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Large optical nonlinearity and fast response time in low-temperature grown GaAs/AlAs multiple quantum wells

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We have investigated optical nonlinearity in low-temperature (LT) molecular-beam-epitaxy-grown GaAs/AlAs multiple quantum wells (MQWs). Minimum saturation intensity, that is, maximum optical nonlinearity, is observed at around the excitonic absorption peak. The saturation intensity of the LT MQW is smaller by an order of magnitude than that of LT bulk GaAs. The response time of the LT MQW is 1/4–1/2 of the LT GaAs, and becomes less than 1 ps, corresponding to ~1/400 of the standard-temperature-grown MQW. These results demonstrate a clear advantage of the room-temperature excitons in the LT MQW having large optical nonlinearity as well as fast response time. © *2000 American Institute of Physics.* [S0003-6951(00)03627-5]

Low-temperature (LT) molecular-beam-epitaxy (MBE) grown III–V semiconductors have found many applications in ultrafast all-optical switching,[1–3] femto- and picosecond laser pulse generation,[4] and high-bit-rate optical communications systems.[5,6] Ultrafast all-optical switching based on the absorption nonlinearity requires materials with two properties, i.e., a fast temporal response of the nonlinear absorption and large optical nonlinearity. From this purpose LT GaAs has been extensively investigated, and beryllium doping and annealing, for example, are reported very recently to improve their properties.[7,8] A LT multiple quantum well (MQW) can be considered as another candidate. It is known that the standard-temperature (ST)-grown GaAs/Al(Ga)As MQW shows optical saturation intensity much smaller than that of the bulk GaAs, which means larger optical nonlinearity of the room-temperature excitons in the MQW.[9] However, LT MBE growth usually brings about high crystal-defect densities, which broadens the excitonic absorption peak. Due to this difficulty, only a few papers have been published so far. They reported on LT GaAs/AlAs MQWs exhibiting 15 ps carrier lifetime,[10] LT InGaAs/InAlAs MQWs demonstrating 1.5 ps time response,[6] LT InGaAs/InAlAs MQWs having 27 ps–sub-ps responses,[11] and LT InGaAs/GaAs MQWs having picosecond responses.[12] Nevertheless, there has been no paper describing both the optical nonlinearity and the temporal response except Ref. 12, or describing a comparison between the MQW and the bulk film. In addition, the wavelength dependence of the nonlinearity showing excitonic enhancement was not described in the literature, although the excitonic property in the LT MQW is very important, as it is in the ST MQW. In this letter, by MBE we grow a LT GaAs/AlAs MQW which shows clear quantum-confined excitons at room temperature, and study both the optical nonlinearity and the picosecond dynamics in comparison with a ST MQW and LT bulk GaAs.

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The GaAs/AlAs MQW and bulk GaAs epilayers were grown by standard MBE on semi-insulating (001) GaAs substrates. The substrates were cleaned by high-temperature treatment without As flux in a preparation chamber.[13] 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 μm/h for AlAs. The substrate temperature, which is the most important parameter in LT MBE, was monitored and controlled by using a thermocouple placed on the substrate holder, a radiation thermometer, and the input electric power into the substrate heater. We grew first a GaAs buffer layer and then an $Al_{0.3}Ga_{0.7}As$ etch-stop layer at 560 °C. In 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 the MQW growth, the substrate temperature was 700 °C in ST growth and 360 or 310 °C in the LT growth. For the bulk GaAs epilayer samples, 700 nm GaAs was grown instead of the MQW. A 100 nm $Al_{0.3}Ga_{0.7}As$ window layer was grown at 450 °C. All of the epilayers were not intentionally doped. The epilayer was, then, *in situ* annealed at 450 °C for 30 min under As pressure. An effort was made to keep the growth condition constant during the course of this study by characterizing all of the grown epilayers with x-ray diffraction, standard photoluminescence, and optical absorption measurement. Transmission electron microscope measurements showed flat heterointerfaces even in the LT MQW. For optical transmission measurements, the substrate was removed by wet-chemical etching up to the etch-stop layer. All measurements were done at room temperature.

In Fig. 1, solid lines show spectra of optical absorption α per unit well thickness (measured at low excitation intensity) of (a) 700, (b) 360, and (c) 310 °C-grown MQWs. In the 700 °C-grown MQW, sharp and well-separated quantum-confined exciton peaks [heavy-hole (HH) and light-hole (LH) excitons] are observed. This is true also for the 360 °C-grown MQW. For the 310 °C-grown MQW, the excitonic peaks become obscure, but still are recognized.

For these samples, we measured optical absorption nonlinearity. As an excitation source, 2 ps pulses from a mode-

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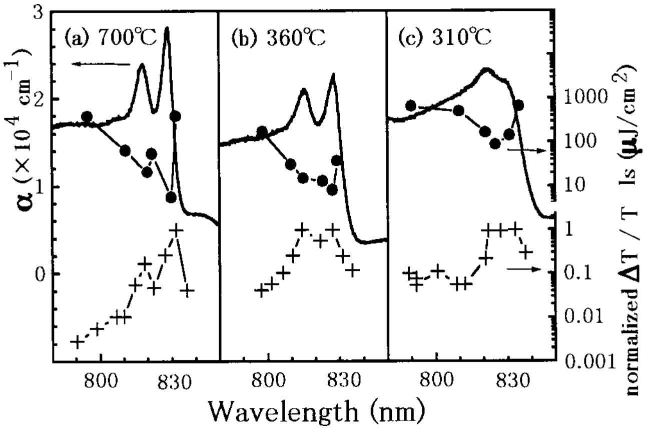

FIG. 1. Absorption spectra (α, solid lines), saturation densities (I_s, circles), and the magnitude of the differential transmission at time zero in the pump–probe measurements (normalized $\Delta T/T$, crosses) in (a) 700 °C, (b) 360 °C, and (c) 310 °C-grown MQWs.

locked titanium–sapphire laser were used. Incident and transmitted laser powers were measured by a calibrated Si photodetector. The size of the excitation spot on the sample was set to 200 μm in diameter. The excitation density I was changed from 0.01 to 100 μJ/cm^2. The maximum I was limited by sample damage. The spectral width of the 2 ps laser pulse is 0.3 nm, which is smaller by a factor of 10 than the exciton linewidth. The pulse repetition rate was set to 80 kHz, because we found that the thermal effect changes the apparent absorption coefficient above 1 MHz at the maximum I.

Figure 2(a) shows I dependence of α measured at various wavelengths λ for the 700 °C-grown MQW. As shown in Fig. 1(a), λ=830 nm and λ=820 nm are HH- and LH-exciton peak wavelengths and are shown by the solid circles and solid squares, respectively, in Fig. 2(a). The wavelength λ=795 nm is located deep in the band, and λ=832 nm is at the long-wavelength-side shoulder of the HH-exciton peak. Among various λ, α begins to be reduced at the smallest I at the HH-exciton peak (circles). We empirically fit these circles by an expression $\alpha=\alpha_1/(1+I/I_s)+\alpha_2$, where α_1 and α_2 are fitting parameters, and I_s is the saturation density.[9,14,15] The least-square fitting gives $1.60/[1+(I/I_s)]+1.32(\times 10^4\ \mathrm{cm}^{-1})$, where $I_s=5.6\pm0.5$ μJ/cm^2, and is shown by the dashed line in Fig. 2(a).

In order to obtain the λ dependence of I_s, we fitted all data obtained with various λ by the same expression. The results are indicated by the dashed lines in Fig. 2(a), and the obtained I_s for various λ is plotted in Fig. 1(a) by solid circles. It is shown that I_s becomes small at the HH- and LH-exciton peaks, and I_s at the HH-exciton peak is smaller than that at the LH-exciton peak.[16] This shows large optical nonlinearity at the exciton peaks.[9,14]

Figure 2(b) shows the I dependence of α at typical λ for the 360 °C-grown MQW. Also for this LT MQW, reduction of α takes place at the smallest I an λ=827 nm (HH-exciton peak) shown by solid circles. The data were fitted as previously, and the result was $1.09/[1+(I/I_s)]+1.00$ $(\times 10^4\ \mathrm{cm}^{-1})$, where $I_s=8.0\pm0.8$ μJ/cm^2.[17] The saturation density I_s is larger by 43% as compared with the 700 °C-grown MQW. Other data at different λ were fitted, and the obtained λ dependence of I_s is plotted in Fig. 1(b) by the solid circles. As is shown, I_s at the LH exciton is comparable to that at the HH exciton.

The dependence of α on I for the 310 °C-grown MQW is shown in Fig. 2(c). In this sample, reduction of α occurs at the smallest I at λ=825 nm shown by the solid circles, which is located at the center of the obscured exciton peak. The least-square fitting for these circles gives $1.06/[1+(I/I_s)]+1.05(\times 10^4\ \mathrm{cm}^{-1})$, where $I_s=90\pm30$ μJ/cm^2.[8,18] The saturation density I_s is larger by more than 10 than those in the previous two samples. In Fig. 1(c), the λ dependence of I_s is shown by the solid circles. As shown in Figs. 1(b) and 1(c) for the LT MQW, I_s becomes minimum at around the HH- and LH-exciton peaks, even though the exciton peak structures are broadened by LT growth. This fact shows that the optical nonlinearity becomes largest at around the excitonic absorption peak even in the LT MQW.

In order to compare the magnitude of the nonlinearity between the bulk GaAs and the MQW, we tried to measure I_s of the bulk GaAs between λ=870 nm (the band-gap wavelength) and 850 nm. However, reduction in α, such as that shown in Fig. 2, was not observed in ST or in LT bulk GaAs in the present experimental condition ($I<100$ μJ/cm^2).[9,19] This means I_s of the MQW is smaller by at least an order of magnitude than that of the bulk GaAs, which demonstrates the enhancement of the optical nonlinearity not only in the ST MQW (Ref. 9) but also in the LT MQW.

Temporal responses of the MQW and the bulk GaAs were measured using a degenerate pump–probe technique with 2 ps laser pulses. The pump beam energy density was 0.01 μJ/cm^2, and the probe density was ~1/5 of the pump. The wavelength λ was set to the HH-exciton peak for the MQW and to 865 nm in the bulk GaAs. Figure 3 shows differential transmission $\Delta T/T$ in the normalized scale for the MQW and the bulk GaAs as a function of the time delay between the pump and the probe beams. We define the response time τ by the 1/e decay time. In the ST-grown

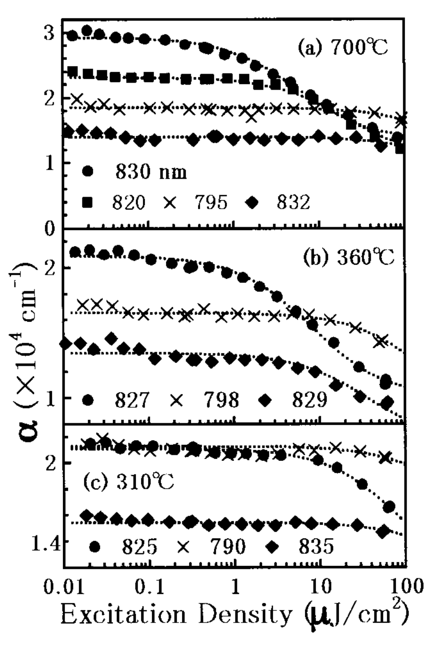

FIG. 2. Excitation density dependence of the optical absorption coefficient in (a) 700 °C, (b) 360 °C, and (c) 310 °C-grown MQWs measured at various wavelengths. Dashed curves are semiempirical saturation fits to the data.

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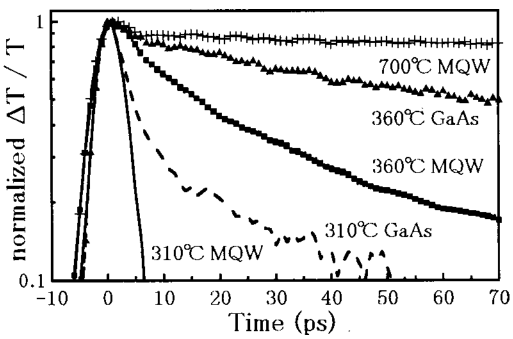

FIG. 3. Normalized temporal changes of differential transmission. The profile for the 310 °C-grown MQW shows the time resolution in this experiment.

samples, τ=340 ps was obtained both for the MQW and the bulk GaAs. However, τ of the 360 °C-grown MQW is decreased to 27 ps, whereas that of the 360 °C-grown bulk GaAs is 100 ps. For the 310 °C-grown MQW, τ is faster than the time resolution. When we used 200 fs pulses for the excitation sources in place of the 2 ps pulses, τ was obtained to be 0.9 ps for this MQW. This is shorter than that of the 310 °C-grown bulk GaAs (τ=2 ps for the 200 fs excitation). These results show that the LT MQW exhibits a faster response as compared with the bulk GaAs grown at the same temperature (τ of the LT MQW is 1/4–1/2 of the bulk GaAs).

It is reported that positively ionized antisite As acting as electron traps is the origin of the fast response time of LT bulk GaAs.[7,8] It is also reported that in the LT GaAs/Al(Ga)As MQW, the antisite As diffuses from the Al(Ga)As barrier layers to the GaAs well layers and As precipitates accumulate in the GaAs layers.[20–22] Therefore, we think that in the LT MQW, the increased density of excess As in the GaAs layers or at the heterointerfaces leads to reduction in τ.

The crosses in Fig. 1 show the λ dependence of the magnitude of the differential transmission $\Delta T/T$ in the pump–probe measurements at the time zero. This value obtained at $I=0.01$ μJ/cm^2 can be used to indicate a measure of the optical nonlinearity at various λ. As shown, the maximum in the λ dependence of $\Delta T/T$ coincides with the minimum in the λ dependence of I_s in all of the three MQW samples. This result again confirms the larger optical nonlinearity around the excitonic peaks in the MQW.

We discuss the measured values of I_s. It was reported that for the standard GaAs/$Al_{0.3}Ga_{0.7}$As MQW, the obtained value of I_s corresponds to the effective exciton density of 3.3×10^{17} cm^{-3} in the GaAs layer.[9] The likely mechanism for the optical absorption nonlinearity was thought to be the filling of the excitons in the phase space density of states.[23] For the 700 °C-grown MQW in the present experiment, the obtained value, I_s=5.6 μJ/cm^2, corresponds to 2.7 $\times10^{17}$ cm^{-3}, which agrees well with the above reported value. For the 310 °C-grown MQW, τ is 0.9 ps, which is smaller roughly by a factor of 2 than the excitation pulse width (2 ps). Thus, the obtained I_s (90 μJ/cm^2) should be regarded as 40 μJ/cm^2, in order to make a fair comparison with the other samples having τ larger than 2 ps. Then, we find that I_s becomes larger (from 5.6 and 8.0 to 40 μJ/cm^2) when the growth temperature decreases (from 700 and 360 to 310 °C). This increase in I_s means the reduction of the optical nonlinearity. It might be due to the degradation of MQW quality coming from the LT growth. However, this increase of I_s by a factor of ~7 is much smaller than the reduction of τ by a factor of ~400 (from 340 and 27 to 0.9 ps). We can say that the LT MQW drastically reduces the response time while it does not reduce the magnitude of the optical nonlinearity as much.

In summary, the LT MQW shows a large optical nonlinearity (small saturation intensity and large differential transmission) at around the excitonic peak at room temperature. The saturation intensity of the MQW is almost the same, within a factor of 7, regardless of the growth temperature. The response time of the LT MQW is ~1/400 of the ST MQW. In addition, the saturation intensity is smaller and the response time is shorter for the LT MQW as compared to the LT bulk GaAs. These results demonstrate the advantage of the LT MQW having large optical nonlinearity as well as fast response time.

[1] S. Gupta, J. F. Whitaker, and G. A. Mourou, IEEE J. Quantum Electron. **28**, 2464 (1992).
[2] E. S. Harmon, M. R. Melloch, J. M. Woodall, D. D. Nolte, N. Otsuka, and C. L. Chang, Appl. Phys. Lett. **63**, 2248 (1993).
[3] H. S. Loka, S. D. Benjamin, and P. W. E. Smith, IEEE J. Quantum Electron. **34**, 1426 (1998).
[4] U. Keller, K. J. Weingarten, F. X. Kartner, D. Kopf, B. Braun, I. D. Jung, R. Fluck, C. Honninger, N. Matuschek, and J. Aus der Au, IEEE J. Sel. Top. Quantum Electron. **2**, 435 (1996).
[5] R. Takahashi, Y. Kawamura, T. Kagawa, and H. Iwamura, Appl. Phys. Lett. **65**, 1790 (1994).
[6] R. Takahashi, Y. Kawamura, and H. Iwamura, Appl. Phys. Lett. **68**, 153 (1996).
[7] M. Haiml, U. Siegner, F. Morier-Genoud, U. Keller, M. Luysberg, R. C. Lutz, P. Specht, and E. R. Weber, Appl. Phys. Lett. **74**, 3134 (1999).
[8] M. Haiml, U. Siegner, F. Morier-Genoud, U. Keller, M. Luysberg, P. Specht, and E. R. Weber, Appl. Phys. Lett. **74**, 1269 (1999).
[9] D. A. B. Miller, D. S. Chemla, D. J. Eilenberger, P. W. Smith, A. C. Gossard, and W. T. Tsang, Appl. Phys. Lett. **41**, 679 (1982) .
[10] I. Lahiri, D. D. Nolte, E. S. Harmon, M. R. Melloch, and J. M. Woodall, Appl. Phys. Lett. **66**, 2519 (1995).
[11] P. W. Juodawlkis, D. T. McInturff, and S. E. Ralph, Appl. Phys. Lett. **69**, 4062 (1996).
[12] U. Keller, Appl. Phys. B: Lasers Opt. **58**, 347 (1994).
[13] K. Iizuka, K. Matsumaru, T. Suzuki, H. Hirose, K. Suzuki, and H. Okamoto, J. Cryst. Growth **150**, 13 (1995).
[14] Y. Masumoto, S. Tarucha, and H. Okamoto, J. Phys. Soc. Jpn. **55**, 57 (1986).
[15] B. Fluegel, M. Joffre, S. H. Park, R. Morgan, Y. Z. Hu, M. Lindberg, S. W. Koch, D. Hulin, A. Migus, A. Antonetti, and N. Peyghambarian, J. Cryst. Growth **101**, 634 (1990).
[16] Some data such as λ=795 nm in Fig. 2(a) do not show enough reduction in α even at the maximum I. Thus, we assumed to fix the ratio $r=\alpha_2/(\alpha_1+\alpha_2)=0.45$ in all λ (the error of r was estimated to be 0.02 in the fitting for the λ=830 nm data). We confirmed that I_s is minimum at the exciton peaks regardless of the assumption.
[17] For the 360 °C-grown MQW, $r=0.48\pm0.02$.
[18] For the 310 °C-grown MQW, $r=0.5\pm0.1$. In this fitting, r is assumed not to increase with the decrease of the growth temperature, because one of the origins of the residual loss α_2 is neutral excess As included in LT-grown layers. The condition $r>0.4$ was assumed.
[19] The present largest value of I not reducing α is consistent with the reported I_s for bulk GaAs.
[20] K. Mahalingam, N. Otsuka, M. R. Melloch, and J. M. Woodall, Appl. Phys. Lett. **60**, 3253 (1992).
[21] M. N. Chang, N. T. Yeh, C. M. Lu, K. C. Hsieh, and J.-I. Chyi, Appl. Phys. Lett. **75**, 52 (1999).
[22] B. Lita, S. Ghaisas, R. S. Goldman, and M. R. Melloch, Appl. Phys. Lett. **75**, 4082 (1999).
[23] S. Schmitt-Rink, D. S. Chemla, and D. A. B. Miller, Phys. Rev. B **32**, 6601 (1985).

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