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Growth of novel broadband high reflection mirrors by molecular beam epitaxy

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Abstract

Novel broadband (Ga,Al)As/CaF_2 Bragg mirrors have been grown on Si(1 1 1) substrates for the first time providing large reflectance bandwidth due to a high ratio of refractive indices. Two types of interface morphology have been observed: a rough one when growing (Ga,Al)As on CaF_2 and a smooth one when growing CaF_2 on (Ga,Al)As. The effect of surface flattening due to CaF_2 overgrowth of (Ga,Al)As prevented the accumulation of interface roughness and provided a smoother surface of the top layer compared to that obtained from (1 0 0) oriented growth. Specular highly reflecting surfaces have been obtained showing no cracks. An absolute reflectance as high as 98% have been determined for a four pair Bragg mirror. © 1999 Elsevier Science B.V. All rights reserved.

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1. Introduction

Epitaxially grown distributed Bragg reflectors (DBR) of high reflectance have been widely used in many active and passive optoelectronic devices like semiconductor saturable absorber mirrors (SESAM), vertical cavity surface-emitting lasers (VCSEL) and light-emitting diodes (LED). Some of these applications require not only high reflectance at a certain wavelength but also high reflectance bandwidth for producing laser pulses in the femtosecond region [1]. The relatively small reflection bandwidth of semiconductor Bragg mirrors limit further pulse shortening. For example, the use of epitaxially grown III–V-semiconductors on GaAs substrates limit the bandwidth to about 60 nm for (Ga,Al)As/AlAs mirrors in the 800 nm region due to a refractive index ratio of only 1.21. However, reflectivity and spectral bandwidth of distributed Bragg mirrors increase rapidly with the ratio of the refractive indices of the materials forming the mirror. (Ga,Al)As/IIa-fluorides are material stacks, which can be epitaxially grown onto each other. With group IIa-fluorides (CaF_2, BaF_2, SrF_2) the ratio of refractive indices is as high as 2.3 due to the low refractive indices of the fluorides. Furthermore, a high reflectivity can be obtained with only four material pairs. In contrast to GaAs/(AlAs thermal

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oxide) DBR providing also large bandwidth due to a refractive index ratio of about 1.8, large area (Ga,Al)As/CaF_2 Bragg mirrors can be grown limited only by substrate size and thickness control.

Crystal quality, interface properties and surface morphology of the Bragg mirror will be influenced by both the high difference in the thermal expansion coefficients and the lattice mismatch of about 3.5% of III–V materials and IIa-fluorides. Crack-free GaAs/(CaF_2–BaF_2–CaF_2) Bragg mirrors of large reflectance bandwidth have been grown on GaAs(1 0 0) substrates only by applying extensive time-consuming temperature cycling [2]. Interface studies by RHEED revealed three-dimensional growth of CaF_2 and (Ga,Al)As as well [3]. Interface roughness accumulated during growth leading to surface roughness causing losses in reflectivity.

Our new approach in the crystal growth of Bragg mirrors was focused on the improvement of the interface quality by changing (I) the substrate orientation from (1 0 0) to (1 1 1) and (II) the substrate material from GaAs to Si. The $\{1\,1\,1\}$ planes are the preferred growth planes for the IIa-fluorides. Lattice and mismatch strain in the epitaxial IIa-fluoride layers relax due to dislocation gliding [4]. Furthermore, CaF_2 can be two-dimensionally grown on silicon. Additionally, Si substrates are inexpensive and easier to handle than GaAs substrates since there will be no As evaporation during heating up.

2. Experimental procedure

(Ga,Al)As/CaF_2 Bragg mirrors with a center wavelength of 780 nm have been grown on 3″ Si(1 1 1) $\pm 1^\circ$ substrates using a home-built MBE system. Quarter-wavelength CaF_2 and (Ga,Al)As layers (60–80% Al contents) have been grown at substrate temperatures of 740 and 540°C, respectively. Growth rates of 0.35 μm/h for CaF_2 and 0.55 μm/h for (Ga,Al)As were used. Exposure time of the impinging As_2 flux before (Ga,Al)As growth was varied from 10 to 90 s.

In situ RHEED and scanning electron microscopy (SEM) were used to study interface and surface morphology. Reflectance spectra of the Bragg mirrors have been measured using a Varian Cary 5E spectrophotometer.

3. Results and discussion

While a two-dimensional (1 1 1) oriented growth of the first CaF_2 layer on Si is easily achieved, the nucleation and growth of the following (Ga,Al)As layer will mainly proceed three-dimensionally for two reasons: (I) CaF_2(1 1 1) has a lower surface energy than (Ga,Al)As. The latter one tends to nucleate in a three-dimensional mode to maintain a minimum energy configuration as known for GaAs [5]. (II) The natural (fluorine terminated) CaF_2(1 1 1) surface has very low sticking coefficients for As. It will poorly wet the CaF_2 surface. In addition, a very narrow growth window is reported for the homoepitaxial and heteroepitaxial growth of GaAs(1 1 1) which will be valid for the growth of (Ga,Al)As too. To improve the surface wetting the CaF_2 surface was exposed to impinging As flux at a temperature of 540°C before (Ga,Al)As film growth. Gaseous AsF_3 will form and evaporate from the surface allowing the formation of the stronger Ca–As bonds [6–8].

Fig. 1a shows the RHEED pattern of the first quarter-wavelength CaF_2 layer grown on the Si substrate. The growth started and proceeded two-dimensionally. After lowering the substrate temperature the CaF_2 layer was exposed to impinging As_2. Subsequent overgrowth with (Ga,Al)As was determined as three-dimensional growth right from the beginning (Fig. 1b and Fig. 1c). Rotational twins can be easily formed in (1 1 1) oriented growth. They were found to be suppressed by growing at a substrate temperature of 540°C using an As_2 pressure lower than 10^{-5} mbar. In contradiction to the (1 0 0) oriented (Ga,Al)As growth where a high As/Ga flux ratio is necessary to achieve good quality material, a high As oversupply in (1 1 1) oriented growth causes beside twinning an increase in the surface roughness which is indicated by diminishing RHEED pattern intensity.

Studies of the next interfaces by in situ RHEED revealed two kinds of interface morphologies (1) the rough (Ga,Al)As/CaF_2 interface and (2) the flat CaF_2/(Ga,Al)As interface. The first one is rough

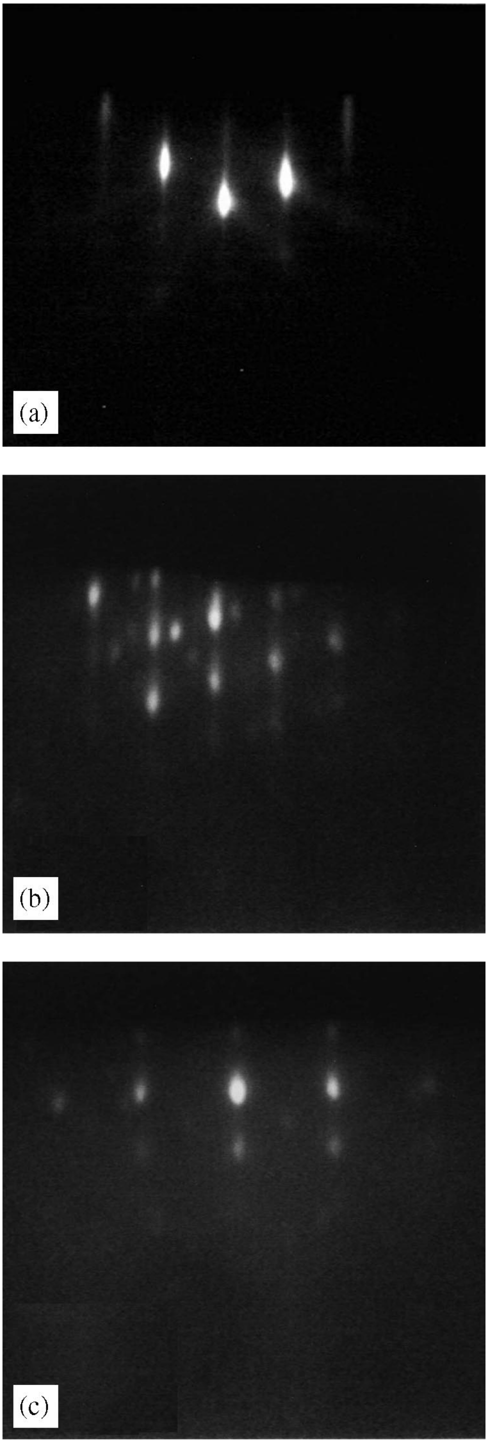

Fig. 1. RHEED pattern of a 2D grown CaF_2 layer (a) and a 3D grown (Ga,Al)As layer in $\langle 1\,\bar{1}\,1\rangle$ azimuth (b) and $\langle 1\,1\,\bar{2}\rangle$ azimuth (c).

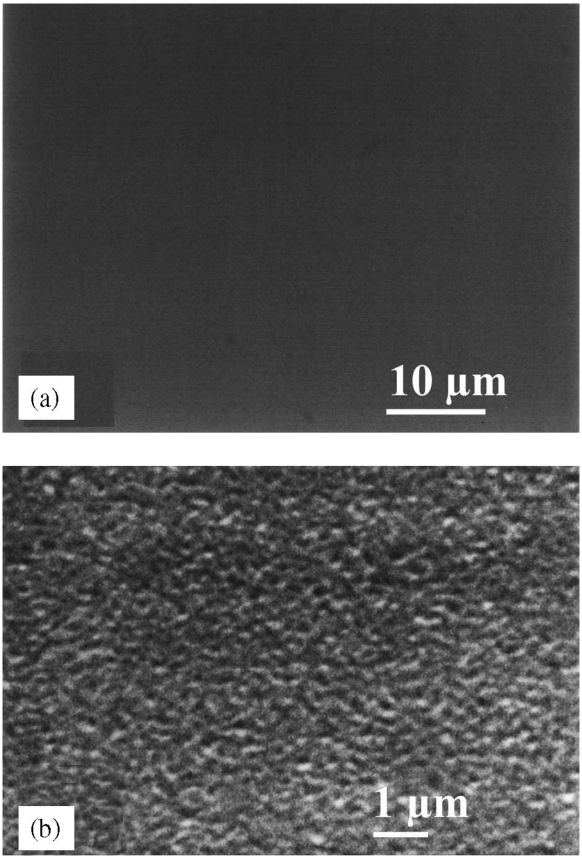

Fig. 2. Surface morphology of a (Ga,Al)As layer studied by optical microscopy (a) and SEM (b).

due to the three-dimensional growth of (Ga,Al)As on CaF_2 whereas the latter one showed a 2D pattern as shown in Fig. 1a. The growth of CaF_2 on a rough (Ga,Al)As surface always started 3D but finished 2D flattening the surface for the subsequent layer overgrowth. This effect of surface flattening by the CaF_2 growth avoided an accumulation of the interface roughness with increasing layer number as we found for the (1 0 0) oriented growth [3].

Despite the difference in the thermal expansion coefficients and the temperature change of 200°C due to the two different growth temperatures, specular surfaces showing no cracks have been observed (Fig. 2a). The surface structure of a 3D

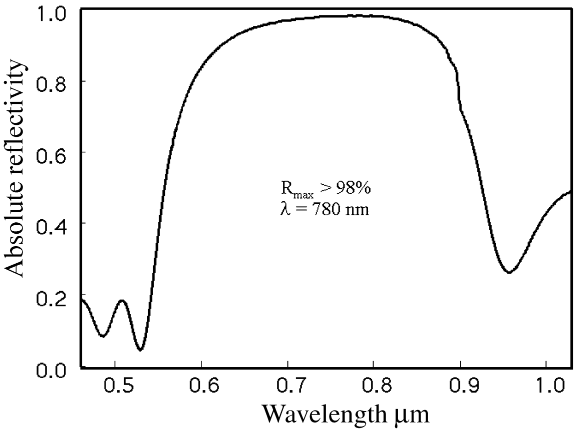

Fig. 3. Absolute reflectance spectrum of a four pair broadband Bragg mirror.

grown (Ga,Al)As layer was revealed by SEM and is shown in Fig. 2b, whereas only smooth surfaces were obtained from CaF_2 layers. The absolute reflectance for such a four pair broadband (Ga,Al)As/CaF_2 Bragg mirror is higher than 98% for a center wavelength of 780 nm (Fig. 3).

The interface flattening effect due to the CaF_2 growth will be applied to SESAM design. A broadband saturable absorber mirror with a CaF_2 top layer, e.g. the absorber layer embedded in two CaF_2 layers on top of a Bragg mirror, will have no surface scattering losses and will provide a very high reflectivity.

4. Summary

(Ga,Al)As/CaF_2 Bragg mirror of large reflectance bandwidth and high reflectivity have been grown on Si(1 1 1) substrates. Despite the three-dimensional growth of (Ga,Al)As on CaF_2 no accumulation of the interface roughness was observed. CaF_2 overgrowth of (Ga,Al)As started 3D but finished 2D smoothing the interface. An absolute reflectivity higher than 98% was achieved for a four pair Bragg mirror.

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