Mode locking using a type II multiple-quantum-well structure as a fast saturable absorber

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We demonstrate the application of a type II $Al_xGa_{1-x}As/AlAs$ multiple quantum well as a fast saturable absorber in a hybridly mode-locked dye laser. Type II multiple quantum wells are promising for this application because of the fast recovery of the saturated absorption with picosecond or even subpicosecond time constants. We obtain almost transform-limited pulses as short as 0.9 psec for a type II sample with a recovery time of 2.3 psec.

Passive or hybrid (active–passive) mode locking requires saturable absorbers with recovery times τ_a that are short in comparison with the recovery time τ_g of the saturable gain.[1,2] Generally, suitable dyes are used in passive or hybrid mode-locked dye laser systems, such as DODCI in combination with Rhodamine 6G in the colliding-pulse mode-locked laser. Semiconductor materials are also attractive for saturable absorbers because of the large optical nonlinearities that can be obtained, particularly in quantum-well structures.[3] However, in high-quality materials the recovery time of the saturated absorption is quite slow owing to the long carrier recombination time constants, which are in the nanosecond range for materials such as GaAs or InP, related alloys, and quantum-well structures. Thus applications as saturable absorbers for mode locking at high repetition rates as well as applications in optical processing, where fast switching at high bit rates is required, are not always possible. To overcome this the recombination lifetime can be reduced by creation of defects, e.g., by ion bombardment, which results in an increased concentration of nonradiative centers.[2] In fact, mode locking of a GaAs semiconductor laser at a repetition rate of 2 GHz with a proton-bombarded quantum-well saturable absorber has been reported, and pulses of 1.6 psec have been achieved.[2] Much shorter pulses, down to 120 fsec, have been obtained by passive mode locking of color-center lasers with HgCdTe multiple-quantum-well (MQW) saturable absorbers.[4]

Saturable absorbers with $\tau_a < \tau_g$ are called either slow or fast depending on whether the recovery time of the bleached absorption is long or short compared with the mode-locked pulse duration.[1,2,5] A slow saturable absorber is able to suppress only the leading part of a pulse, whereas the trailing edge can only be suppressed by saturation of the gain.[2] In contrast, a fast saturable absorber is able to suppress both the leading and the trailing edges of the laser pulses without the help of gain saturation.[5,6]

In this Letter we demonstrate that type II $Al_xGa_{1-x}As/AlAs$ MQW structures behave as fast saturable absorbers and that no further treatment for the reduction of recombination lifetimes is required. We employ a type II $Al_xGa_{1-x}As/AlAs$ MQW sample for active–passive mode locking of a DCM dye laser pumped by a mode-locked frequency-doubled Nd:YAG laser. We obtain almost transform-limited pulses as short as 0.9 psec for this type II sample with a recovery time of 2.3 psec. Much shorter laser pulses should be possible using all binary type II GaAs/AlAs short-period superlattices, since their recovery times of the bleached absorption can be as fast as 100 fsec.[7]

The nonlinearity responsible for saturable absorption in semiconductors is due to exciton bleaching. In semiconductor quantum wells like GaAs/AlGaAs electronic states are confined in one dimension, which results in an increase of exciton binding energy and oscillator strength compared with that of bulk material. In the normal, so-called type I structures, the upper valence band and lowest conduction band states are spatially located in the material with the smaller band gap, i.e., electrons and holes are confined in the same slab, e.g., in GaAs if the type I GaAs/AlGaAs structures are considered. The recovery time of the optical nonlinearity in these type I structures is generally determined by the recombination lifetime, which is of the order of 1 nsec at room temperature. Type II structures, in contrast, are characterized by a spatial separation within the different slabs of the quantum-well structure of the upper valence band and the lowest conduction band states. The lowest-lying electronic conduction band states of type II $Al_xGa_{1-x}As/AlAs$ MQW structures originate from X-conduction band states of the indirect-gap AlAs material[8] and thus are basically confined in the AlAs barrier material. The upper valence band states with Γ symmetry, however, are confined to the AlGaAs well. The absorption spectrum in these type II samples is governed by the direct optical transitions at higher energies, involving the Γ valence band and the X-conduction band states of $Al_xGa_{1-x}As$, since the oscillator strength for the indirect Γ–X transition is smaller by orders of magnitude.[8] Optical pump and probe experiments at the resonance peak of the lowest-lying direct heavy-hole excitonic transition reveal a partial recovery of the bleached absorption with a picosecond or even subpicosecond time constant[7] that is much

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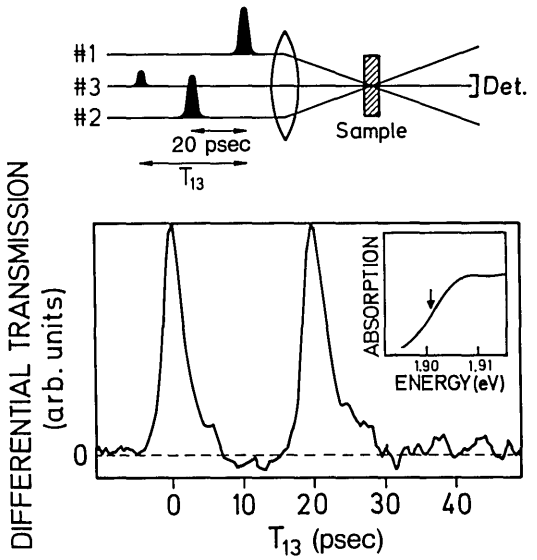

Fig. 1. Differential transmission of a type II MQW sample (pump and probe experiment) at $T = 300$ K. The sample is excited by two pump pulses separated by 20 psec. The laser photon energy is at the spectral position indicated by the arrow in the absorption spectrum shown in the inset.

smaller than the carrier lifetime. This fast response of the excitonic nonlinearity is due to scattering of the electrons from the Γ-related states of the $Al_xGa_{1-x}As$ layer to the X-related states of the AlAs layer.

If the laser photon energy is tuned to the lower-energy part of the direct band-gap excitonic resonance, a complete recovery of the bleached absorption is observed. As an example, this effect is illustrated in Fig. 1 for a type II $Al_xGa_{1-x}As$/AlAs MQW sample consisting of 25 periods of 68 monlayers of $Al_{0.37}Ga_{0.63}As$ and 36 monolayers of AlAs (monolayer thickness $a_0 = 0.283$ nm). This sample was mounted on sapphire, and the GaAs substrate was removed by selective chemical etching. The surface of the MQW sample was antireflection coated. Laser pulses from a cavity-dumped hybridly mode-locked DCM/DQTCI dye laser[9] with a pulse duration of 1.5 psec are used for this pump and probe experiment at a repetition rate of 500 kHz. The laser photon energy in this experiment is indicated by the arrow in the room-temperature absorption spectrum illustrated in the inset of Fig. 1. As depicted in the upper part of Fig. 1, the pump and probe experiment is performed with two pump pulses separated by 20 psec. Strong bleaching of the absorption with an instantaneous increase and a fast and complete recovery occurs when the probe pulse coincides in time with one of the pump pulses. The complete recovery of the bleached absorption in the low-energy part of the band-gap excitonic resonance is due to an enhanced collision broadening of the excitonic resonance after the spatial Γ–X transfer of electrons.[10] Consequently, the response is determined by the Γ–X transfer time of ~2 psec for this particular type II sample at room temperature.[7] Obviously, the response to the two successive pump pulses is virtually identical, demonstrating that this type II MQW sample fulfills the requirements for a fast saturable absorber, i.e., to provide loss for both the leading and the trailing edges of a mode-locked pulse and to permit operation at high repetition rates.

In order to check the application for mode locking we replaced the organic saturable absorber DQTCI in the Coherent 702 dye laser with DCM as the gain medium by the type II MQW sample, as schematically depicted in Fig. 2. A frequency-doubled actively mode-locked Nd:YAG laser is used for synchronous pumping (80-psec pulse width, 76-MHz repetition rate, 1.2-W cw pump power). A one-plate birefringent filter allows one to tune the laser emission wavelength. The optical spectrum as well as the intensity autocorrelation using phase-matched second-harmonic generation are measured for the characterization of the dye-laser pulses.

The autocorrelation and the optical spectrum of the synchronously mode-locked DCM laser without a saturable absorber in the resonator are depicted in Fig. 3. The autocorrelation [Fig. 3(a)] exhibits a sharp spike at $t = 0$, which indicates a rapidly varying substructure of the laser pulse.[11] The broad peak reflects the total duration t_p of the light pulses. We obtain $\tau_p = 4.9$ psec assuming a secant-hyperbolic pulse shape. The corresponding emission spectrum [Fig. 3(b)] reveals a spectral width (FWHM) of 7.5 meV, which results in a time–bandwidth product of $\tau_p \Delta\nu = 8.9$. Obviously,

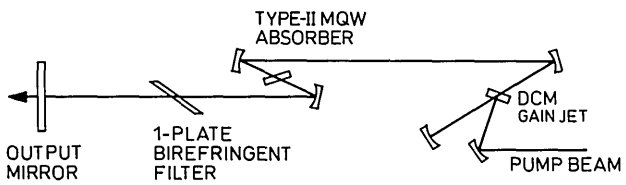

Fig. 2. Setup for the mode-locking experiment.

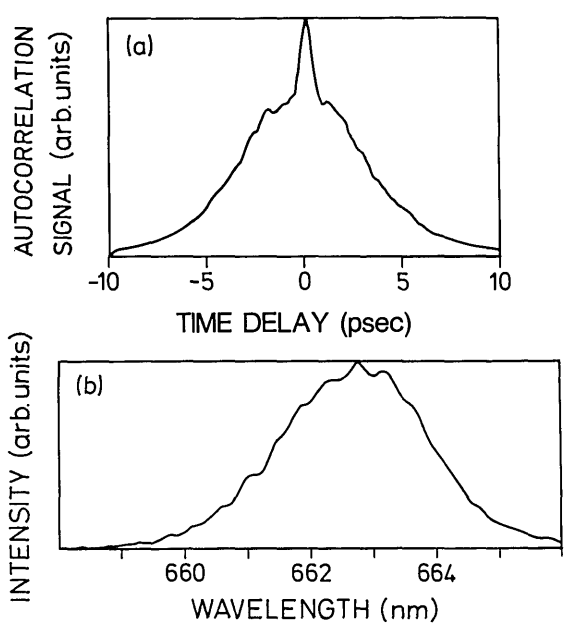

Fig. 3. (a) Intensity autocorrelation trace and (b) the optical spectrum of the mode-locked dye-laser output without a saturable absorber.

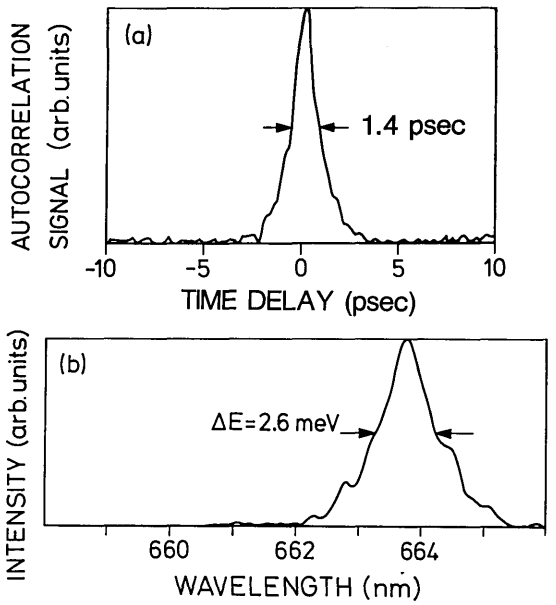

Fig. 4. (a) Intensity autocorrelation trace and (b) the optical spectrum of the mode-locked dye-laser output with the type II MQW absorber installed.

the pulses are far from being transform limited. This temporal and spectral behavior of the dye-laser output is maintained over the whole tuning range from 620 to 690 nm. The average cw output power without a saturable absorber amounts to 120 mW at an emission wavelength of ~650 nm.

The operation characteristics of the dye laser drastically change when the type II MQW sample is installed as a saturable absorber. The corresponding intensity autocorrelation and the optical spectrum of the dye-laser output are shown in Figs. 4(a) and 4(b), respectively, for the same position of the birefringent filter as in the previous case without a saturable absorber. At this position of the birefringent filter the shortest laser pulses are obtained. If we assume the pulse shape to be secant hyperbolic,[5,11] a pulse duration of 0.9 psec is derived from the autocorrelation trace in Fig. 4(a). Together with the spectral width of 2.6 meV as seen in the optical spectrum, a time–bandwidth product of $\tau_p \Delta\nu = 0.57$ is obtained. Consequently, the dye-laser pulses are nearly transform linked without any additional chirp corrections. The average cw output power with the type II saturable absorber installed is 5 mW. A higher output power seems feasible with improved antireflection coating of the MQW sample (in the present experiments only the front surface had been coated) and by careful optimization of the thickness of the absorbing layers of the structure.

A comparison of the optical spectra of Figs. 3(b) and 4(b) demonstrates that the direct band-gap absorption profile of the type II MQW sample forces the dye laser to operate at longer wavelengths, which indicates that the dye laser is operating in the low-energy part of the type I excitonic resonance. Consistently, the laser pulses broaden for operation at longer wavelengths, where the type II MQW absorber becomes transparent, whereas for shorter wavelengths the stability of the pulses decreases and the dye laser ceases to operate at wavelengths shorter than 650 nm. However, the central wavelength in Fig. 4(b) is ~8 nm longer than the wavelength indicated by the arrow in the inset of Fig. 1. We believe that the intracavity dye-laser power of 50 mW focused to a spot of ~30 μm on the type II MQW sample leads to appreciable local heating of the sample, which shifts the absorption spectrum of the type II MQW sample to longer wavelengths. This thermal shift was also observed in mode-locking experiments using type I MQW absorbers.[2]

In conclusion, we have demonstrated that type II MQW structures can be employed as saturable absorbers for mode locking of dye lasers without any further treatment for reduction of recombination lifetimes. We obtain almost bandwidth-limited pulses with pulse widths of 0.9 psec in a hybridly mode-locked DCM dye laser using a type II $Al_xGa_{1-x}As/AlAs$ MQW absorber with a recovery time of 2.3 psec. By using type II GaAs/AlAs short-period superlattices with subpicosecond Γ–X transfer times as short as 100 fsec (Ref. 7) as fast saturable absorbers it should be possible to produce much shorter laser pulses.

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