Fluoride semiconductor saturable-absorber mirror for ultrashort pulse generation

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Received March 12, 2002

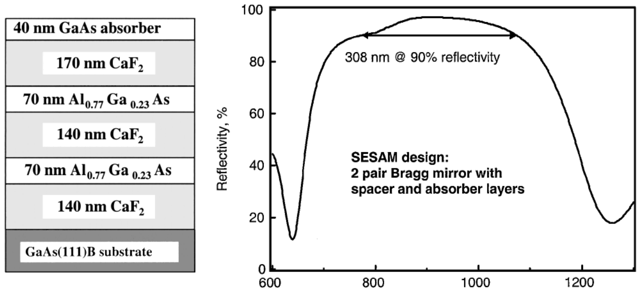
We demonstrate what is to our knowledge the first ultrabroadband monolithically grown AlGaAs/CaF$_2$ semiconductor saturable-absorber mirror (SESAM) that covers nearly the entire gain spectrum of a Ti:sapphire laser. A large high-reflectivity bandwidth of more than 300 nm is provided by a device consisting of only six material layers. This fluoride SESAM had a modulation depth of 2.2%, a fast recovery time constant of less than 150 fs, and a slow recovery time constant of 1.2 ps. Using this SESAM inside a Ti:sapphire laser produced self-starting sub-10-femtosecond pulses. © 2002 Optical Society of America

OCIS codes: 320.7080, 310.3840, 160.4330, 320.7100.

Semiconductor saturable-absorber mirrors (SESAMs) have been used extensively for passive pulse generation in solid-state lasers.[1,2] So far, generation of ultrashort pulses by use of monolithically grown SESAMs for center wavelengths near 800 nm have suffered from the bandwidth limitation of the AlGaAs/AlAs Bragg mirror. Pulse durations were limited to 34 fs.[3] A dielectric top reflector allowed for slightly shorter pulses, with 19-fs duration.[4] Therefore, increasing the reflection bandwidth with metal mirrors[5] and ultimately supporting sub-6-fs pulse generation required postgrowth processing.[6] Approximately 3% nonsaturable losses have typically been observed for silver bottom reflectors,[7] which can be reduced to ~1% by insertion of a dielectric layer between the semiconductor and the metal layers.[8] Without postgrowth processing, however, new ultrabroadband SESAMs are required for full exploration of the gain spectrum of a Ti:sapphire laser and, hence, for support of self-starting sub-10-fs pulse generation with Ti:sapphire lasers. To overcome the refractive-index limitation we studied the material pair AlGaAs and CaF$_2$. The big advantage of this compared with all other material choices was the large difference in the refractive indices of the two compounds, which are 1.43 for CaF$_2$ and 3.14 for AlGaAs with 77% Al at 800 nm.[9] Furthermore, fluoride is nonabsorbing, with no relevant dispersion in the wavelength range of interest. However, the two materials are different in their structural, chemical, and thermal properties, e.g., lattice constants and thermal expansion, which makes epitaxial growth in multilayer stacks challenging. Here, we demonstrate what is to our knowledge the first monolithically grown ultrabroadband AlGaAs/CaF$_2$ SESAM that covers nearly the entire Ti:sapphire gain spectrum and thus supports sub-10-fs pulse generation.

SESAM devices were grown by solid source molecular beam epitaxy upon GaAs (111)B-oriented substrates. (111) orientation was chosen because it allows for strain relaxation owing to dislocation gliding in CaF$_2$.[10] The linear coefficient of thermal expansion of CaF$_2$ is $19.2 \times 10^{-6}\,\mathrm{K}^{-1}$, i.e., three times higher than that of GaAs. High thermal strain is added to the strain already caused by the 3.5% lattice mismatch, which results in cracking of the layers during cooling from the growth temperature for (100)-oriented samples.[11,12] However, the successful strain management by growth on (111)-oriented substrates prevented cracks and guaranteed a long-term stability of more than two years. CaF$_2$ growth was carried out at a substrate temperature of 600 °C and at a growth rate of 0.2 μm/h. AlGaAs layers with 77% aluminum concentration and the GaAs absorber layer were grown at a rate of 0.7 μm/h at 600 °C. We interrupted growth to anneal the surface under As$_2$ flux several times. The CaF$_2$ surface was exposed to a high-energy electron beam of 20 keV at grazing incidence before the GaAs absorber overgrowth because the fluorine-terminated surface showed improved wettability for GaAs after exposure to an electron beam.[13]

A two-pair Bragg mirror consisting of 140-nm CaF$_2$ and 70-nm Al$_{0.77}$Ga$_{0.23}$As with a 170-nm CaF$_2$ spacer layer and a 40-nm GaAs saturable-absorber



(a) (b)

Fig. 1. (a) Design and (b) reflectivity spectrum of an ultrabroadband AlGaAs/CaF$_2$ SESAM with a GaAs semiconductor saturable absorber. The reflectivity spectrum was measured with a Cary 5E photospectrometer with a standard silver mirror of 97% reflectivity used as a reference.

0146-9592/02/201845-03$15.00/0

layer on top has been designed and monolithically grown [Fig. 1(a)]. This SESAM has a bandwidth of more than 300 nm at 90% reflectivity for the linear reflectivity, as shown in Fig. 1(b), which is approximately five times larger than that of conventional AlGaAs/AlAs SESAMs with a much smaller number of mirror pairs. Note that this antiresonant SESAM has neglible group delay dispersion with less than 20 fs^2 over the entire high-reflection bandwidth. The GaAs saturable-absorber layer showed higher absorption than had been calculated for the linear reflectivity, which indicates that the high concentration of defects in the GaAs increased the absorption coefficient of the material. Growth of a GaAs layer upon fluoride in (111) orientation followed the Volmer–Weber growth mode as a result of the low free surface energy of the fluoride surface and the lattice mismatch. Therefore it provided a high concentration of defects at typical growth temperatures. Hence no further procedures were necessary to shorten the lifetime. In contrast, conventional SESAMs based on AlGaAs/AlAs require low-temperature growth or ion implantation of the GaAs saturable absorber to introduce defects for fast response times. The absorber layer was studied by saturation fluence and pump–probe measurements. All measurements were carried out at room temperature with an 80-MHz, 150-fs pulse train from a Ti:sapphire laser centered at 830 nm. The saturable absorber is described in terms of modulation depth ΔR, nonsaturable losses ΔR_{ns}, saturation fluence F_{sat}, and impulse response or recovery time τ_A.[2,14] The modulation depth is the maximum amount of saturable loss that can be bleached. The saturation fluence describes the light intensity that is necessary for bleaching the saturable losses. The recovery time is a measure of the carrier relaxation processes. The approximately bitemporal recovery time has two components that are attributed to carrier thermalization and to trapping–recombination. For the AlGaAs/$\mathrm{CaF_2}$ SESAM a linear reflectivity of 95.1% was measured, which is less than the calculated value of 96.1%. The loss of 1% in reflectivity may be caused by light scattering at the surface owing to the Volmer–Weber growth mode. A saturated reflectivity of 97.3%, a modulation depth of 2.2%, and nonsaturable losses of 2.7% have been obtained (see Fig. 2). These losses can be partially attributed to the mirror design, to surface roughness, and to additional absorption caused by defect states. The SESAM consists of only two mirror pairs, spacer and absorber layers with a calculated saturated reflectivity of 99%. Thus nonsaturable losses of 1% are already provided by the mirror design. Together with the observed reduction in linear reflectivity of 1% by light scattering, design and fabrication of the SESAM produce 2% nonsaturable losses. Therefore the nonsaturable losses caused by absorption by additional defect states were estimated to be less than 0.7%. A saturation fluence F_{sat} of 36 $\mu\mathrm{J/cm^2}$ was measured for the AlGaAs/$\mathrm{CaF_2}$ SESAM device, which can be compared with that of standard AlGaAs/AlAs SESAMs. A biexponential recovery time was measured, as demonstrated in Fig. 3. The first time constant, of ~150 fs, is attributed to thermalization limited by the temporal resolution of the experimental setup. The second time constant describes the process of carrier trapping. This fast time response of the GaAs saturable absorber is excellent for application of the saturable absorber as an all-optical switch in generation of ultrashort pulses. So far, the defect mechanism responsible for the fast carrier trapping has not been revealed. However, it can be assumed that the defects are different from those found in low-temperature GaAs because growth at high substrate temperatures does not allow for the incorporation of excess arsenic. Preliminary studies by high-resolution transmission electron microscopy and x-ray diffraction indicate a high number of stacking faults and rotational twins.[15]

For generation of short pulses, the SESAM was inserted into the cavity of the Kerr-lens mode-locked Ti:sapphire laser described in Ref. 6. The AlGaAs/$\mathrm{CaF_2}$ SESAM successfully started and supported mode locking in the laser. The pulse buildup time in the given configuration was less than 1 s. First experiments with the laser yielded output powers of as much as 180 mW for a pump

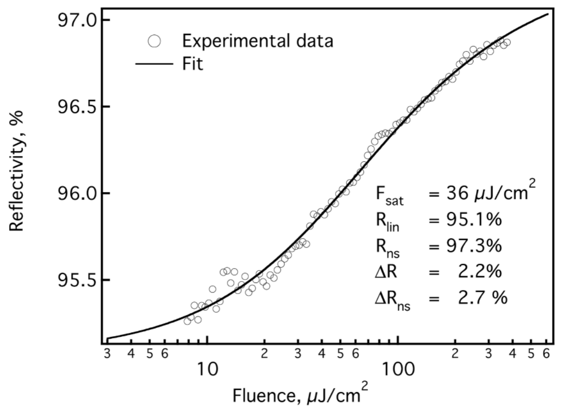


Fig. 2. Nonlinear reflectivity versus pulse energy fluence. A modulation depth ΔR_{lin} of 2.2% and nonsaturable losses ΔR_{ns} of 2.7% were measured.

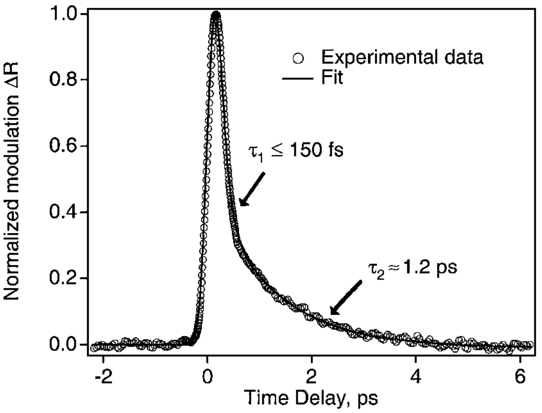


Fig. 3. Nonlinear reflectivity modulation ΔR as a function of time measured with a standard degenerate pump–probe setup at 830 nm. Biexponential recovery times of ~150 fs (thermalization) and ~1.2 ps (carrier trapping–thermalization) were measured.

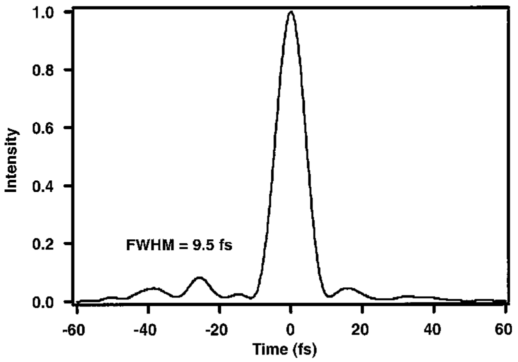


Fig. 4. Temporal pulse shape reconstructed by SPIDER of a sub-10-fs pulse generated by a Kerr-lens mode-locked Ti:sapphire laser with an AlGaAs/CaF_2 SESAM. The corresponding pulse spectrum has a transform limit of 8.2 fs.

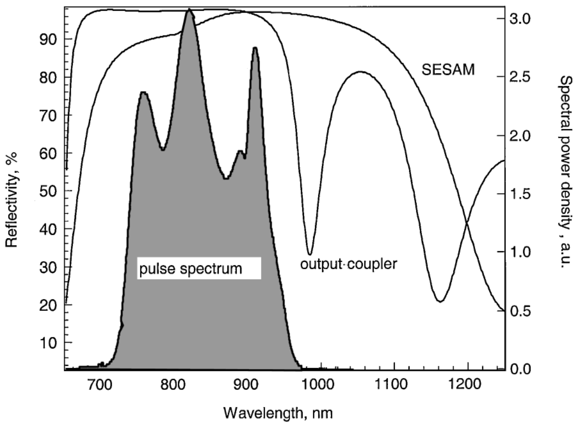


Fig. 5. Reflectivity spectra (left axis) of the AlGaAs/CaF_2 SESAM and of the output coupler and pulse spectrum (right axis) with a transform limit of 8.2 fs.

power of 5 W at 532 nm. In another oscillation pumped at 9.5 W, we obtained 380-mW mode-locked output power using the SESAM compared with 450-mW cw output with a high reflector instead. We obtained self-starting pulses with a pulse spectrum supporting transform-limited 8.2-fs pulses. Spectral phase interferometry for direct electric-field reconstruction (SPIDER) measurements[16] determined a pulse duration of 9.5 fs (Fig. 4). For the first time to our knowledge, the generation of ultrashort pulses below 10 fs with an AlGaAs/CaF_2 SESAM has been demonstrated. Further pulse shortening was limited only by the laser cavity configuration. In Fig. 5 the reflectivity spectra of the AlGaAs/CaF_2 SESAM and of the output coupler, in addition to the pulse spectrum, are drawn. The bandwidth of the output coupler clearly limits the pulse spectrum toward longer wavelengths and thereby demonstrates the potential for the generation of shorter pulses.

In conclusion, we have demonstrated a novel ultrabroadband SESAM based on an AlGaAs/CaF_2 material system with a GaAs saturable absorber. This SESAM device supports sub-10-fs pulse operation in a Ti:sapphire laser.

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