PHYSICAL REVIEW B VOLUME 40, NUMBER 17 15 DECEMBER 1989-I

Dynamical aspects of luminescence from GaAs-AlAs single quantum wells under hydrostatic pressure

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(Received 22 May 1989)

Time-integrated and time-resolved photoluminescence studies of GaAs-AlAs single quantum wells under hydrostatic pressure were performed at 77 K. The Γ-point exciton luminescence quenches when the luminescence peak energy approaches 1.83 eV under hydrostatic pressure. The quenching is ascribed to the interlayer Γ-X crossing. Photoluminescence shows anomalously slow decay ranging from several nanoseconds to a few tens of nanoseconds under low hydrostatic pressure. With the further increase of hydrostatic pressure, the luminescence rise decreases drastically and is less than a few tens of picoseconds around the interlayer Γ-X crossover. The appearance of the slow decay component and the sudden decrease in the rise time are explained by the interlayer Γ-X transfer of electrons.

I. INTRODUCTION

Recently, much effort has been devoted to clarify the electronic structure of the staggered-alignment GaAs-AlAs, GaAs-$Al_xGa_{1-x}As$, and $Al_xGa_{1-x}As$-AlAs quantum-well structures.[1-15] Many distinct optical properties concerning the interlayer Γ-X crossing have been studied by changing the aluminum content,[2-4] the layer thickness,[5-9] application of the hydrostatic pressure,[10-14] or the electric field.[15] Most of the previous studies concern the static optical properties. Some time-resolved measurements of luminescence were reported, but were done in the fairly slow time domain ranging from nanoseconds to microseconds,[5-7,14] where the main interest is focused on the X-point exciton luminescence. In contrast, we obtained the detailed temporal information about the lowest-energy Γ-point exciton luminescence around the interlayer Γ-X crossover. In this work we have used hydrostatic pressure to induce the interlayer Γ-X crossing in GaAs-AlAs single quantum wells. The temporal profile of the Γ-point exciton luminescence was studied by means of picosecond and nanosecond time-resolved spectroscopy. On the basis of the experimental results, we clarify the dynamical relaxation processes of carriers inherent in the interlayer Γ-X mixing.

II. EXPERIMENTAL PROCEDURES

The sample used in this study was grown by metalorganic chemical vapor deposition. The sample consists of five single GaAs well layers separated by wide AlAs barrier layers 48 nm thick. Well thicknesses are 2.9, 4.2, 5.8, 9.3, and 15 nm, respectively. Single quantum wells were grown on a GaAs substrate and were covered with an $Al_{0.5}Ga_{0.5}As$ cap layer 100 nm thick. The GaAs substrate was thinned to 50 μm. A small piece of the thinned sample was placed in a Merrill-Bassett diamond-anvil cell. As the pressure-transmitting fluid, liquid nitrogen was used. The energy shift of the R_1 luminescence line from a ruby crystal was used to calibrate the pressure. The cell was directly immersed in liquid nitrogen. The energy shift of the R_1 line was measured by an 85-cm double monochromator.

One of three following excitation light sources was used depending on the purpose of the experiments. It was either a 514.5-nm (2.409 eV) line of a steady-state argon-ion laser, or 532-nm (2.33 eV) pulses (82 MHz) obtained by the second harmonics of the fiber-compressed output of the mode-locked Nd^{3+}:YAG (YAG denotes yttrium aluminum garnet) laser, or 574-nm (2.16 eV) pulses (4 MHz) generated by a cavity-dumped synchronously pumped dye laser. In all the cases, the GaAs wells are selectively photoexcited because the direct energy gap of AlAs is 3.125 eV at 77 K.[16] For the time-resolved luminescence measurement, a synchroscan streak camera and single-photon counting electronics were used together with a 25-cm monochromator for the picosecond and nanosecond time regimes, respectively. The synchroscan streak camera was operated with the repetition rate of 82 MHz by using 532-nm pulses (5 ps) as the excitation source, while the single-photon counting electronics were operated with the repetition rate of 4 MHz by using 574-nm pulses (1 ps) as the excitation source. The time resolution of the former system was 74 ps, while that of the latter system was 0.85 ns.

III. EXPERIMENTAL RESULTS AND DISCUSSIONS

A. Luminescence under hydrostatic pressure

The luminescence spectrum at 77 under K atomospheric pressure is shown in Fig. 1 together with the

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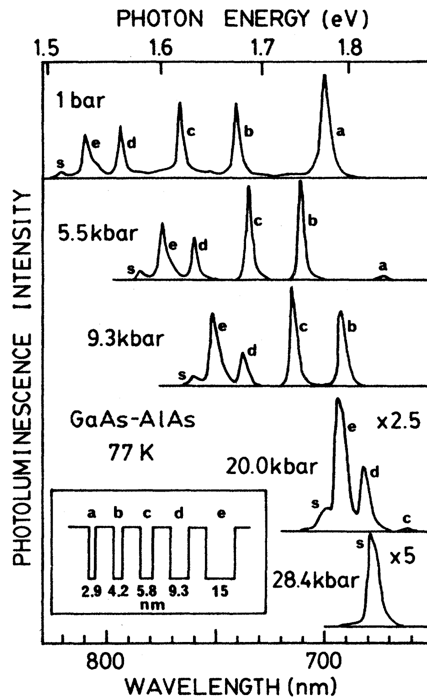

FIG. 1. Luminescence spectra of the 77 K under various hydrostatic pressures. Luminescence peaks labeled by *a*, *b*, *c*, *d*, *e*, and *s* come from the 2.9-, 4.2-, 5.8-, 9.3-, and 15-nm GaAs wells and the GaAs substrate, respectively.

luminescence under various hydrostatic pressures. At atmospheric pressure, the sample showed six distinct luminescence peaks which are ascribed to the Γ excitons associated with the lowest Γ subband in five GaAs single quantum wells and the luminescence from a GaAs substrate. The lowest quantum levels in 2.9-, 4.2-, 5.8-, 9.3-, and 15-nm GaAs quantum wells were calculated by the envelope-function approximation in the finite-well model,[17] and their lowest-energy intersubband transitions were found to be 1.797, 1.685, 1.620, 1.562, and 1.533 eV, respectively.[18] Here, exciton binding energy was not taken into account. The estimated values agree with the observed values, 1.772, 1.675, 1.618, 1.561, and 1.527 eV with the error of 25, 10, 2, 1, and 6 meV, respectively. The agreement is good. In order to check the sample quality, the relative luminescence intensity and the lifetime of excitons were studied between 4.2 and 80 K at atmospheric pressure. The reduction in the luminescence intensity of excitons in all the GaAs wells from 4.2 to 80 K was at most 30%. The lifetime of excitons was found to increase with the increase of the well width and the rise of the temperature. The increase of the lifetime is well explained by the increase of the exciton coherent volume with the increase of the well width and temperature consistently with the result by Feldmann *et al.*[19] Therefore, the measured exciton recombination rate is the radiative rate, and is affected little by nonradiative contributions.

With the application of hydrostatic pressure, the exciton luminescence shifts toward higher energy, as shown in Figs. 1 and 2. There is no significant broadening of luminescence peaks. The energy shift with pressure of the luminescence from the 5.8-, 9.3-, and 15-nm wells were well fitted by straight lines, but those from the 2.9- and 4.2-nm wells were fitted by quadratic curves. The pressure dependence of the quantum-well luminescence energy is similar to that observed by previous authors.[10–13] The exciton luminescence quenches when its energy approaches 1.83 eV. We have measured the integrated intensity of the luminescence peaks with careful correlation of the wavelength dependence of the sensitivity of the experimental system by using a standard light source. In Fig. 2, the hatched part shows the region where the luminescence intensity changes from 50% to less than 0.1% of that in atmospheric pressure. The luminescence quench is ascribed to the interlayer Γ-*X* crossing, as reported by previous authors.[10–14] Wolford *et al.* reported the reduction in luminescence intensity to be 1% just after the interlayer Γ-*X* crossover.[10] Venkateswaran *et al.* reported a reduction in intensity of 10%.[13] The hatched region shows this range with some allowance. The pressure coefficient of the Γ-*X* transition in GaAs-AlAs is reported to be −2.4 meV/kbar.[20] Using this value, the *X*-state energy in AlAs is drawn so as to pass through all of the hatched regions. The procedure yields the band offset ratio of 0.77:0.23. The Γ-*X* interlayer crossing occurs at the cross points. The pressures at the crosses denoted by P_c are 5.7±1, 14.1±1, 18.1±1,

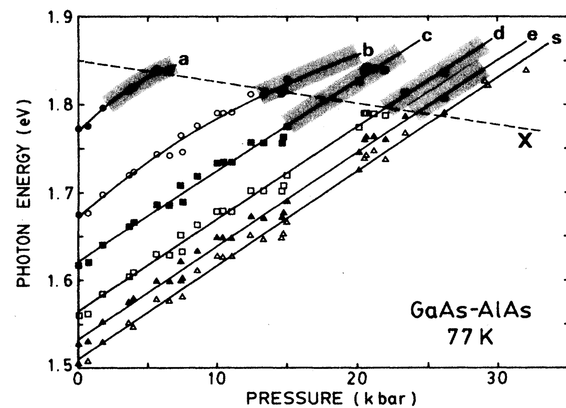

FIG. 2. Luminescence peak energies as a function of hydrostatic pressure at 77 K. Solid circles, open circles, solid squares, open squares, solid triangles, and open triangles show luminescence from the 2.9-nm GaAs well, the 4.2-nm GaAs well, the 5.8-nm GaAs well, the 9.3-nm GaAs well, the 15-nm GaAs well, and the GaAs substrate, respectively. Experimental points are fitted by six lines, *a*, *b*, *c*, *d*, *e*, and *s*, which are represented by $1.77+1.590\times10^{-2}P-7.4\times10^{-4}P^2$ eV, $1.67+1.358\times10^{-2}P-2.3\times10^{-4}P^2$ eV, $1.62+1.028\times10^{-2}P$ eV, $1.56+1.051\times10^{-2}P$ eV, $1.53+1.065\times10^{-2}P$ eV and $1.51+1.071\times10^{-2}P$ eV, respectively. Here P is the hydrostatic pressure in units of kbar. The hatched region corresponds to the luminescence intensity changes from 50% to less than 0.1% of that in atmospheric pressure. The dashed line shows pressure dependence of the *X*-point energy in AlAs.

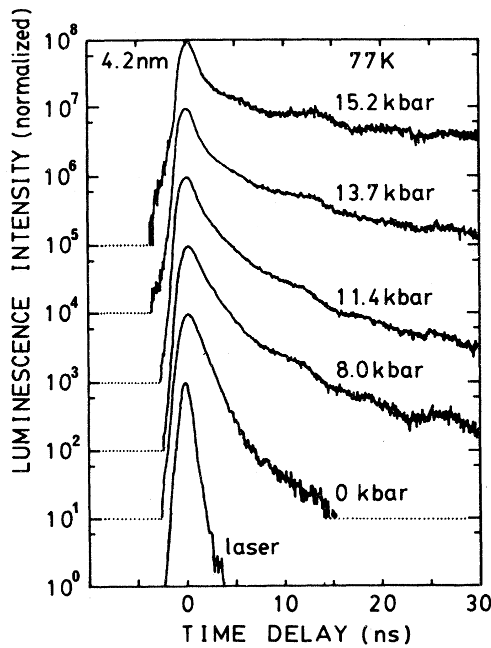

FIG. 3. Normalized temporal profile of the lowest-energy Γ-point exciton luminescence from the 4.2-nm GaAs well under various hydrostatic pressures at 77 K. Data are shifted in turn by a decade. The data denoted by "laser" show the temporal resolution of the experimental system.

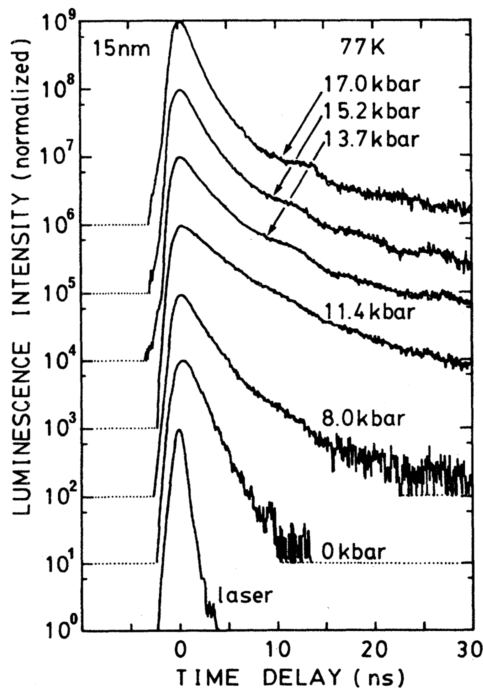

FIG. 4. Normalized temporal profile of the lowest-energy Γ-point exciton luminescence from the 15-nm GaAs well under various hydrostatic pressures at 77 K. Data are shifted in turn by a decade. The data denoted by "laser" show the temporal resolution of the experimental system.

22.5±1, and 24.5±1 kbar for the 2.9-, 4.2-, 5.8-, 9.3-, and 15-nm wells, respectively.

B. Time-resolved study of luminescence under hydrostatic pressure

Time-resolved study of exciton luminescence was done in picosecond and nanosecond time domains. The temporal response was investigated in the nanosecond time domain by using picosecond light pulses and time-correlated single-photon counting. Figure 3 shows the results for the 4.2-nm GaAs quantum well. With the application of 8–15 kbar, a slow decay component ranging between several nanoseconds and a few tens of nanoseconds in decay time appears. For the 4.2-nm well, P_c is almost equal to 14.1±1 kbar. As the pressure increases and approaches P_c, the decay time of the slow decay component increases. Figure 4 shows the results for the 15-nm GaAs quantum well. In this case, the slow decay component is observed again. The slow decay component contributes most at $P=11.4$ kbar. This pressure corresponds to an energy separation between the lowest Γ quantum state in GaAs and the X state in AlAs of 170

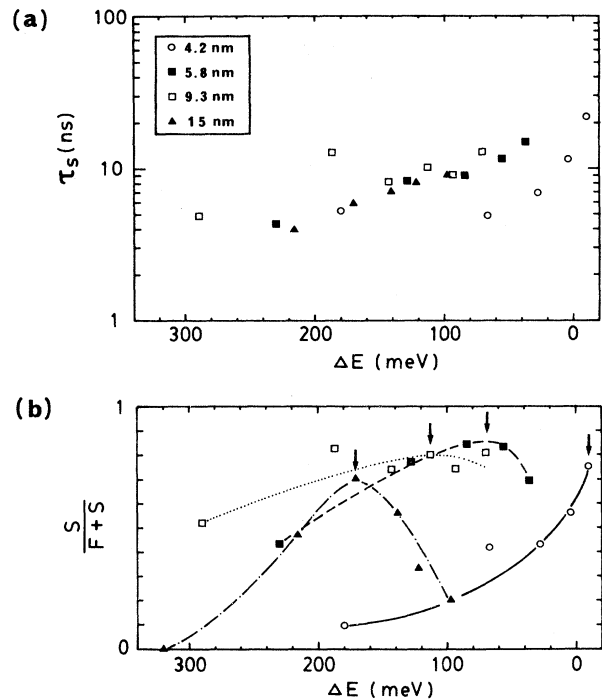

FIG. 5. (a) Decay time of the slow decay component of the exciton luminescence as a function of the energy separation ΔE from the lowest Γ subband energy in GaAs to the X-state energy in AlAs. Data of 4.2-, 5.8-, 9.3- and 15-nm wells are plotted by open circles, solid squares, open squares, solid triangles, and open triangles, respectively. (b) The relative contribution of the slow decay component, $S/(F+S)$, as a function of ΔE. Here, S and F denote the time-integrated luminescence intensity of the slow $(3.9<\tau_s<24$ ns) and fast $(1.2<\tau_f<1.5$ ns) decay components, respectively. Lines are guides for the eye. The arrows indicate apparent maxima.

meV. With the further increase of the pressure, the slow decay component becomes gradually smaller between 11 and 17 kbar. Similar characteristics were observed for the 5.8- and 9.3-nm GaAs quantum wells. We fitted the data by two exponential decays, $a_f \exp(-t/\tau_f) + a_s \exp(-t/\tau_s)$, where $a_{f(s)}$ is the amplitude of the fast (slow) decay component and $\tau_{f(s)}$ is the decay time constant of the fast (slow) component. Here, the fast decay time constant τ_f was found to range from 1.2 to 1.5 ns and the slow decay time constant from 3.9 to 24 ns. Figure 5 shows the slow decay time constant, τ_s, and the relative contribution of the slow decay component, $S/(F+S) = a_s\tau_s/(a_f\tau_f + a_s\tau_s)$, as a function of the energy separation ΔE between the lowest Γ subband in GaAs and the X state in AlAs. As the energy of the lowest Γ quantum state in GaAs well increases and approaches the X state in AlAs, the time constant of the slow decay component increases, but its relative contribution is at its maximum at the positions indicated by arrows which define ΔE_m. With the decrease of well width, ΔE_m increases and approaches zero.

The growth of the exciton luminescence shows a more drastic change. The rise part of the exciton luminescence was studied in the picosecond time domain by using the picosecond light pulses and the streak camera. Figure 6 shows the rise of the exciton luminescence for all wells around the interlayer Γ-X crossover. The figure clearly shows that the rise time drastically decreases when pressure increases and approaches P_c. We approximated the temporal change of the exciton luminescence at the curve $[1-\exp(-t/\tau_r)]\exp(-t/\tau_d) \otimes \mathrm{sech}^2[t(\text{in ps})/42]$ by taking account of the time resolution of the present experimental system, where $\otimes$ denotes the convolution and $\tau_{r(d)}$ is rise (decay) time constant. The deconvolution procedure was necessary, if the rise time is smaller than 100 ps. In Fig. 7, the rise time τ_r of the exciton luminescence is plotted as a function of ΔE. Figure 7 indicates that the sudden decrease in the rise time constant takes place when $-\Delta E$ decreases and becomes less than 20 meV. Around the Γ-X crossover, the rise time is reduced to be less than a few tens of picoseconds.

To explain the time-resolved experiments, we present a model shown schematically in Fig. 8. All of the laser sources used in our experiments excite Γ electrons in the GaAs wells selectively because the direct gap of AlAs barriers is 3.125 eV at 77 K which is larger than the photon energy of the laser sources. Some excited Γ electrons in GaAs wells will be scattered to the X valley in GaAs wells. Some of them will be scattered to the Γ valley in GaAs wells again or to the X valleys in AlAs barriers via the interlayer transfer. Then, the X electrons in AlAs barriers will be energetically relaxed to the bottom of the X valley in AlAs. If the X valley in AlAs is higher than the lowest Γ quantum states in GaAs, the X electrons in AlAs will be scattered to the Γ quantum states in GaAs. The interlayer process will contribute to slowing the rise of the lowest exciton luminescence. Conversely, if the X valley in AlAs is lower than the lowest Γ quantum states in GaAs, the interlayer transfer from the X state in AlAs to the Γ state in GaAs does not work, so that the rise of

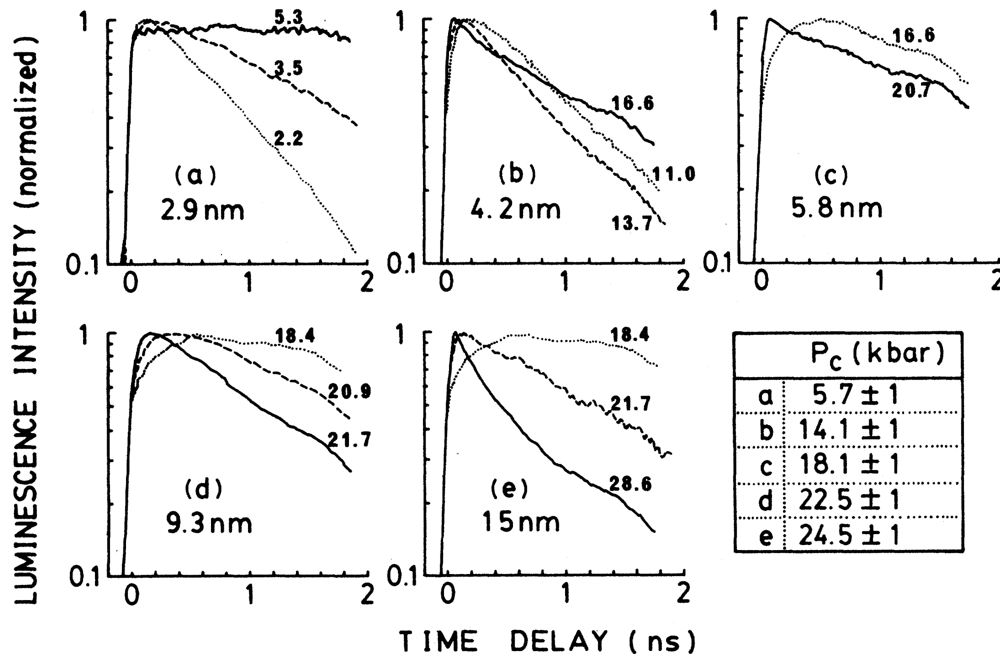

FIG. 6. Normalized temporal profiles of the lowest-energy Γ-point exciton luminescence in five quantum wells around the interlayer Γ-X crossing in the picosecond time domain. Numbers in the figure represent the hydrostatic pressure in kbar. Note that the rise of the luminescence drastically decreases around the Γ-X crossover. In the figure P_c denotes the critical pressure bringing about the Γ-X crossover estimated from Fig. 2.

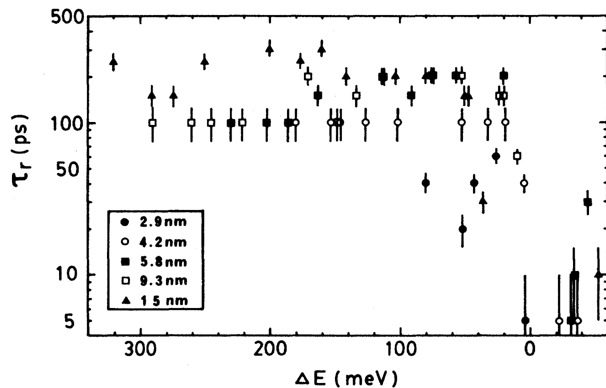

FIG. 7. Rise time of the exciton luminescence as a function of the energy separation ΔE between the lowest Γ subband energy and the X energy in AlAs. Data for the 2.9-, 4.2-, 5.8-, 9.3-, and 15-nm wells are plotted by solid circles, open circles, solid squares, open squares, and solid triangles, respectively.

the lowest exciton luminescence is determined only by the intrawell relaxation processes.

The interlayer processes described in Fig. 8 are expected to affect also the decay of the lowest exciton luminescence, because the X states in AlAs barriers have longer lifetimes than the Γ states in the GaAs wells. The X states in AlAs work as a reservoir. The radiative recombination between the X electrons in AlAs and the Γ holes in GaAs is strongly reduced because of the small overlap of the wave functions in the real space as well as the k space. The slow decay component observed in Figs. 3 and 4 is probably due to the retransfer from the X states in AlAs to the Γ states in GaAs, as is described in Fig. 8.

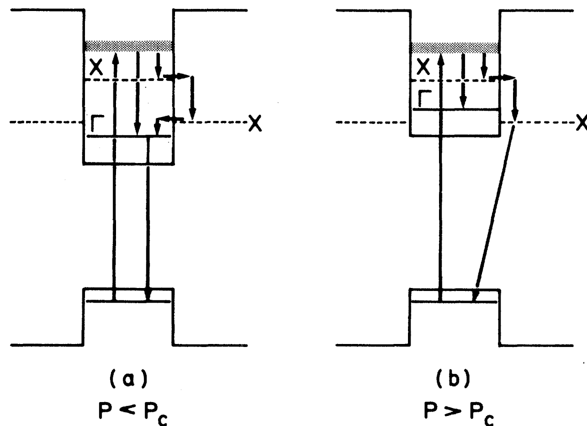

FIG. 8. Schematic figure showing mechanisms of the interlayer Γ-X transfer and the energy relaxation in the well and the barrier. The Γ state and the X state are represented by solid and dashed lines, respectively. The hatched region represents the photoexcited Γ state in the well. (a) denotes the case that the lowest Γ quantum state in GaAs is lower than the X state in AlAs corresponding to the condition $P < P_c$. (b) denotes the case that the lowest-Γ quantum state in GaAs is higher than the X state corresponding to the condition $P > P_c$.

The slow decay arises when the X states in AlAs are higher than the Γ states in GaAs by 0–300 meV. However, we cannot explain the observed systematic correlation between ΔE_m and the well thickness. Leroux *et al.* proposed a model which explains the lifetime of the Γ electrons around the Γ-X crossover in the bulk GaAs.[21] In the model, Γ electrons are in thermal equilibrium with X electrons. However, their expression cannot explain our data systematically. Especially, the model cannot explain the observed fact that ΔE_m is not always zero. We examined several other ideas to account for the data, for example, valence-band mixing due to hydrostatic strain,[22] and the reduction of the coherent length of excitons due to the pressure dependence of mass or uniaxial strain[19,23,24] coming from the different compressibilities of GaAs and AlAs. However, estimated effects are too small to explain the data.

Steiner *et al.* studied the Γ-X mixing effects by using the time-resolved luminescence with a resolution of 20 ns.[14] They observed the common slow decay in both the Γ-Γ and Γ-X luminescence, when the X states in $Al_{0.24}Ga_{0.76}As$ are lower than the Γ quantum states in GaAs. They clarified that the link between the Γ states in GaAs and the X states in $Al_{0.24}Ga_{0.76}As$ is observed, even if the X states are lower than the Γ quantum states by 40 meV. They proposed the Auger decay of the X states and the interlayer transfer of hot electrons. Similar processes may work in our sample. However, their observation dealt with a different situation than ours because their barrier layers were photoexcited, the X state was lower than the Γ state, and their time resolution was longer than 20 ns.

IV. CONCLUSIONS

Time-integrated and time-resolved luminescences of GaAs-AlAs single quantum wells under hydrostatic pressure were measured at 77 K. All of the luminescence from GaAs single quantum wells 2.9, 4.2, 5.8, 9.3, and 15 nm thick shifted to higher energy with the application of the hydrostatic pressure and quenched when the luminescence approached 1.83 eV. The quenching is ascribed to the interlayer Γ-X crossing, where the lowest Γ quantum state in GaAs crosses the X state in AlAs. With the application of hydrostatic pressure, the rise and decay times of the lowest Γ exciton were modified in the picosecond and nanosecond time domains. The slow decay component, ranging from several nanoseconds to a few tens of nanoseconds, was observed when the X state in AlAs is higher than the lowest Γ quantum state in GaAs by 0–300 meV. The rise of the Γ exciton luminescence was drastically reduced around the interlayer Γ-X crossing. The results were explained by taking account of the interlayer transfer between the Γ states in GaAs and the X states in AlAs.

ACKNOWLEDGMENTS

High-quality samples used in this study were kindly supplied by Dr. Y. Mori and Dr. N. Watanabe at Sony Corporation. The authors wish to thank Professor J.

Nakahara at Hokkaido University for valuable discussions. They also wish to thank Dr. A. Ono and Dr. F. Sakuma at National Research Laboratory of Metrology for the kind loan of a standard lamp. This work was in part supported by Grant-in-Aid No. 01604515 for Scientific Research on Priority Areas, New Functionality Materials—Design, Preparation, and Control—by the Ministry of Education, Science and Culture of Japan. A part of this work was done at the Cryogenic Center, University of Tsukuba.

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