this letter, single layer TiO_2 and Al_2O_3 are first deposited on BK7 at different substrate temperatures. Further analysis shows that the optical properties of Al_2O_3 maintain approximately the same between 120 and 200 °C. However, optical loss of TiO_2 at 200 °C increases drastically, compared with the one at 120 °C, which is mainly induced by the crystallization in TiO_2 above 140 °C[10]. To obtain better optical performance, 120 °C is selected as the deposited temperature. The practical performance of this anti-reflection (AR) coating matches well with the designed model and compares favorably with that by other deposition technologies.

The experimental equipment is TFS 200 reactor made by Beneq, and its deposition vacuum is approximately 6 mbar. In TiO_2 deposition, the pulsing time is 200 ms for both $TiCl_4$ and water. Meanwhile, for Al_2O_3, the pulsing time is 600 ms for trimethylamine (TMA) and water, respectively. 2 s is set as the purging time for all reactants. During the whole fabricated process, the temperature of the liquid precursors is kept at 20 °C to guarantee the repeatability of experiments. All the precursors are liquid, thus the surface vapor pressure of them are high enough, so that extra carried gas is not necessary. To acquire the uniform distribution of the reactants in short time, the flow rate of N_2 is maintained as 250 sccm during the whole deposited process.

At the substrate temperatures of 120 and 200 °C, the single layer TiO_2 and Al_2O_3 are deposited, and their transmittances are illustrated in Fig. 1. In previous experiment, it has been verified that Al_2O_3 is amorphous below 800 °C[11,12]. The average transmittance is above 91.4% from 400 to 1 200 nm while the peak value is approximately 93%, which approaches to that of bare BK7 substrate. It represents that the Al_2O_3 has good homogeneity irrespective of the substrate temperature. Moreover, between 120 and 200 °C, the increasing deposited temperature almost has no effect on the homogeneity of Al_2O_3, except advancing the refractive index a little.

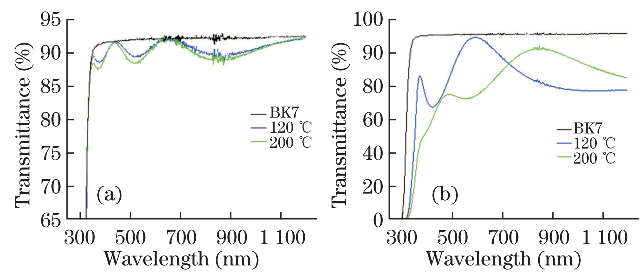

Fig. 1. (Color online) (a) Transmittance of Al_2O_3 deposited at 120 and 200 °C; (b) the transmittance of TiO_2 deposited at 120 and 200 °C.

However, for the single layer TiO_2 deposited by ALD, the transmittance is closely relevant to the deposited temperature. With the substrate temperature of 120 °C, the optical loss in TiO_2 is negligible, but the transmittance shrinks sharply when the substrate temperature reaches 200 °C. Such results can directly ascribed to the fact that TiO_2 is amorphous at 100–140 °C, while over 165 °C, the crystallization in TiO_2 results in worse surface roughness and larger scatter[13].

To deeply understand the effect of surface roughness on optical properties, we measure their surface morphologies by atomic force microscopy (AFM), and the results are shown in Figs. 2 and 3. At 120 and 200 °C, the Al_2O_3 surface is relatively smooth, including some tiny particles. The particle fluctuations are small, while the maximum size of these particles is less than 4 nm. Diverse substrate temperatures perform little influence on the surface morphology of Al_2O_3. The RMS is 0.803 nm for Al_2O_3 with 120 °C deposited temperature , and 1.078 nm for the one with 200 °C deposited temperature. The value is almost the same as the Al_2O_3 fabricated by electron beam evaporation, which can satisfy the requirement in optics[14]. On the other hand, for the TiO_2 growth, the surface morphology is closely related to the substrate temperature. In Fig. 3(a), the surface of TiO_2 grown at 120 °C is comparatively flat, and the RMS is only 0.928 nm, but some peaks still exist which may originate from the residual precursors, as comparatively low deposited temperature makes the purging process more difficult. However, the surface morphology of the TiO_2 worsens when the deposited temperature increases to 200 °C. It is well known that at 165 °C and or above temperature, TiO_2 are polycrystalline, and lattice scattering and surface scattering is drastically strengthened, resulting in the decline of transmittance (Fig. 1(b))[10]. Therefore, compared with the surface of TiO_2 at 120 °C, the one at 200 °C is rougher, and the particles on it are larger. Its RMS even reaches 8.529 nm, which can hardly meet the requirement in optics. To obtain eligible optical performance, 120 °C is chosen as the deposited temperature during the AR coating fabrication.

The optical refractive indexes of TiO_2 and Al_2O_3 are illustrated in Fig. 4, which are slightly higher compared with the one deposited by electron beam, owing to the excellent packing density of ALD. With such indexes, AR coating at 550 nm is designed, whose coating type is H=50.41 nm, L=38.78 nm, H=22.53 nm. Here H represents TiO_2 and L represents Al_2O_3, respectively. Without considering the extinction

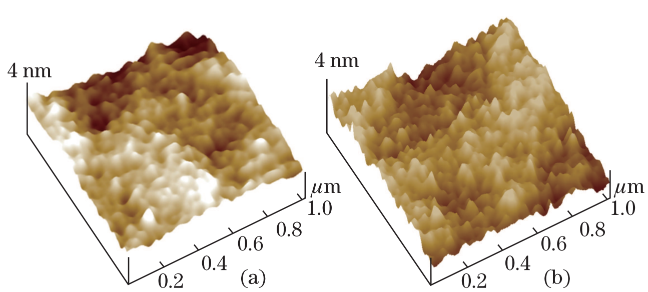

Fig. 2. Morphological images of Al_2O_3 grown at (a) 120 and (b) 200 °C. The surface roughness (RMS) is 0.803 nm for Al_2O_3 with 120 °C deposited temperature, and 1.078 nm for the one with 200 °C deposited temperature.

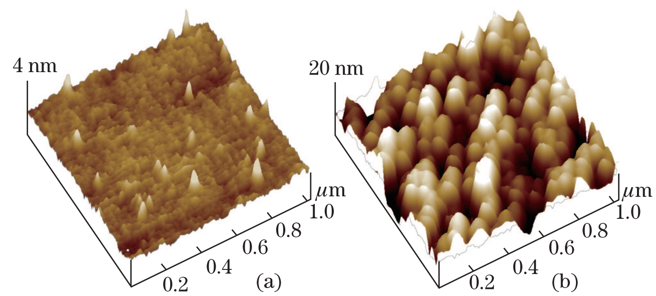

Fig. 3. Morphological images of TiO_2 grown at (a) 120 and (b) 200 °C. The RMS is 0.928 nm for TiO_2 with 120 °C deposited temperature, and 8.529 nm for the one with 200 °C deposited temperature.

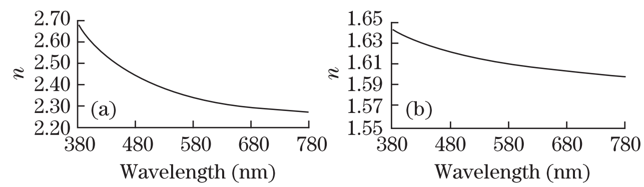

Fig. 4. (a) Optical refractive index of TiO_2 at 120 °C; (b) optical refractive index of Al_2O_3 at 120 °C.

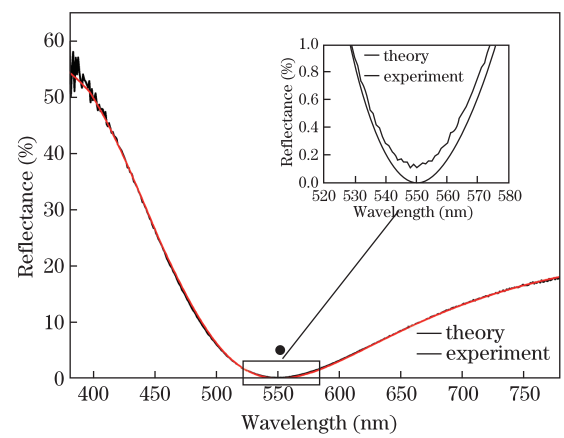

Fig. 5. (Color online) Reflectance of AR coating at 550 nm; at the top right corner, the reflectance is demonstrated in detail, and the reflectance is no more than 0.2% at 550 nm.

coefficient, the designed reflectance is 0.02% at 550 nm (Fig. 5). From the single layer experiments above, we confirm that the growth rate in ALD is almost a constant when the grown film is thicker. It is approximately 0.59 nm/cycle for TiO_2, and 0.13 nm/cycle for Al_2O_3 at 120 °C. Because the coating growths in ALD have excellent stability and repeatability, the thickness control can simply be realized by counting the numbers of cycle. The experimental result is also shown in Fig. 5, and no apparent divergence can be observed between the theoretical reflectance and the practical one. The magnified comparison of detailed data is sketched at the top right corner of Fig. 5. Only a little deviation exists, which hints the experimental reflectance is no more than 0.2% at 550 nm, and approximates the optimal prediction. The good result represents that, in the aspects of both growth rates and refractive indexes, the calibrations are more precise.

In conclusion, for ALD, which is seeing rapid growth in optics domain, its applications on multilayer fabrication also attract increasing attentions. Due to the mechanism of ALD, most corresponding deposition facilities lack of on-line monitoring, while time monitoring is regarded as an alternative to be adopted to realize the thickness control. Hence, the growth rate calibration is critical for precise film fabrication. Furthermore, to acquire a multilayer coating with a good performance, it is also necessary to build up accurate refractive index models. In this letter, the technologies of TiO_2 and Al_2O_3 deposited by ALD are given, and the growth rate and refractive index are measured precisely. With these data, AR coating is designed and fabricated. The practical reflectance of the AR coating at 550 nm is 0.2%, which can even reach the precision level of mature deposition technology. Although fabricating multilayer coating with ALD is just at its initial stage, according to the experimental results above, it may bring other properties, which may potentially expend the application scope of other complicated multilayer coating.

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