Design and preparation of frequency doubling antireflection coating with different thicknesses of interlayer for LiB_3O_5 crystal

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Received August 10, 2006

Design and preparation of frequency doubling antireflection coating with different thicknesses of interlayer were investigated for LiB_3O_5 (LBO) substrate. The design was based on the vector method. The thickness of the inserted SiO_2 interlayer could be changed in a wide range for the four-layer design with two zeros at 1064 and 532 nm. The coatings without any interlayer and with 0.1 quarter-wave ($\lambda/4$), 0.3 $\lambda/4$, 0.5 $\lambda/4$ SiO_2 interlayer were deposited respectively on LBO by using electron beam evaporation technique. All the prepared coatings with SiO_2 interlayer indicated satisfying optical behavior. This expanded our option for the thickness of an interlayer when coating on LBO substrate. The prepared films with SiO_2 interlayer showed better adhesion than that without any interlayer. The thickness of the interlayer affected the adhesion, the adhesion for the coating with 0.5 $\lambda/4$ SiO_2 interlayer was not as good as the other two.

OCIS codes: 220.0220, 310.6860, 160.4330.

Lithium triborate LiB_3O_5 (LBO) is a nonlinear-optical crystal that has high transmission in the ultraviolet (UV) region and a wide transparency range (from 160 to 3500 nm)[1]. In particular, the crystal has a high damage threshold, a large acceptance angle, and a small walk-off angle. All these properties, along with its mechanical hardness, chemical stability, and nonhydroscopicity, make LBO an attractive material for harmonic generation of high-power density laser beams[2−5].

To reduce the additional losses due to Fresnel reflection, it is necessary to deposit antireflection (AR) coatings on the crystal surfaces[6]. One of the most frequent coatings is frequency doubling, where AR is required at two wavelengths, one of which is twice the other, such as 1064 and 532 nm, 946 and 473 nm, 1319 and 660 nm, 1342 and 671 nm, and so on[7]. Compared with the commonly used glassy substrate, LBO is an anisotropic crystal and combines such properties as changing optical and thermal properties with the direction, partially soluble material which is characterized by its surface growing turbidly under long action of the atmosphere. All these cause additional difficulties in coating on the crystal and the coatings often show poor adhesion. Interlayer inserted between coating and LBO substrate is expected to improve greatly the adhesion. But in many designed coatings, the application of interlayer always makes the optical performance worse, so the thickness of interlayer has to be controlled strictly, which increases the difficulty of the production. In the other hand, the adhesion between coating and substrate can be affected by the thickness of the interlayer[8].

In this paper, frequency doubling coating, in which the thickness of SiO_2 interlayer can be added in a wide range, was designed on LBO substrate using the vector method. The fabrication result of the designed coating with different thicknesses of SiO_2 interlayer showed promising optical performance and the adhesion of these coatings after annealing was investigated.

The vector method is a valuable technique, especially in design work associated with antireflection coatings. The specific calculation using this method for frequency doubling coating on LBO was given by Tan *et al.*[7]. The coating, which was a three-layer quarter-wave stack, had two zeros at λ and $\lambda/2$ and the reference wavelength λ_0 was equal to $\frac{2}{3}\lambda$. When the refractive index of the LBO substrate n_s was set in 1.562, the designed residual reflectance at wavelength of 1064 nm could be 0.014% and 0.0004% at 532 nm in the absence of interlayer. Unfortunately, the optical performance was sensitive to the thickness of the interlayer. Besides, the designed index of the middle layer ($n_M = 1.69$) was larger than that of the available material Al_2O_3 ($n = 1.60$), which affected the experimental performance badly. Consequently, we established a four-layer quarter-wave frequency doubling AR coating instead of the usual three-layer one as a starting coating for interlayer-added consideration based on the vector method. The LBO crystals with dimension of $10 \times 10 \times 3$ (mm) were supplied by Fujian CASTECH Crystals, Inc. (CASTECH), China. The principal refractive indices n_X, n_Y, and n_Z at wavelength of 532 nm were 1.562, 1.578 and 1.589 respectively, which were measured by spectrophotometer[9]. Here, based on the vector method, we determined the indices of the materials for the four layers when the substrate index n_s was set in 1.562 and two zeros were situated at 1064 and 532 nm. The residual reflectance at this two wavelengths when n_s was equal to the other two indices was also calculated. The calculated indices and residual reflectance were shown in Table 1. It indicated that the error was acceptable for the other two indices of LBO when we established the indices of the materials only considering one direction of index.

For a coating on LBO substrate, a useful material of interlayer is SiO_2 in amorphous structure as interlayer,

1671-7694/2007/010060-03

Table 1. The Starting Four-Layer Frequency Doubling AR Coating Design for LBO without Any Interlayer

Air	n_1	n_2	n_3	n_4	n_s	R_{532}	R_{1064}	λ_0 (nm)
1.00	1.46	1.63	1.82	2.04	1.562	0	0	532
1.00	1.46	1.63	1.82	2.04	1.578	5.6×10^{-6}	1.8×10^{-5}	532
1.00	1.46	1.63	1.82	2.04	1.589	3.3×10^{-5}	5.8×10^{-5}	532

n_i: the refractive index of the ith layer from the air.

Table 2. Residual Reflectance at 1064 and 532 nm of the Designed Frequency Doubling AR Coatings with Different Thicknesses of SiO_2 Interlayer

Thickness of SiO_2 Interlayer ($\lambda/4$)	R_{532}	R_{1064}	Optimized Stack
0.1	1.7×10^{-5}	2.0×10^{-6}	0.1LHYAL
0.3	3.0×10^{-8}	1.0×10^{-7}	0.30L0.76H0.99Y0.99A1.23L
0.5	7.0×10^{-8}	1.0×10^{-8}	0.50L0.62H0.98Y1.0A1.36L
0.8	7.2×10^{-7}	3.0×10^{-8}	0.80L0.43H0.92Y1.0A1.54L
1.0	2.0×10^{-8}	1.0×10^{-8}	1.0L1.20H1.04Y1.42A0.41L

H, Y, A, L being n_1, n_2, n_3, n_4 respectively.

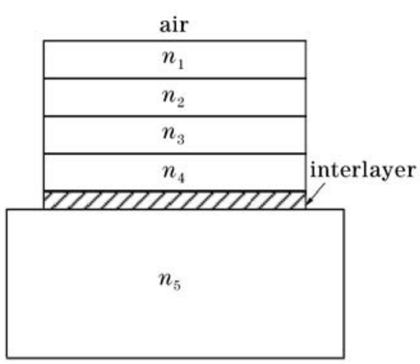

Fig. 1. Schematic diagram of a four-layer frequency doubling AR coating with interlayer between coatings and substrate.

which has been proved to improve the adhesion between coating and LBO by the formation of some compound. Here, different thicknesses of the SiO_2 interlayer were considered based on the starting four-layer quarter-wave stack. Figure 1 illustrated a four-layer frequency doubling AR coating with interlayer between coatings and substrate. The residual reflectance of coatings at 1064 and 532 nm with different thicknesses of SiO_2 interlayer and the optimized stacks were listed in Table 2.

Table 2 shows that the ideal residual reflectance at 1064 and 532 nm can be accessible by the adjustment of every layer except the interlayer. That is to say, theoretically the thickness of this four-layer stack can be changed in a wide range with satisfying optical behavior.

To confirm the design, some experimental examples were given subsequently. Frequency doubling AR coatings with two zeros at 1064 and 532 nm according to the above design were deposited on LBO crystals by electron beam evaporation. The thickness of the SiO_2 interlayer varied from 0 to 0.5. The materials used for each layer from LBO to air were silicon dioxide (SiO_2) as interlayer, zirconium dioxide (ZrO_2) for n_4, mixture of zirconium dioxide (ZrO_2) and yttrium trioxide (Y_2O_3) (4:1) for n_3, aluminium trioxide (Al_2O_3) for n_2, silicon dioxide (SiO_2) for n_1. All the materials were supplied by General Research Institute for Nonferrous Metals (GRINM), China, with purity of 99.99%. The substrates were cleaned ultrasonically in acetone, alcohol, and ligroin before coating. The base vacuum of all depositions was in 5.0×10^{-3} Pa. The substrates were rotated and heated to 503 K by a radiant heater for 3 h inside the vacuum chamber. This temperature was maintained during the deposition. The prepared coatings were annealed at a temperature of 473 K for 2 h. The reflectance of the coated LBO versus wavelength in the range of 400—1200 nm was measured by the Perkin Elmer lambda 900 spectrophotometer. Since many available methods for adhesion test are destructive, here, the adhesion properties of the series of prepared coatings are characterized by their performance after annealing. The micrographs of the coatings after annealing were observed by the Leica microscope with same magnification. The residual reflectances at wavelengths of 532 and 1064 nm were given in Table 3. The optical microscope results were presented in Fig. 2.

For frequency doubling AR coatings on LBO, a common criterion is that the residual reflectance at 1064 nm should be less than 0.2% and the transmittance at 532 nm should be larger than 95%. Obviously, the optical behavior prepared based on the above design satisfies the requirements. Because the optical monitoring technique is involved, the designed optical thickness of the film has to be adjusted to an integral number of quarter wavelengths, which causes large error between design and preparation. The deviation of the optical behavior between the design and experiment was mainly caused by

Table 3. Residual Reflectances at 1064 and 532 nm of the Prepared Frequency Doubling AR Coatings with Different Thicknesses of SiO_2 Interlayer

Thickness of Interlayer ($\lambda/4$)	R_{532} (%)	R_{1064} (%)
0	0.05	0.05
0.1	0.11	0.16
0.3	0.07	0.15
0.5	0.10	0.13

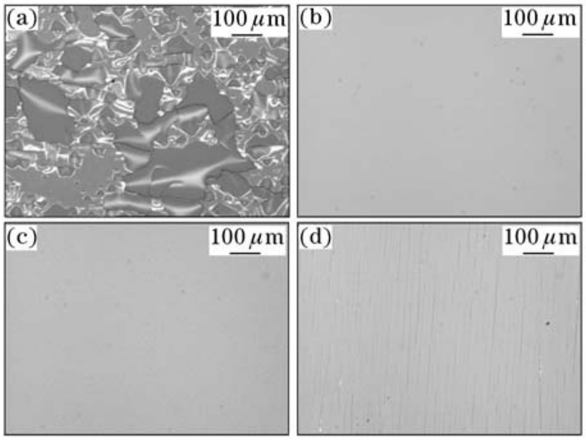

Fig. 2. Optical micrographs of the prepared frequency doubling AR coatings with different thicknesses of SiO_2 interlayer. (a) Without any interlayer; (b) 0.1 $\lambda/4$ SiO_2 interlayer; (c) 0.3 $\lambda/4$ SiO_2 interlayer; (d) 0.5 $\lambda/4$ SiO_2 interlayer.

the index difference of the hired materials from the ideal value. However, this inevitable deviation will be acceptable when the thickness of SiO_2 interlayer is less than 0.6 $\lambda/4$, which expands the option for the thickness of interlayer. Before annealing, it has been found that the coating without any interlayer is directly torn off after preparation, and there are no apparent cracks observed from the coatings with the three different thicknesses of SiO_2 interlayer, which indicates that SiO_2 interlayer can improve the adhesion between coatings and LBO substrate. However, after annealing, the coatings with different thicknesses of SiO_2 interlayer perform differently. The thickness of the interlayer affects the adhesion. The coating with 0.5 $\lambda/4$ SiO_2 interlayer shows some cracks and its adhesion is not as good as the other two. This is because the adhesion between coatings and substrate can be affected by interface composition and structure including any interlayer, and the coatings with different thicknesses of interlayer have different interface attributes.

In conclusion, based on the vector method, we have designed a four-layer quarter-wave frequency doubling coating for LBO substrate. Though optimization, the designed coating was characterized by that the thickness of SiO_2 interlayer could be changed in a wide range with ideal residual reflectance at the two given wavelengths. The result of the preparations of the four thicknesses of SiO_2 interlayer which were 0, 0.1, 0.3, and 0.5 $\lambda/4$ confirmed the design. All the experimental optical performances were satisfying. Furthermore, the coatings with the three different thicknesses of SiO_2 interlayer showed better adhesion than those without any interlayer. Besides, the coatings with different thicknesses of SiO_2 interlayer showed different adhesion performances after annealing and the adhesion for the coating with 0.5 $\lambda/4$ SiO_2 interlayer is not as good as the other two coatings containing interlayer.

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References

1. F. Xie, B. Wu, G. You, and C. Chen, Opt. Lett. **16,** 1237 (1991).
2. A.-Y. Yao, W. Hou, X.-C. Lin, Y. Bi, R.-N. Li, D.-F. Cui, and Z.-Y. Xu, Opt. Commun. **231,** 413 (2004).
3. H. Q. Li, H. B. Zhang, Z. Bao, J. Zhang, Z. P. Sun, Y. P. Kong, Y. Bi, X. C. Lin, A. Y. Yao, G. L. Wang, W. Hou, R. N. Li, D. F. Cui, and Z. Y. Xu, Opt. Commun. **232,** 411 (2004).
4. L. Liu and K. Nagashima, Opt. & Laser Technol. **31,** 283 (1999).
5. Z. Sun, R. Li, Y. Bi, X. Yang, Y. Bo, Y. Zhang, G. Wang, W. Zhao, H. Zhang, W. Hou, D. Cui, and Z. Xu, Opt. Commun. **241,** 167 (2004).
6. E. A. Levtchouk, V. V. Novopachine, and A. V. Shestakov, Proc. SPIE **3739,** 118 (1999).
7. T. Tan, J. Huang, M. Zhan, J. Shao, and Z. Fan, Chin. J. Laser (in Chinese) **33,** 242 (2006).
8. J. Tang, L. Feng, and J. S. Zabinski, Sur. & Coatings Technol. **99,** 242 (1998).
9. T. Tan, K. Yi, J. Shao, and Z. Fan, Chin. J. Lasers (in Chinese) **32,** 1678 (2005).