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Femtosecond response time in beryllium-doped low-temperature-grown GaAs/AlAs multiple quantum wells

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We have investigated optical nonlinearity in beryllium-doped low-temperature (LT) molecular-beam-epitaxy-grown GaAs/AlAs multiple quantum wells (MQWs). The response time of the nonlinearity is reduced by Be doping in the MQW. While the undoped LT MQW shows a 0.7–0.9 ps response, the response time of the Be-doped LT MQW is as short as 0.25 ps. The saturation density of the Be-doped MQW is almost the same as that of the undoped MQW, and is smaller than that of bulk GaAs. These results demonstrate that the Be-doped LT MQW exhibits a faster response than the undoped LT MQW, and a faster response as well as larger nonlinearity than LT bulk GaAs. © *2001 American Institute of Physics.* [DOI: 10.1063/1.1390478]

Optical nonlinear materials having an ultrafast photoresponse are indispensable in fabrication of optical devices necessary for ultrafast optical information processing technology and ultra-high-speed optical communication. Low-temperature (LT) molecular-beam-epitaxy (MBE) grown III–V semiconductors have attracted much attention because of the potential for sub-ps response time and large optical nonlinearity.[1–3] In the recent letter, we demonstrated the larger nonlinearity of undoped LT GaAs/AlAs multiple quantum wells (MQWs) as compared to bulk GaAs, and reported their picosecond dynamics.[4] The obtained minimum response time was 0.9 ps in the 310 °C-grown MQW, and further improvement is required.

Beryllium doping has been reported to reduce the response time in LT bulk GaAs (Refs. 5 and 6) and LT InGaAs/InAlAs MQWs.[7] However, a complex dependence of decay time on the Be-doping level was observed in time-resolved photoluminescence in LT bulk GaAs.[8] Furthermore, no paper, except Ref. 5, reports Be-doping dependence of the magnitude of the optical nonlinearity, though it is another important property. In addition, there has not been a published paper describing the effect of Be doping on the dynamics of the nonlinearity in the most fundamental material system such as the GaAs/AlAs MQW. In this letter, we study the femtosecond dynamics and the magnitude of the nonlinearity in the Be-doped LT GaAs/AlAs MQW. By comparing the results of the undoped MQW, the Be-doped MQW shows a reduced response time and a negligible amount of a slow-decay component. Saturation optical density F_{sat} of the Be-doped MQW is the same as that of the undoped one up to the modest Be-doping concentration, and it is smaller than that of Be-doped bulk GaAs. These results demonstrate the advantage of the Be-doped LT MQW over the undoped MQW and Be-doped bulk GaAs, with respect to the fast response time as well as the large optical nonlinearity (small F_{sat}).

The Be-doped GaAs/AlAs MQW and bulk-GaAs epilayers were grown by standard MBE on semi-insulating (001) GaAs substrates. The substrate was cleaned by high-temperature treatment without As flux in a preparation chamber.[9] The As_4/Ga beam equivalent pressure ratio was 5–15, and the growth rate was 1.8 μm/h for GaAs and 1.4 μ/h for AlAs. The substrate temperature was monitored by an infrared thermometer and the input electric power into the substrate heater, and the temperature was controlled by using a thermocouple placed behind the substrate holder. We grew first a GaAs buffer layer (200 nm) and then an $Al_{0.3}Ga_{0.7}As$ etch-stop layer (200 nm) at 560 °C. For the MQW samples, they were followed by a MQW layer consisting of 100 periods of 7 nm GaAs wells and 7 nm AlAs barriers. During this MQW growth, the substrate temperature (T_g, hereafter) was kept constant between 700 and 280 °C, and Be was uniformly doped. For the bulk-GaAs epilayer samples, GaAs with a thickness of 700 nm, which was equal to the total thickness of the GaAs layers in the MQW, was grown instead of the MQW. Finally, a 100 nm $Al_{0.3}Ga_{0.7}As$ window layer was grown at T_g. No post-growth annealing was carried out. For optical transmission measurements, the substrate was removed by wet-chemical etching up to the etch-stop layer. After that, the surface of this layer was made somewhat rough by etching, in order to avoid the Fabry–Pérot effect. All measurements were done at room temperature.

Figure 1 shows the absorption spectra of Be-doped ($7.8\times10^{17}\,cm^{-3}$) MQWs grown at $T_g=600\,°C$ (a), 360 °C (b), and 310 °C (c). Also shown is a spectrum of a Be-doped ($2\times10^{19}\,cm^{-3}$) MQW grown at $T_g=280\,°C$ in (d). The ordinate is absorption coefficient α per unit well thickness measured at low excitation intensity, and the curves are shifted vertically for clarity. In undoped MQWs, heavy-hole and light-hole exciton peaks were clearly observed (Fig. 1 in Ref. 4). Though their peaks are broadened and merged in these Be-doped MQWs, a single peak is still recognized. This shows that the excitonic feature is preserved in Be-doped MQWs.

The temporal responses of the optical nonlinearity were measured by using a degenerate pump–probe technique with

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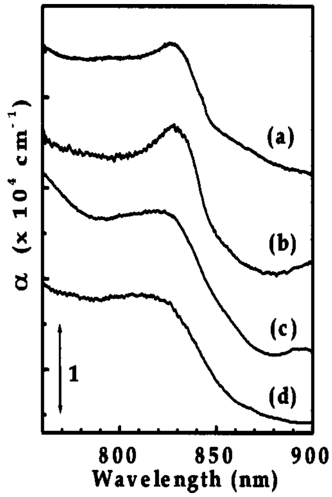

FIG. 1. Absorption spectra of Be-doped ($7.8\times10^{17}\,\mathrm{cm}^{-3}$) GaAs/AlAs MQWs grown at (a) 600, (b) 360, and (c) 310 °C. Also, shown a spectrum of Be-doped ($2\times10^{19}\,\mathrm{cm}^{-3}$) MQW grown at 280 °C in (d).

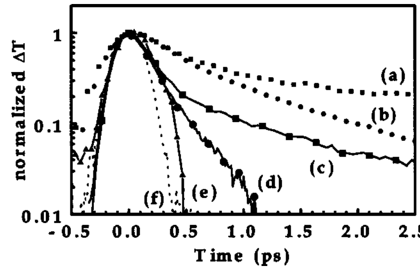

FIG. 2. Normalized temporal changes of differential transmission. The samples of curves (a)–(e) are shown in Table I. Curve (f) shows the system repsonse.

200 fs laser pulses from a mode-locked titanium sapphire laser. The pump beam energy density was 0.01 μJ/cm^2, and the probe density was ~1/5 of the pump.[10] Figure 2 shows differential transmission ΔT normalized to the peak at zero delay for undoped MQWs [(a) and (b)], Be-doped MQWs [(c) and (d)], and Be-doped bulk GaAs [(e)] as a function of time delay t between the pump and the probe pulses (see Table I). The excitation wavelength was set at around the excitonic absorption peak for MQWs and at 860 nm for bulk GaAs. We define the response time τ by the 1/e decay time. The response time τ in undoped MQW are 0.9 and 0.7 ps for $T_g=310\,°\mathrm{C}$ (a) and 280 °C (b), respectively. In these undoped LT MQWs, the decay dynamics is determined by the trapping time of electrons to antisite $\mathrm{As}_{\mathrm{Ga}}^{+}$ defects and the following recombination time between the electrons and holes.[11] By Be doping with the concentration $[\mathrm{Be}]=7.8\times10^{17}\,\mathrm{cm}^{-3}$, the response time is reduced to 0.3 ps in the 310 °C-grown MQW (c). The fastest decay with $\tau=0.25$ ps is obtained in the MQW grown at 280 °C with $[\mathrm{Be}]=2\times10^{19}\,\mathrm{cm}^{-3}$ (d). For 310 °C-grown bulk GaAs, τ was reduced from 2 ps (not shown) to 0.3 ps by Be doping with $[\mathrm{Be}]=7.8\times10^{17}\,\mathrm{cm}^{-3}$ [(e)]. In this Be-doped bulk GaAs, the value of the normalized ΔT goes to -0.17 at $t>1$ ps (Table I), which means the presence of induced absorption.[11]

In LT bulk GaAs, Be doping is reported to decrease neutral antisite $\mathrm{As}_{\mathrm{Ga}}^{0}$ and to increase ionized $\mathrm{As}_{\mathrm{Ga}}^{+}$.[5] The reduction in τ shown in Fig. 2 can be ascribed to increased electron traps of antisite $\mathrm{As}_{\mathrm{Ga}}^{+}$. In undoped LT GaAs, it was reported that electrons and holes are trapped rapidly by point defects located in spatially separated sites, which induces the following slow recombination.[11] This slow component sometimes accompanies the induced absorption ($\Delta T<0$),[11] which was also observed in the present Be-doped bulk GaAs to a significant extent. As shown in Fig. 2 and Table 1, the slow-decay component or normalized ΔT at $t\gg1$ ps is smaller in the Be-doped LT MQWs [(c) and (d)]. This is desirable for high-speed and high-repetition-rate optical switching devices. In LT InGaAs/InAlAs MQWs, the shortening of the lifetime by Be doping was ascribed to the formation of complexes composed of Be acceptors and antisite $\mathrm{As}_{\mathrm{Ga}}^{+}$ donors, which act as the recombination centers trapping both holes and electrons in the same real space.[7] It is probable that the same complexes are formed also in the present Be-doped GaAs/AlAs MQWs, and thus the slow-decay component is suppressed.

In order to obtain optical absorption nonlinearity in Be-doped MQW and bulk GaAs, the excitation-density dependence of α [apparent α obtained from $\alpha=-(\ln T)/d$, where T is the transmission and d is the thickness] was measured. As an excitation source, we used the 200 fs pulses. The wavelength was set around the excitonic absorption peak for MQWs and at 860 nm for bulk GaAs. A low-duty-cycle (1:3000) acousto-optic modulator was used to eliminate heating effects. Incident and transmitted laser powers were measured by a calibrated Si photodetector. The size of the excitation spot on the sample was 70 μm in diameter. The excitation density F was changed from 0.01 to 100 μJ/cm^2. The maximum F was limited by sample heating.

Figure 3 shows the F dependence of α measured for Be-doped ($7.8\times10^{17}\,\mathrm{cm}^{-3}$) MQWs and bulk GaAs grown at 600 (triangles), 360 (circles), and 310 °C (squares). In the MQWs (closed symbols), α begins to decrease at around 1 μJ/cm^2, while the reduction of α in the bulk GaAs (open symbols) takes place at around 10 μJ/cm^2. In addition, the magnitude of the reduction of α at the maximum F in the experimental range is much larger for MQWs than for bulk GaAs. These results indicate the larger optical nonlinearity of MQWs as compared with bulk GaAs. The data of each curve are fitted to the traveling-wave rate-equation model for a

TABLE I. Samples used in Fig. 2. The last column denotes the value of normalized ΔT when the signal becomes almost constant ($t\sim10$ ps).

Symbol	Doping	T_g (°C)	Bulk/MQW	τ (ps)	Normalized ΔT at $t\gg1$ ps
(a)	undoped	310	MQW	0.9	0.07
(b)	undoped	280	MQW	0.7	−0.06
(c)	Be-doped ($7.8\times10^{17}\,\mathrm{cm}^{-3}$)	310	MQW	0.3	0.008
(d)	Be-doped ($2\times10^{19}\,\mathrm{cm}^{-3}$)	280	MQW	0.25	−0.015
(e)	Be-doped ($7.8\times10^{17}\,\mathrm{cm}^{-3}$)	310	Bulk GaAs	0.3	−0.17

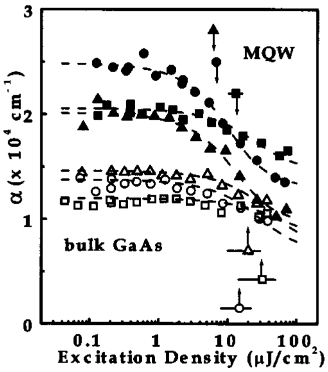

FIG. 3. Excitation density dependence of the apparent optical absorption coefficient α in Be-doped ($7.8\times10^{17}\,\mathrm{cm}^{-3}$) bulk GaAs (open symbols) and GaAs/AlAs MQWs (closed symbols) grown at 600 (△▲), 360 (○●), and 310 °C (□■). Dashed curves are fits to the data. Arrows show the saturation density F_{sat} for each sample, and error ranges are indicated by bars.

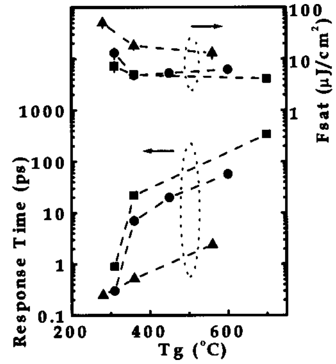

FIG. 4. Response time τ and saturation density F_{sat} of GaAs/AlAs MQWs plotted against growth temperature T_g: undoped (■), $[\mathrm{Be}]=7.8\times10^{17}\,\mathrm{cm}^{-3}$ (●), and $[\mathrm{Be}]=2\times10^{19}\,\mathrm{cm}^{-3}$ (▲).

two-level absorber.[12,13] We can determine F_{sat} and $r=\alpha_{\mathrm{ns}}/\alpha_{\mathrm{lin}}$ (relative nonsaturable losses, where $\alpha_{\mathrm{lin}}=-(\ln T)/d$ at $F\rightarrow0$: linear absorption, and $\alpha_{\mathrm{ns}}=-(\ln T)/d$ at $F\rightarrow\infty$: nonsaturable absorption). The small values of F_{sat} and r are preferable for large optical modulation. For the MQWs, r is in the range of 0.4–0.7. Nearly the same values were obtained for MQWs with higher doping $[\mathrm{Be}]=2\times10^{19}\,\mathrm{cm}^{-3}$. In Be-doped LT bulk GaAs, r is reported to be decreased by Be doping in the reflection measurements.[5,14] However, we did not observe such a decrease for Be-doped bulk GaAs in the present absorption measurements, presumably because of the limitation of maximum F. We observed only a small reduction in α even at the maximum F for all Be-doped bulk GaAs samples, as is shown in Fig. 3. Here, we assumed the same value of r for bulk GaAs as that for the MQW grown at the same temperature, in order to compare F_{sat} between bulk GaAs and the MQW. The possible variation of r may lead to some uncertainty in F_{sat}.

The saturation density F_{sat} is shown by the arrows in Fig. 3. Although the error range of F_{sat} for bulk GaAs is rather large, we can find a smaller F_{sat} in the Be-doped MQWs than that of the Be-doped bulk GaAs grown at the same temperature. This result shows the larger optical nonlinearity of Be-doped MQWs than that of Be-doped bulk GaAs. This was also confirmed in the pump–probe experiments where $\Delta T/T$ was ~5 times larger for Be-doped MQWs than for Be-doped bulk GaAs. The saturation density F_{sat} was found to be minimum at around the excitonic peak wavelength even in Be-doped MQWs, as is the case in the undoped MQW.[4,15] This result means that the enhancement of the optical nonlinearity is due to the excitonic effect.

The response time τ of the MQW with different [Be] is plotted against the growth temperature T_g in the lower part of Fig. 4. In all the samples, τ is decreased as T_g is decreased. In the whole region of T_g, τ is decreased as [Be] is increased. Decrease of τ is especially prominent at high T_g. In the upper part of Fig. 4, F_{sat} of these MQWs is plotted. As T_g is decreased, F_{sat} is increased. This may be due to the degradation of the MQW quality coming from the LT growth. It is found that Be doping at $[\mathrm{Be}]=7.8\times10^{17}\,\mathrm{cm}^{-3}$ induces almost no increase in F_{sat}, and higher doping at $[\mathrm{Be}]=2\times10^{19}\,\mathrm{cm}^{-3}$ increases F_{sat} by a factor of 3–4. This factor of the increase in F_{sat} is much smaller than that of the decrease in τ (except $T_g=280\,°\mathrm{C}$), as is shown in Fig. 4. It is concluded that τ of the Be-doped MQW is drastically reduced while the magnitude of the optical nonlinearity is not reduced so much. While the 280 °C-grown Be-doped ($2\times10^{19}\,\mathrm{cm}^{-3}$) MQW shows a 0.25 ps response time with large F_{sat} ($50\,\mu\mathrm{J/cm}^2$), the 310 °C-grown Be-doped ($7.8\times10^{17}\,\mathrm{cm}^{-3}$) MQW has τ of 0.3 ps with moderate F_{sat} ($13\,\mu\mathrm{J/cm}^2$). We should carefully choose T_g and [Be] for obtaining the desired time response and the magnitude of the nonlinearity.

In summary, Be doping shortened the response time of the LT GaAs/AlAs MQW down to $\tau=0.25$ ps, and the slow-decay component often observed in undoped LT bulk GaAs was suppressed. The saturation density of the Be-doped MQW was smaller than that of Be-doped bulk GaAs, regardless of the growth temperature. These results demonstrate the superior properties of the Be-doped LT MQW for applications in ultrafast nonlinear optics having a fast time response and a large optical nonlinearity.

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