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# GaAs absorber layer growth for broadband AlGaAs/fluoride SESAMs

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## Abstract

Semiconductor saturable absorber mirrors (SESAMs) are used for ultrashort pulse generation. They are required to support self-starting sub-10-fs pulse generation with Ti:sapphire lasers. So far conventional AlGaAs/AlAs SESAMs have been limited by their small reflection bandwidth of about 60 nm. Broadband SESAMs with high reflection bandwidth were needed. In this paper, we report for the first time on the successful growth and operation of a broadband AlGaAs/$CaF_2$ SESAM with a high reflection bandwidth of more than 300 nm for ultrashort pulse generation with GaAs used as saturable absorber. We will demonstrate a pulse spectrum supporting sub-6-fs pulses obtained from an AlGaAs/$CaF_2$ SESAM in a Ti:sapphire laser. Furthermore, we have studied the formation of twin defects depending on the growth mode for the GaAs absorber layer heteroepitaxially grown on $CaF_2$. Defects in the GaAs absorber are necessary to achieve fast response times. However, they can generate surface roughness causing nonsaturable losses by light scattering. A GaAs saturable absorber was grown on a two pair AlGaAs/$CaF_2$ Bragg mirror with $CaF_2$ spacer layer without significant degradation of the saturable absorber parameters relevant for passive mode-locked solid-state lasers. © 2001 Elsevier Science B.V. All rights reserved.

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## 1. Introduction

Broadband semiconductor saturable absorber mirrors (SESAMs) are required to support self-starting sub-10-fs pulse generation with Ti:sapphire lasers [1]. Conventional AlGaAs/AlAs SESAMs limit further pulse shortening to 34 fs due their small high reflection bandwidth of 60 nm [2]. The bandwidth limitation can be overcome by replacing the lower Bragg mirror by a silver mirror [3,4]. However, the semiconductor saturable absorber cannot be epitaxially grown on a silver mirror or amorphous $SiO_2/TiO_2$. Therefore, postgrowth processing is applied to fabricate broadband SESAMs. To overcome the bandwidth limitation of conventional Bragg mirrors and to avoid postgrowth processing we have studied the heteroepitaxy of GaAs and AlGaAs on $CaF_2$ in multilayer stacks [5–7]. Recently, we have demonstrated an operating GaAs saturable absorber grown on $CaF_2$ by MBE [8,9]. In this paper, we report for the first time on the successful growth

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and operation of a broadband AlGaAs/$CaF_2$ SESAM with a high reflection bandwidth of more than 300 nm for ultrashort pulse generation with a GaAs absorber. We have demonstrated a pulse spectrum supporting sub-6-fs pulses obtained from an AlGaAs/$CaF_2$ SESAM in a Ti:sapphire laser.

The homoepitaxial growth of GaAs on (1 1 1) oriented substrates is more challenging than the growth on (1 0 0) oriented substrates since surface morphology and defect formation are very sensitive to the As/Ga flux ratio. The window for good epitaxial growth is very small [10–12]. Furthermore, heteroepitaxial growth of GaAs on a lattice mismatched material with very different linear thermal expansion coefficient, crystal structure and crystal chemistry such as $CaF_2$ is even more demanding [13–15]. Defects in the material are very important for the operation as a saturable absorber, since they are responsible for fast response times. While ion-implanted or low-temperature (LT) grown GaAs is used for conventional AlGaAs/AlAs SESAMs to achieve fast response times, the (1 1 1) oriented heteroepitaxial growth of GaAs on $CaF_2$ is already combined with the formation of a high number of defects. Here, the most important issue is to grow surfaces with low surface roughness to avoid nonsaturable losses by scattering light. We have studied the formation of twin defects depending on the growth mode for the GaAs on $CaF_2$. Since the thickness of the absorber layers is 30 nm or less, defects like rotational twins originated at the interface cannot grow out.

## 2. Experimental procedure

AlGaAs/$CaF_2$ SESAMs were grown by MBE on GaAs (1 1 1)B oriented substrates. $CaF_2$ growth was carried out at a substrate temperature of 620°C and a growth rate of 0.2 μm/h. $Al_xGa_{1-x}As$ layers with $x = 0.77$ were grown at 600°C with 0.7 μm/h. Growth was interrupted to anneal the surface under $As_2$ flux several times. An electron beam was used to modify the surface of the $CaF_2$ spacer layer for GaAs overgrowth. Details about electron modification were already reported elsewhere [9]. Cross sections of the SESAMs were studied by standard and high-resolution transmission electron microscopy (TEM, HRTEM). A Varian Cary 5E spectrophotometer was used to measure the spectral reflectivity. The absorber layer was tested by saturation fluence and degenerate time-resolved pump–probe measurements. All measurements were carried out at room temperature using an 80 MHz, 150 fs pulse train from a Ti : sapphire laser centered at 830 nm. A SESAM assisted Kerr-lens mode-locked Ti:sapphire laser described in Ref. [1] was operated by an AlGaAs/$CaF_2$ SESAM.

## 3. Results and discussion

AlGaAs and $CaF_2$ were epitaxially grown in stacks to fabricate the bottom Bragg mirror. The heteroepitaxial growth of the ternary compound AlGaAs on $CaF_2$ is less critical than the growth of the binary GaAs on $CaF_2$ for the absorber application. It seemed that Al has a higher wettability on $CaF_2$ than Ga, and the AlGaAs growth was less sensitive to the $As_2$ flux. A streaky RHEED pattern indicated nearly two-dimensional growth. However, TEM images revealed some surface roughness of the AlGaAs layers.

A crucial point during growth is always the growth interruption when the substrate temperature $T_1$ of the AlGaAs growth is increased to the substrate temperature $T_2$ of the $CaF_2$ overgrowth. During that time $As_2$ is impinging on the surface to prevent As from desorption. In (1 0 0) oriented growth, it is sufficient to apply the flux $F_{g,100}$ used for the previous AlGaAs growth. For the (1 1 1) oriented growth, $F_{g,111}$ is much smaller than $F_{g,100}$, but still higher than the flux needed to prevent As from desorption. However, the subsequent overgrowth of $CaF_2$ on an AlGaAs surface exposed to flux $F_{g,111}$ was accompanied by the formation of rotational twins already in the first monolayers. Since $CaF_2$ is known to form penetration twins according to spinell law and not rotational twins, it must have replicated surface features. Therefore, it is assumed that the rotational twins have already formed during annealing even though we did not notice any change in the RHEED pattern during the growth interruption. That means, that the $As_2$ flux $F_{g,111}$ used for the AlGaAs growth did not stabilize the surface. This result is supported by an observation during GaAs growth: growth interruptions only improve surface morphology under Ga-rich growth conditions. Otherwise rotational twinning was supported. Consequently, for the growth of $CaF_2$ on AlGaAs it is necessary to lower the $As_2$ flux to avoid long periods of growth interruptions. $CaF_2$ growth was initiated on a Ga-rich surface. Streaks were typically found in the RHEED. Electron channeling carried out on $CaF_2$/GaAs revealed that $CaF_2$ always grows "type A" under the chosen conditions. "B-type" epitaxy originates misfit dislocations in $CaF_2$ causing morphology problems [14,16].

The most challenging part of the growth was that of the GaAs absorber layer. We have already reported on the growth and characterization of a test structure where a GaAs absorber layer was sandwiched by two $CaF_2$ layers [9]. The next step was now to grow a GaAs absorber layer on top of AlGaAs/$CaF_2$ Bragg mirror. In this context, the increasing roughness with increasing number of layers has to be taken into account. A $CaF_2$ layer directly grown on the substrate provides a much better surface to grow on than the surface of a $CaF_2$ spacer layer grown on top of a two pair AlGaAs/$CaF_2$ Bragg mirror. A certain roughness always appears with increasing number of layers since the change in growth temperature for each layer introduces strain and misfit dislocations to the layers. To improve the wettability of GaAs on $CaF_2$ we modified the $CaF_2$ surface by an electron beam [9]. It was suggested that the electron beam removes the uppermost F-layer of $CaF_2$ while As is impinging on the $CaF_2$ surface allowing for the formation of stronger Ca–As bonds [17]. However, we noticed that the As on the $CaF_2$ surface always causes the formation of rotational twins. Rotational twinning can be considered as "B-type" epitaxy where the symmetry relation of the growing layer to the previous layer is determined by a rotation of 180° in the (1 1 1) plane and will be achieved by a disorder in the ABC sequence of the atomic layers in [1 1 1] direction. Rotational twins can be identified during growth by RHEED. They present themselves by double spots arranged one right below the other as shown in Fig. 1a. An example for rotational twinning at the $CaF_2$/GaAs interface is given by the HRTEM image in Fig. 2. The twins originate directly at the interface and do not continue from the $CaF_2$ layer.

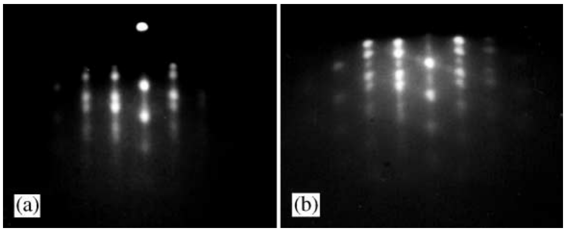

Fig. 1. Growth defects in GaAs grown on $CaF_2$ studied by RHEED: (a) rotational twinning, (b) microtwinning.

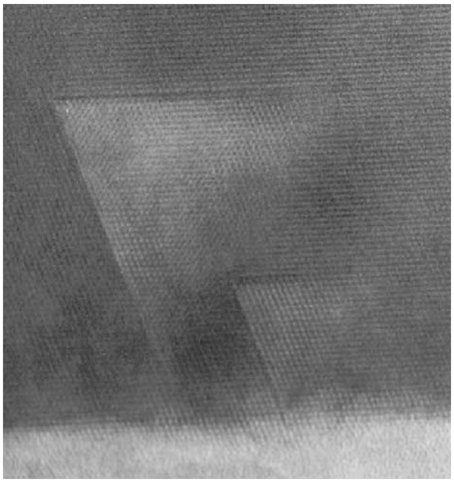

Fig. 2. Rotational twins originated at the GaAs/$CaF_2$ interface observed by HRTEM.

We now tried a different approach by growing Ga-rich. In that case, the electron modification of the $CaF_2$ surface was carried out without impinging As, and was extended to the deposition of the first monolayers. During the growth of the first monolayers microtwinning was noticed. A certain surface roughness was observed in the form of light scattering caused by a white light beam hitting the surface at normal incidence. This surface roughness was decreased by growth interruptions for annealing: for a short period of time (30 s to 1 min) only As was supplied to the surface. Additionally, microtwinning started to disappear.

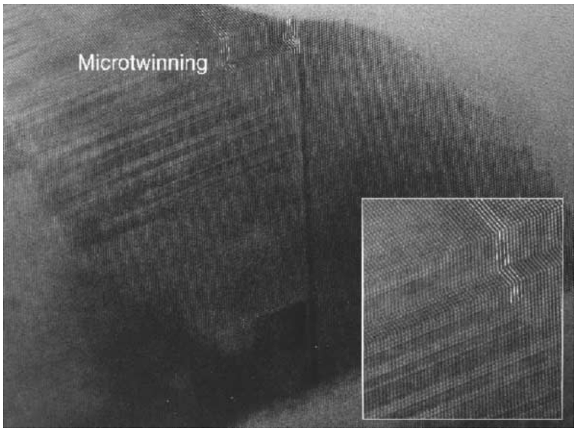

Fig. 3. Microtwinning in GaAs grown on $CaF_2$. Inset presents the enlarged microtwinned region.

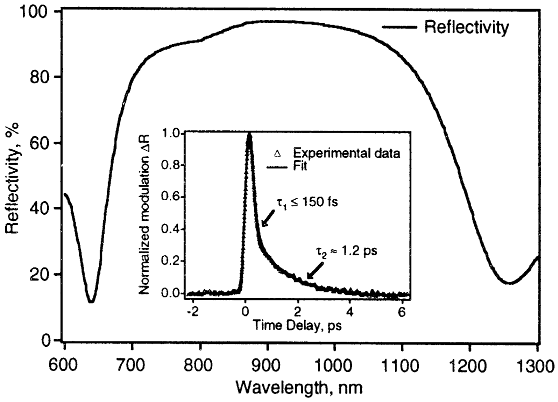

Fig. 4. Reflectivity spectrum of an AlGaAs/$CaF_2$ SESAM with GaAs absorber layer. The inset presents the differential reflectivity vs. time delay between pump and probe pulse.

Microtwinning can also be observed in situ by RHEED. Here, multiple spots arranged on a line and not pairs of spots are imaged on the RHEED screen (Fig. 1b). Microtwinning is the accumulation of a high number of twins consisting of only a few monolayers as shown in Fig. 3 by HRTEM. Microtwinning is always an indication for excess Ga. Opposite to rotational twinning, microtwinning can be overcome by annealing with As flux or by increasing the As/Ga flux ratio. Spectral reflectivity was measured to judge surface morphology in terms of reflectivity losses caused by scattering light. A reflectivity spectrum of a SESAM consisting of a 70/140 nm AlGaAs/$CaF_2$ Bragg mirror with 170 nm $CaF_2$ spacer layer and 40 nm GaAs absorber layer is shown in Fig. 4. A maximum reflectivity of more than 97% is obtained, which is less than 2% below the theoretical prediction. The high reflection bandwidth is 308 nm at 90% reflectivity. There is a pronounced drop in reflectivity due to the absorption of the GaAs top layer starting at 870 nm. It is higher than expected from the SESAM design, and can point to an increase in the absorption coefficient due to the high defect concentration in the absorber material. Measurements of the pulse-energy-dependent reflectivity of the SESAMs were carried out to verify the operation of the GaAs absorber layer. A linear reflectivity of 95.1% was measured at a wavelength of 830 nm. With a nonsaturable reflectivity $R_{ns}$ of 97.3% a modulation depth $\Delta R$ of 2.2% was obtained. Nonsaturable losses $\Delta R_{ns}$ of 2.7% were determined. A saturation fluence of $36\,\mu J/cm^2$ was achieved. Pump–probe measurements revealed a fast recovery time $\tau_1$ of less than 150 fs and a slower one $\tau_2$ of 1.2 ps needed for self-starting operation shown in the inset of Fig. 4. Small nonsaturable losses and a fast recovery time demonstrate that the growth conditions were chosen appropriately. Compared to the absorber in the test structure, the absorber on top of the Bragg mirror shows faster response times indicating a higher defect concentration. That result is assumed to be caused by the Bragg mirror underneath providing a less good surface to grow on compared to that in the test structure. Nonsaturable losses can be mostly attributed to scattering light caused by surface roughness and still need to be improved further.

The AlGaAs/$CaF_2$ SESAM successfully started and stabilized a SESAM assisted Kerr-lens mode-locked Ti : sapphire laser. A broad pulse spectrum with a transform limit of 5.9 fs was measured shown in Fig. 5. That is the first time that a AlGaAs/$CaF_2$ SESAM was fabricated and operated in a Ti : sapphire laser. The broad pulse spectrum demonstrates the great potential of AlGaAs/$CaF_2$ SESAMs for ultrashort pulse generation.

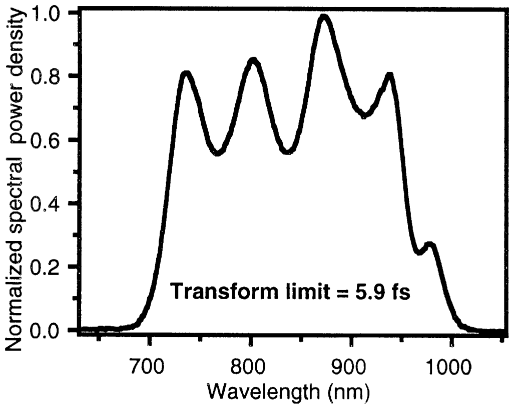

Fig. 5. Pulse spectrum of a AlGaAs/$CaF_2$ SESAM with GaAs saturable absorber measured in a SESAM assisted Kerr-lens mode-locked Ti:sapphire laser supporting sub-6-fs pulses.

## 4. Summary

We have showed for the first time the growth and operation of broadband AlGaAs/$CaF_2$ SESAMs with GaAs absorber layer grown by MBE. Due to the high reflectivity bandwidth of more than 300 nm and the fast response time of the GaAs absorber a broad pulse spectrum supporting sub-6 fs was measured. The GaAs absorber was grown Ga-rich causing less surface roughness than As-rich growth.

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