Wide bandwidth (100) GaAs/fluorides quarter-wavelength Bragg reflectors grown by molecular beam epitaxy

Z. Shi,[a] H. Zogg,[b] P. Müller,[b] I. D. Jung, and U. Keller
Swiss Federal Institute of Technology, Institute of Quantum Electronics, ETH Hönggerberg, CH-8093 Zurich, Switzerland

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Broadband quarter-wavelength Bragg reflectors that consist of periodic stacks of fluorides (CaF_2–BaF_2–CaF_2) and GaAs, centered at 1.4 μm, were grown by molecular beam epitaxy. Despite a total fluoride thickness as high as 720 nm, crack-free surface morphology was obtained. In this letter, we report a crack-free standard quarter-wavelength III–V semiconductor-fluoride Bragg reflector. With only three stacks, the bandwidth with reflectance above 95% is about 650 nm (1.15–1.80 μm), while, near the center wavelength, the reflectivity is as high as 99%. Both important wavelengths of 1.3 and 1.55 μm for optical communication are included in the very wide high reflectance plateau. These mirrors are expected to have wide applications for optical and optoelectronic devices. © *1996 American Institute of Physics.* [S0003-6951(96)02449-7]

Epitaxially grown distributed Bragg reflectors (DBRs) of high reflectance have been widely used in many active and passive optoelectronic devices. They include vertical cavity surface emitting lasers (VCSELs),[1] light emitting diodes (LEDs),[2] reflection modulators[3] and, recently, antiresonant Fabry–Perot saturable semiconductor absorbers (A-FPSA) for mode-locked solid-state lasers.[4] Some applications require not only high reflectance at a certain wavelength but also high reflectance bandwidth, e.g., LED and A-FPSA for producing laser pulses in the femtosecond regime.[5] Attention has been focused on GaAs/AlAs DBRs since high quality epitaxial layers for DBRs and other device layers can all be obtained in the same run. However, the reflectance bandwidth of GaAs/AlAs DBRs is basically limited by the ratio of their refractive indices of 1.21.[6] Progress in the expansion of the high reflectance bandwidth has included fabricating GaAs/$Ga_xAl_{1-x}As$ tandem DBRs[6] and chirped structures.[7,8] Disordered multilayers of alternating dielectric materials with random thicknesses, based on the localization of light for one-dimensional disordered systems, are also used to obtain a broadband optical reflection.[9] Drawbacks in these designs include (1) many layers must be grown which need rigorous control of each layer and which lead to increased absorption; (2) since these mirror structures are asymmetric, the reflections are, to a certain extent, out of phase depending on the deviation from the quarter-wavelength thickness.

$Ga_xAl_{1-x}As$/IIa fluorides are another material pair which can be grown epitaxially onto each other.[10–16] The advantage of this material system is that the ratio of refractive indices can be as high as 2.3 because of the low fluoride refractive indices. Standard $\lambda/4$ CaF_2/GaAlAs DBRs have been grown by molecular beam epitaxy (MBE), and promising results have been obtained.[10] However, to achieve good material quality for devices, two difficult points must be overcome. One is that the fluoride layers tend to crack when their total thickness exceeds about 300 nm. This is due to the thermal stress between both materials caused by the large difference in their thermal expansion coefficients. To alleviate this problem, an asymmetric design with thinner fluoride layers was proposed and grown with (Ga,Al)As/$(Ca,Sr)F_2$ by MBE.[11] This structure was reported to have a crack-free surface morphology. However, to take full advantages of standard $\lambda/4$ GaAs/fluorides DBRs, one needs to be able to grow such thick epitaxial crack-free fluorides. Furthermore, since these layers should be grown alternatively with GaAs, there is a second difficult point of getting mirrorlike surfaces of GaAs on fluoride which must be overcome.

In this letter, we use CaF_2–BaF_2–CaF_2 three-layer stacks as the fluoride $\lambda/4$ layer. The advantages of such a fluoride stack are: (1) The first thin CaF_2 layer on GaAs can be grown [100] oriented at relatively low temperature (about 400 °C), while BaF_2 can only be grown in [100] orientation when substrate temperature is higher than 580 °C.[16] This low substrate temperature can reduce the thermal lattice mismatch when the sample cools down to room temperature. (2) BaF_2 can be epitaxially grown on CaF_2 and has a smaller elastic constant, which is expected to help overcome the crack problem caused by the thermal mismatch strain.[17] Since in our experiments GaAs overgrowth on CaF_2 has better material quality than on BaF_2, the second thin CaF_2 is used. It is worthwhile to point out that, as soon as good quality GaAs can be grown directly on BaF_2, this thin CaF_2 layer can be omitted. Since the difference between the refractive indices of CaF_2 (1.42) and BaF_2 (1.46) is very small, the reflection at the CaF_2–BaF_2 interfaces can be neglected.

The mirror structure is shown in Fig. 1, where each fluoride $\lambda/4$ layer consists of CaF_2 (4 nm)–BaF_2 (232 nm)–CaF_2 (4 nm). The designed center wavelength is 1.4 μm. The thicknesses of the quarter-wavelength layers are calculated by using refractive indices for CaF_2, BaF_2, and GaAs of 1.42, 1.46, and 3.47, respectively. Theoretical simulations of such structures were performed using the standard matrix method with appropriate GaAs parameters.[18] The fluorides are assumed nonabsorbent in this wavelength range.

The structure was grown in a home built MBE system, equipped with CaF_2, BaF_2, Ga, and As_2 effusion cells. The

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[a] Electronic mail: shi@iqe.phys.ethz.ch

[b] AFIF at Swiss Federal Institute of Technology, ETH-Teil Technopark, CH-8005 Zurich, Switzerland.

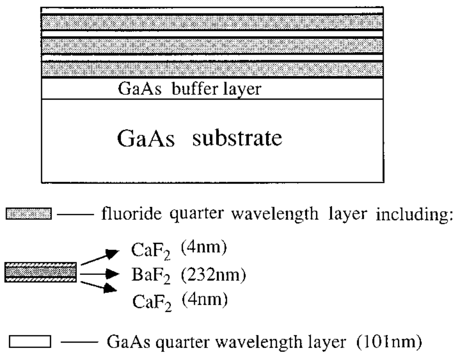

FIG. 1. Illustration of the grown structure and thickness of three stack GaAs/(CaF_2–BaF_2–CaF_2) quarter-wavelength Bragg reflector centered at 1.4 μm.

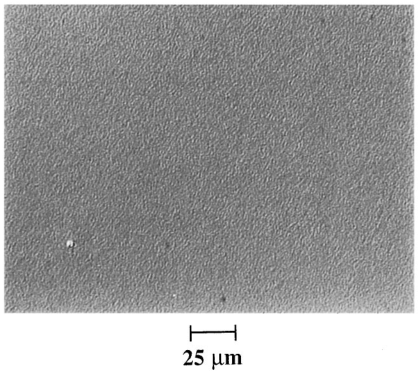

FIG. 2. Surface morphology of the top GaAs layer in the mirror structure observed in an optical microscope with Normarski interference contrast. A growth pit (lower left corner in the picture) is intentionally chosen to give the correct focus.

As_2 is produced by using GaAs effusion cell with a Ga trap cap.[19] The substrates are epiready semi-insulating 2-in GaAs (100) wafers. The substrate temperature is calibrated by the deoxidizing temperature (580 °C) of the GaAs substrate. After heating the substrate at 600 °C for 5 min under As_2 atmosphere, a 0.5 μm GaAs buffer layer is grown at 580 °C. The growth procedure of the CaF_2–BaF_2–CaF_2 is as follows: First, 4 nm CaF_2 is grown at 360 °C. The substrate temperature is then increased to 560 °C, and 6 nm BaF_2 layer is grown on top at this temperature. Afterwards, the substrate temperature is decreased back to 360 °C. The rest of the BaF_2 layer and the 4 nm CaF_2 top layer are grown at 360 °C. As pointed out by Stumborg *et al.*,[15] the BaF_2 overgrowth at low temperature will keep the [100] orientation due to the initial high temperature [100] BaF_2 layer. The growth rates of both CaF_2 and BaF_2 are 4 nm/min. GaAs is then overgrown on CaF_2 at a substrate temperature of 560 °C with a growth rate of 20 nm/min. The As_2/Ga flux ratio is about 40 for GaAs growth on CaF_2. During the temperature increase and decrease, the ramp rate is kept at 15 °C/min. The background pressure during fluoride growth is about 1×10^{-8} mbar. The procedure is repeated three times to obtain the mirror structure shown in Fig. 1. The exact growth rates of both materials were calibrated by fitting the experimental data of multilayer reflectance to the theoretical simulations. Reflection high energy electron diffraction (RHEED) was used in the *in situ* observation of the growth. All the layers grown show [100] orientation and good crystalline quality. Although the total fluoride thickness is 720 nm, these samples show, besides mirrorlike surface, a crack-free surface morphology (Fig. 2). To our knowledge, such crack-free stacks with such large thicknesses have not been reported before. The low growth temperature of the BaF_2 layers and the temperature cycling during the growth at different growth temperatures seem to be responsible for the crack-free nature. Further investigations are needed for a more detailed explanation.

The absolute reflectance spectrum of such a three pair fluoride-GaAs stack was measured with a Varian Cary 5E spectrophotometer and is shown together with theoretical simulation in Fig. 3. Near the center wavelength, the reflectivity is as high as 99%. The bandwidth with reflectance larger than 95% is about 650 nm, while that with reflectance larger than 90% is more than 750 nm. As can be seen in Fig. 3, the measured spectrum agrees well with the theoretical simulation. The minima of measured reflectance is about 7% higher than the simulated one. This is because the substrate is assumed to have infinite thickness in the simulation program. The overall measured spectrum was about 20 nm less than the calculated spectrum due to the thickness variation of about 5%.

Results reported here provide an excellent choice for devices requiring a high reflectance band, especially for those requiring perfect phase coherence as well, e.g., an A-FPSA. By using coated silver as the wideband rear mirror for the A-FPSA, self-starting Kerr-lens mode-locked pulses as short as 10 fs have been obtained with a Ti:sapphire laser.[5] Since A-FPSA with a wider reflection bandwidth and lower losses could be fabricated with the results reported here by using $Ga_{1-x}Al_xAs$/fluorides for the shorter wavelengths, one may expect shorter pulses to be produced. Another key feature is the crack-free surface which makes it possible to grow structures on top. It is expected that $Ga_{1-x}Al_xAs$/fluorides broadband reflectors could have wide applications for optical and optoelectronic devices. For applications in which higher reflectance is required, four or more stacks should be used. Work towards this goal is in progress.

In summary, crack-free standard quarter-wavelength GaAs/(CaF_2–BaF_2–CaF_2) mirror structures were fabricated. A very wide high reflectance bandwidth together with coher-

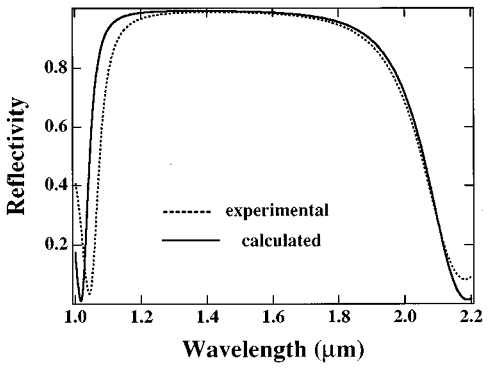

FIG. 3. Measured and calculated reflectance spectra of the structure shown in Fig. 1.

ent phase is obtained. Epitaxial layers of integrated structures can be grown on the crack-free surface. The excellent features of such mirror structures provide a promising choice for many optical and optoelectronic devices.

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[1] D. L. Huffaker, J. Shin, and D. G. Deppe, Electron. Lett. **30**, 1946 (1994).

[2] T. Kato, H. Susawa, M. Hirotani, T. Saka, Y. Ohashi, E. Shichi, and S. Shibaba, J. Cryst. Growth **107**, 832 (1991).

[3] H. Fouckhardt, M. Walther, Th. Hackbarth, and K. J. Ebeling, Opt. Laser Technol. **22**, 23 (1990).

[4] U. Keller, D. A. B. Miller, G. D. Boyd, T. H. Chiu, J. F. Ferguson, and M. T. Asom Opt. Lett. **17**, 505 (1992); U. Keller, Appl. Phys. B **58**, 347 (1994).

[5] R. Fluck, I. D. Jung, G. Zhang, F. X. Kärtner, and U. Keller, Opt. Lett. **21**, 743 (1996).

[6] W. I. Lee, Appl. Phys. Lett. **67**, 3753 (1995) and references therein.

[7] T. Saka, M. Hirotani, T. Kato, H. Susawa, and N. Hamauchi, J. Appl. Phys. **73**, 380 (1993).

[8] R. Szipöcs, K. Ferencz, C. Spielmann, and F. Krausz, Opt. Lett. **19**, 201 (1994).

[9] D. Zhang, Z. L. Li, W. Hu, and B. Y. Cheng, Appl. Phys. Lett. **67**, 2431 (1995).

[10] C. W. Tu, S. A. Ajuria, and H. Temkin, J. Cryst. Growth **81**, 545 (1987).

[11] C. Fontaine, P. Requena, and A. Munoz-Yagüe, J. Appl. Phys. **68**, 5366 (1990).

[12] P. W. Sullivan, J. E. Bower, and G. M. Metze, J. Vac. Sci. Technol. B **3**, 500 (1985).

[13] T. Minemura, J. Asano, K. Tsutsui, and S. Furukawa, J. Cryst. Growth, 114 (1991).

[14] W. Li, T. Anan, and L. J. Schowalter, Appl. Phys. Lett. **65**, 595 (1994).

[15] M. F. Stumborg, F. Santiago, T. K. Chu, J. L. Price, and J. R. Leibowitz, J. Appl. Phys. **77**, 2739 (1995).

[16] H. Clemens, U. Stromberger, P. C. Weilguni, and G. Bauer, J. Appl. Phys. **66**, 1680 (1989).

[17] See, for instance, H. Zogg, S. Blunier, A. Fach, C. Maissen, P. Müller, S. Teodoropol, V. Meyer, G. Kostorz, A. Dommann, and T. Richmond, Phys. Rev. B **50**, 10 801 (1994).

[18] L. R. Brovelli and U. Keller, Opt. Commun. **116**, 343 (1995).

[19] T. Shitara and K. Eberl, Appl. Phys. Lett. **65**, 365 (1994).