Extremely slow energy relaxation of a two-dimensional exciton in a GaAs superlattice structure

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Time-resolved photoluminescence spectra of a two-dimensional exciton in a GaAs superlattice structure were measured as a function of photon energy from a picosecond dye laser. When the photon energy of the excitation laser is close to the luminescence of the heavy-hole exciton, the rise time of the photoluminescence is very short. However, in a sample of high quality, an extremely slow rise time of 400 ps was observed during subband excitation. Three different relaxation processes, such as the relaxation of the heavy-hole exciton band, the relaxation from the electron-hole pair to the heavy-hole exciton, and the relaxation from the light-hole exciton to the heavy-hole exciton, are proposed. The relaxation time determined in the heavy-hole exciton band is (45±20 ps)/meV, the generation rate of the heavy-hole exciton from the bottom of the electron-hole pair is 1/(190±20 ps), and the generation rate of the heavy-hole exciton from the light-hole exciton is 90±20 ps.

I. INTRODUCTION

Excitons in semiconductors play an important role in optical properties as elementary excitations at low temperature. The relaxation process and the generation rates of the excitons are important parameters in the study of the dynamics of excitons. In bulk GaAs, the binding energy of the exciton is about 4 meV and the excitonic nature is not so strong. However, in the case of an exciton in a quantum-well structure of GaAs, the two-dimensional (2D) exciton has a large binding energy[1] and strong oscillator strength[2] in comparison to those of a three-dimensional exciton in a bulk crystal. These are explained in terms of the quantum confinement of the wave functions of the electron and hole by the potential barrier. As a result, the excitonic nature of the absorption and luminescence is enhanced. In addition, the band structure of the superlattice changes according to the change of the point-group symmetry. The degenerate valence band at $K=0$ in the 3D structure decomposes into the heavy-hole band and the light-hold band. Both the heavy-hole exciton and the light-hole exciton exist in the quantum-well structure. In such systems, the heavy-hole exciton, the light-hole exciton, and the electron-hole pair in the subband can be selectively generated according to the photon energy of the excitation source. Thus, in the 2D structure, the dependence of the relaxation process on the type of initially photogenerated carrier can be studied.

In order to investigate the relaxation processes and the generation mechanism of a 2D exciton in a GaAs superlattice structure, time-resolved photoluminescence spectra of the $n=1$ heavy-hole exciton (the 2D exciton) were measured as a function of the photon energy of the excitation laser. The measurement system has a time resolution of better than 15 ps and this allowed the rise time of the luminescence to be determined with high precision. The time response of the photoluminescence in the high-quality superlattice, which was characterized by the broadening of Landau level in the excitation spectrum of the photoluminescence under the magnetic field, showed a very slow rise time. This slow rise time was strongly dependent on the photon energy of the excitation laser. In contrast, the decay time was independent of the excitation photon energy. The rise time of the photoluminescence indicates that in the energy relaxation process, the initial photogenerated carrier relaxes the excess energy with subsequent generation of the 2D exciton. The experimental results were analyzed by a rate-equation approach with the following process considered: the generation rate of the 2D exciton from the electron-hole pair, that from the light-hole exciton, and the intraband relaxation time in the 2D exciton band. The extremely slow energy relaxation process of the 2D exciton originated to the initial photogenerated carrier has been discussed.

II. EXPERIMENTAL RESULTS

A. Characterization of samples

Two types of GaAs superlattice structures, referred to as sample I and sample II in this paper, were grown by a molecular-beam epitaxy (MBE) technique.[3] Sample I was a GaAs (86 Å)–AlAs (75 Å) superlattice structure composed of 100 periods. Sample II was a GaAs (100 Å)–$Al_{0.4}Ga_{0.6}As$ (100 Å) of 30 periods. The quality of sample I is superior to that of sample II, and this judgment is based on criteria which will be described below.

The time-integrated photoluminescence spectrum collected by picosecond dye-laser excitation and the excitation spectrum of the photoluminescence obtained with a monochromatic Xe lamp for sample I are shown in Fig. 1. The photoluminescence due to the radiative recombination of the $n=1$ heavy-hole exciton (the 2D exciton) was observed at 1.5710 eV and the full width at half maximum (FWHM) was 5.3 meV at lattice temperatures of 1.8 and 10 K. The excitation spectra were detected at the peak photon energy of the heavy-hole exciton (the 2D exciton) luminescence. When a magnetic field is applied perpendicular to the heterointerface, Landau levels are generated in the continuum subband. As shown in Fig. 1 by the excitation spectra, subband bunching is observed and the peak structures due to the transition between the Landau level of the electron and that of the heavy hole appear. The excitation spectra were measured as a function of the magnetic field up to 6 T. Linear extrapolation of the peak photon energies of the $N=2\sim5$ Landau levels to zero field converges to the subband edge.[4,5] The subband edge is determined to be 1.5870 eV$\pm$1.1 meV. The same experiments were also performed on sample II. The photoluminescence of the 2D exciton was observed at 1.561 eV with a FWHM of 7.0 meV. Therefore, sample II has a large fluctuation of the potential well due to the interface roughness in comparison with that of sample I.

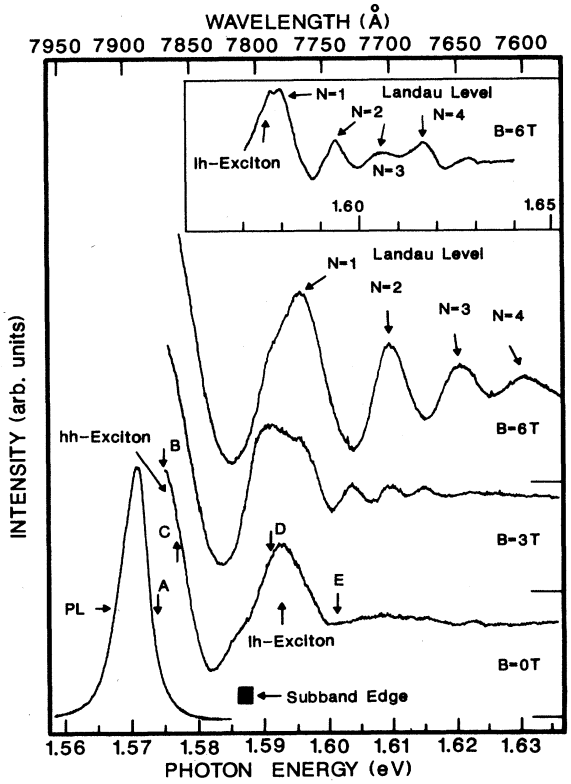

FIG. 1. Time-integrated photoluminescence spectrum of the heavy-hole exciton obtained by picosecond dye-laser (7740 Å) excitation and excitation photoluminescence spectra by a Xe lamp at zero field, 3 T, and 6 T of sample I. The inset represents the excitation spectrum of sample II at 6 T. The excitation spectra were detected at the peak photon energy of the luminescence. "*hh*-exciton" and "*lh*-exciton" represent the heavy-hole exciton band and the light-hole exciton band, respectively. Landau levels are indicated by arrows *N*. The shaded rectangle represents the subband edge. Time responses at the photon energy of the dye laser marked by arrows *B*, *C*, *D*, and *E* are shown in Figs. 5–7. The arrows *A*, *B*, *C*, *D*, and *E* are used in Figs. 2–4 with the same meaning.

The relaxation time strongly depends on the quality of the crystal and the quantitative characterization of a sample is a necessary condition when the relaxation rate is presented. However, it is difficult to characterize the quality of the undoped well layer by conventional methods. Accordingly, the sample characterization was performed by the magnet optics method along with the determination of the subband edge.

Since the broadening of the Landau level[6–9] has been attributed to impurity scattering, carrier-phonon scattering, and the fluctuation of the local potential, the appearance of the Landau levels strongly depends on the quality of the crystal. Although the absolute value of the impurity concentration is not clear, it is possible to select a high-quality sample by comparison of the magnetic field strengths when Landau levels first begin to appear. The broadening parameter of each Landau level was obtained from the excitation spectra. In the case of sample I, Landau levels were apparent at 3 T, as shown in Fig. 1. The density of states of the Landau levels was calculated by the path-integral method[10] and represented as a Gaussian function making use of the broadening parameter. The broadening parameter at 6 T of sample I for the $N=2$ Landau level is 2.5$\pm$0.5 meV at a lattice temperature of 10 K, when the inhomogeneous broadening of the 2D structure is equivalent to the width of the photoluminescence of the 2D exciton. This value agrees with the experimental result for the high-mobility sample.[9] It is concluded that this superlattice structure has high-quality well layers. In contrast, the Landau levels of sample II began to appear at 5 T and the distinct subband bunching structure was not observed until 6 T, as shown in the inset of Fig. 1. Therefore, the overall quality of sample II together with the interface roughness is worse than that of sample I.

B. Time-resolved photoluminescence spectra

Time-resolved photoluminescence spectra at low temperature were obtained by the excitation of a picosecond dye laser. A tunable picosecond dye laser (LD 700) was synchronously pumped by a cw mode-locked Kr laser (6471 Å). The repetition rate was 82.4 MHz and the linewidth of the dye laser was about 4 Å. The detection system was a synchronous scanning streak camera with a 25-cm monochromator. The observed FWHM of the picosecond dye laser was 15 ps, when 2.5×10^9 pulses were accumulated. The actual pulse width of the dye laser is about 2 ps, therefore the time resolution is limited by the time jitter of the dye laser. Since experiments were per-

formed under weak excitation conditions, the average intensity of incident dye-laser pulses was 2.5 W/cm^2 and the density of photoexcited carriers per unit plane was estimated to be 2×10^8/cm^2 $\sim 2\times10^9$/cm^2. The spectrum excited by the picosecond dye laser was identical to that obtained by a He-Ne laser (160 mW/cm^2). Therefore, the renormalization of the band gap[11] and the rise of the lattice temperature by the short pulse excitation did not occur. Interactions between the exciton-exciton and the exciton carrier are neglected in the remaining discussion.

The two-dimensional image (time delay versus photon energy) of the intensity of the photoluminescence of the 2D exciton of sample I is shown as a contour representation in Fig. 2, for a dye-laser wavelength of 7740 Å. The same measurements were carried out as a function of the photon energy of the dye laser, ranging from resonant excitation of the 2D exciton to $\Delta E = 150.6$ meV. This point is below the $n=2$ heavy-hole exciton band. ΔE is defined as the interval between the photon energy of the dye laser and the peak photon energy of the luminescence (1.5710 eV). The rise time was quite sensitive to the excitation photon energy. The decay time, which was measured at slower speed of the streak camera, was independent of the excitation photon energy. As shown in Fig. 2, the rise time of the high energy side of the luminescence band was equal to that of the center of the luminescence band. In the following discussion, the time response of the photoluminescence of the 2D exciton is assumed to be equal to the time response of the photoluminescence band, which is determined with a spectral width of 2 meV around the peak. The rise-time dependence upon the energy interval (ΔE) is represented in Fig. 3. The rise time changes drastically at the excitation of the subband edge and at the light-hole exciton band. In the case of excitations above $\Delta E = 30.4$ meV (the excitation of the subband), the rise time was independent of energy interval. When the wavelength of the dye laser was 7875 Å ($\Delta E = 3.0$ meV, the arrow *A* in Fig. 1), the rise time nearly exceeded the detection limit of the system (subpicosecond).

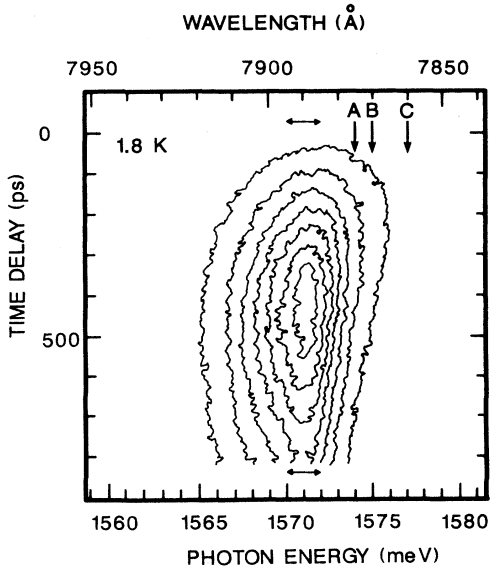

FIG. 2. Time-resolved photoluminescence spectrum of the heavy-hole exciton of sample I. The luminescence intensity is represented by the contour map in the two-dimensional (the time delay and the photon energy) space. The wavelength of the dye laser was 7740 Å. The time responses of the photoluminescence which are shown in Figs. 5–7 are obtained at the peak photon energy within a spectral range of 2 meV. This spectral range is represented by the horizontal arrows in this figure. The arrows *A*, *B*, and *C* correspond to those of Fig. 1.

III. DISCUSSIONS

The model for the relaxation process in the superlattice structure is shown schematically in Fig. 4. The relaxation process of the 2D exciton is composed of three channels according to initial photogenerated carriers: (1) the heavy-hole exciton, (2) the light-hole exciton, and (3) the electron-hole pair. The rate equation is used to quantitatively evaluate the relaxation process. Although the parameters in the rate equations are relaxation times (T_s, T_g, T_{lh}, T_h) and a decay time (T_d), the experimentally observed decay time of 230±20 ps of sample I at 1.8 K is substituted into the rate equations. There exists a ground state of the exciton, an excited state of the exci-

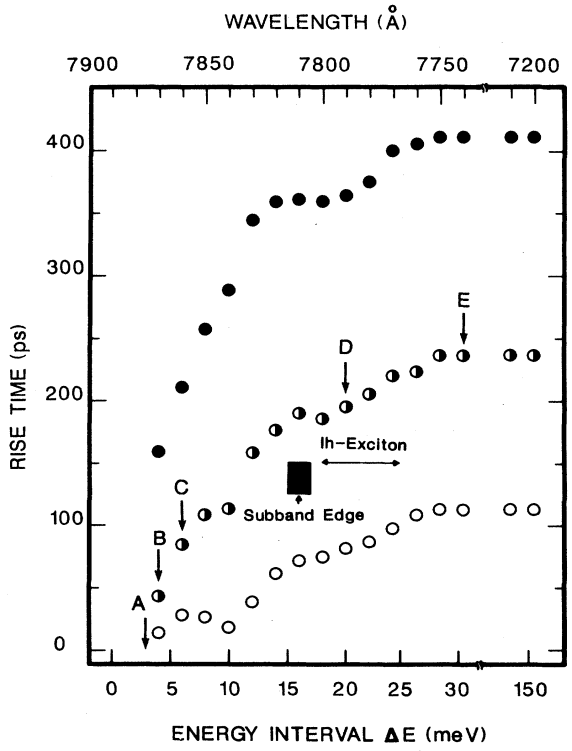

FIG. 3. Rise times of sample I as a function of the energy interval ΔE. ●, ◑, and ○ represent time intervals from the time zero to the peak (100%), 70% of the peak intensity, and 30% of the peak intensity, respectively. The horizontal arrow *lh*-exciton indicates the FWHM of the light-hole exciton band. The shaded rectangle represents the subband edge.

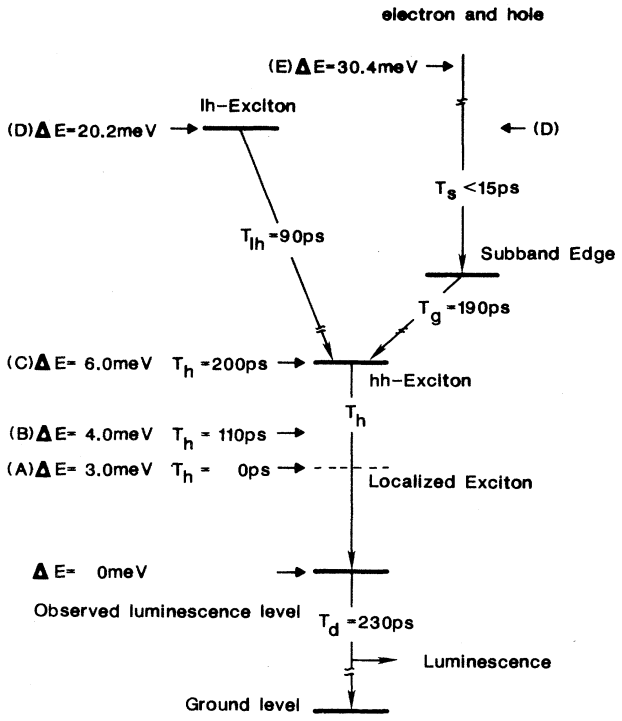

FIG. 4. Energy relaxation process in the superlattice structure. The horizontal lines represent the energy levels for the rate equation. The horizontal arrows *A*, *B*, *C*, *D*, and *E* correspond to those in Fig. 1. At the excitation of the light-hole exciton (*lh*-exciton) band (the arrow *D*), two parallel relaxation processes, (1) the light-hole exciton process and (2) the electron and the hole process, are proposed. The relaxation times (T_s, T_g, T_{lh}, T_h, T_d) indicate as follows: T_s, the relaxation time of the electron and the hole in the subband; T_s, shorter than the time resolution of our detection system; $(T_g)^{-1}$, the generation rate of the heavy-hole exciton (*hh*-exciton) from the electron and the hole; T_{lh}, the relaxation time from the light-hole exciton to the heavy-hole exciton; T_h, the relaxation time of the heavy-hole exciton in the heavy-hole exciton band. T_h depends on the excitation photon energy; T_d, the decay time.

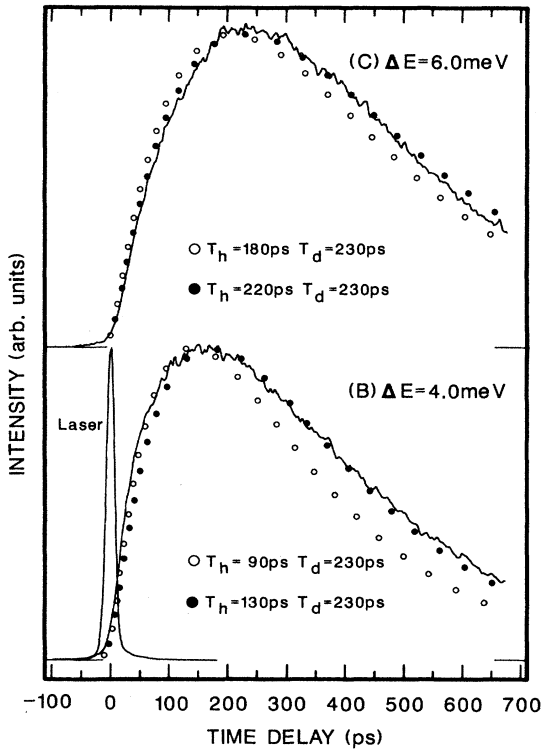

FIG. 5. Time responses of the luminescence of sample I at 1.8 K and the dye laser at the excitation of the 2D exciton band. Time responses of (*B*) and (*C*) are obtained by the excitation photon energies marked by the arrow *B* and *C* in Fig. 1, respectively. Open and solid circles represent calculated results from the three-level rate equation.

ton, and a subband edge near the bottom of the subband. The energy level has an inhomogeneous linewidth and overlap of the energy levels occur at the boundaries of each state. Analyses are performed for the excitation photon energies, shown in Fig. 1 arrows as *B*, *C*, *D*, and *E*, where the type of the photogenerated carrier is clearly determined.

A. Relaxation process in the 2D exciton band

In the case of resonant excitation of the 2D exciton (the heavy-hole exciton) band, the 2D exciton is directly photogenerated. Time responses of the photoluminescence are shown in Fig. 5. The rise time becomes faster with decreasing ΔE. This ΔE-dependent relaxation process is explained by a rate equation composed of three levels: (1) the excitation laser level (the initial photogenerated 2D exciton), (2) the exciton level (the observed luminescence photon energy), and (3) the ground level. The single fitting parameter is the relaxation time in the 2D exciton band (T_h). As shown in Fig. 5, the calculated results give good agreement with experimental results. Relaxation times (T_h) are found to be 200±20 ps at $\Delta E = 6.0$ meV, 110±20 ps at $\Delta E = 4.0$ meV, and 0 ps at $\Delta E = 3.0$ meV for a decay time (T_d) of 230±20 ps. The relaxation rate of (45±20 ps)/meV is also determined for the relaxation in the heavy-hole exciton band.

With increasing lattice temperature, the rise time became faster and the FWHM of the luminescence band increased. The relaxation time (T_h) became zero at a lattice temperature of 20 K, for excitation photon energies on the higher energy side of the 2D exciton band. The FWHM of the luminescence band at 20 K was found to be 7.9 meV due to the enhancement of the interaction between the 2D exciton and thermally populated phonon modes.

The rise time of sample II became faster when the excitation was below the subband edge. The relaxation time in the 2D exciton band (T_h) at 1.8 K, however, is almost zero when the excitation is on the high energy side of the

2D exciton band. The slow relaxation in the 2D exciton band is not observed. Therefore, the slow relaxation is inherent in the high-quality superlattice.

A heterointerface has a 1-ML fluctuation with some lateral size.[12] The excitonic nature of a quantum-well structure is affected by such a random potential. It depends on the ratio of the Bohr radii of the 2D exciton and the lateral size of the island. If the lateral size is very large in comparison with the Bohr radii of the 2D exciton, a distinct splitting structure in the photoluminescence spectrum originates from the 1-ML fluctuation. In such a case, the relaxation process from the upper level to the lower level in the 2D exciton band should be observed.[13] In sample I, as shown in Fig. 2, no splitting was observed and the 2D exciton band is inhomogeneously formed. Moreover, the red shift of the luminescence peak against the absorption line is observed, as shown in Fig. 1. This Stokes shift is approximately 3 meV. The photogenerated excitons above the center of the absorption line, where these points are marked by arrows B and C in Fig. 1, do not recombine radiatively before losing their kinetic energy. Excitons at B and C relax toward the low energy side of the 2D exciton band. This relaxation process represents the relaxation time (T_h). The relaxation time (T_h) becomes shorter with decreasing ΔE and the relaxation process disappears when the excitation is at the center of the absorption line ($\Delta E = 3.0$ meV). The dynamics of the 2D exciton in the 2D exciton band also changes across the absorption-line center. The difference of the excitonic nature in the 2D exciton band is explained by Hegarty *et al.*,[14] who state that the excitons were effectively localized below the line center and delocalized above it; the delocalized exciton can move along the heterointerface like a free exciton in a bulk crystal. Therefore, the photogenerated exciton above the center of the absorption line is the delocalized exciton and the luminescence is due to the radiative recombination of the localized exciton. The relaxation process represented as the relaxation time (T_h) indicates that the delocalized exciton relaxes the excess kinetic energy by intraband scattering and localizes into the potential fluctuation. The center of the absorption line of the 2D exciton at $\Delta E = 3.0$ meV corresponds to the mobility edge.

The main relaxation processes of the 2D exciton in the exciton band during weak excitation are results of impurity scattering and phonon scattering. The interaction between the 2D exciton and the acoustic phonon (the deformation-potential coupling and the piezoelectric coupling)[15] is the dominant relaxation process, since the LO phonon (36 meV) does not contribute to the relaxation. With increasing lattice temperature, the number of the phonon modes increases and, as a consequence, the exciton-phonon interaction is enhanced. This is consistent with the observed fast relaxation at 20 K. The drastic decrease of the relaxation time (T_h) from the high-quality superlattice (sample I) to the lower-quality superlattice (sample II) is explained as a result of the increase of the roughness of the heterointerface and the impurity concentration. With increasing potential fluctuation, the 2D exciton cannot move along the heterointerface and localizes immediately. Such a slow relaxation process in the exciton band can be observed in the high-quality sample as a broadening exciton line. Therefore, only the GaAs superlattice structure satisfies the above-mentioned two conditions.

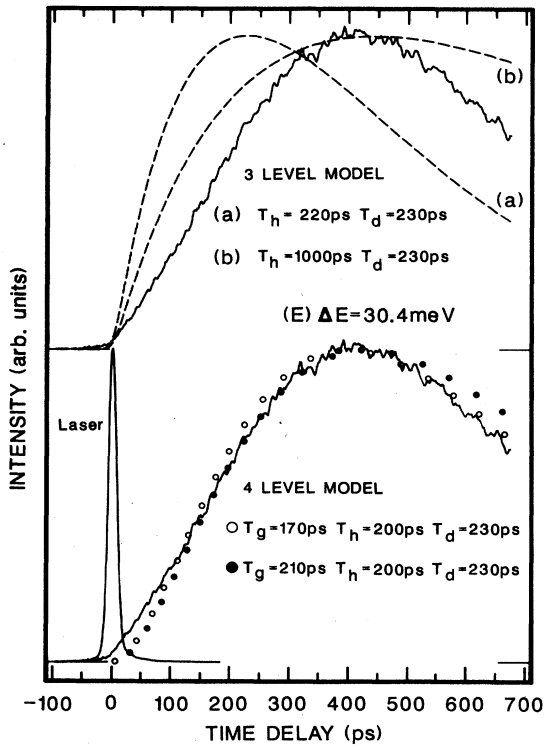

FIG. 6. Time responses of the luminescence of sample I at 1.8 K from dye-laser excitation of the subband. The time response of (E) is obtained by the excitation photon energy marked by the arrow E in Fig. 1. Open and solid circles represent calculated results from the four-level rate equation. Dashed lines [(a) and (b)] are calculated results from the three-level rate equation.

B. Generation process of the 2D exciton from the electron and the hole

Under subband excitation, the drastic delay of the rise time was observed in comparison with that of the 2D exciton band, as shown in Fig. 6. This delay is attributed to the different nature of the photogenerated initial carrier; the electron-hole pair and the 2D exciton. At the excitation of the subband, both the relaxation process of the electron and the hole in the subband and the generation process of the 2D exciton from the electron and the hole should be considered in addition to the relaxation process in the 2D exciton band, as shown in Fig. 4 (T_s, T_g, T_h, and T_d). The rise time is so slow that the time response of the photoluminescence cannot be explained by the rate equation composed of the three levels, as shown in Figs. 6(a) and 6(b). The experimental result is explained by a rate equation composed of four levels. The relaxation process at the excitation of the subband is proposed as

follows. The photogenerated electron and hole in the subband relax by interaction with the LO phonon and the LA phonon. In our experiment, the rise time was independent of the photon energy of the dye laser for ΔE greater than 30.4 meV. The relaxation time of the LO-phonon interaction (T_s), as shown in Fig. 4, is equal to or less than a picosecond;[16,17] the dependence of relaxation time (T_s) on the photon energy could not be observed with present detection systems. After relaxation to the bottom of the subband, the electron and the hole are thermally distributed and generate the 2D exciton at a rate of ($1/T_g$). The 2D exciton relaxes (T_h) and recombines radiatively (T_d). Therefore, the parameters of the four-level rate equations are the generation rate of the 2D exciton ($1/T_g$), the relaxation time of the 2D exciton in the 2D exciton band (T_h), and the decay time (T_d). The best fit is shown in Fig. 6, when relaxation times are taken as $T_g = 190 \pm 20$ ps, $T_h = 200 \pm 20$ ps, and $T_d = 230 \pm 20$ ps.

As previously mentioned, the slow relaxation process under subband excitation is attributed to the slow generation rate of the 2D exciton ($1/T_g$). An exciton is generated from an electron and a hole by Coulomb capture. In our experiments, since the intensity of the dye laser was weak, this generation process is very slow. The relaxation time in the 2D exciton band (T_h) coincides with that of the excitation at $\Delta E = 6.0$ meV. This point is the high energy level of the 2D exciton band. Therefore, the relaxation process from the subband and that of the 2D exciton band are both of the cascade type. The 2D exciton generated from the electron-hole pair is the delocalized exciton and has a large kinetic energy.

C. Relaxation process from the light-hole exciton to the heavy-hole exciton

During excitation of the light-hole exciton band, both the light-hole exciton and the electron-hole pair are generated as per their transition probabilities. As shown in Fig. 1, the structure of the light-hole exciton is easily observed and luminescence of the light-hole exciton is not observed under weak excitation conditions. Therefore, the light-hole exciton contributes to the luminescence of the heavy-hole exciton (the 2D exciton) and a generation channel of the heavy-hole exciton from the light-hole exciton exists. If the 2D exciton is generated by the dissociation of the light-hole exciton, the rise time at the resonant excitation of the light-hole exciton band should be equal or slower in comparison to that of the subband. The experimental result, however, shows that the rise time is fast, as shown in Fig. 3. Therefore, the generation process of the 2D exciton from the light-hole exciton and that from the electron hole differ remarkably and proves that the heavy-hole exciton is directly generated from the light-hole exciton. The time response at $\Delta E = 20.2$ meV (arrow D in Fig. 1) consists of two parallel relaxation processes as represented in Fig. 7 (D). The luminescence from the light-hole exciton [Fig. 7 (a)] is resolved by subtracting the time response of the luminescence at the excitation of the subband [$\Delta E = 30.4$ meV, Fig. 7 (E)] from the observed luminescence [$\Delta E = 20.2$ meV, Fig. 7 (D)]. The ratio of the intensities from the light-hole exciton and the subband (heavy-hole) is found to be 1:1.25, which is obtained from the excitation spectrum shown in Fig. 1. When the relaxation process in the 2D exciton band from the light-hole exciton is assumed to be of the cascade type as well as in the case of the excitation of the subband, for $T_h = 200 \pm 20$ ps and $T_d = 230 \pm 20$ ps, the relaxation time between the light-hole exciton and the heavy-hole exciton (T_{lh}) is found to be 90 ± 20 ps by the fitting procedure shown in Fig. 7 (a).

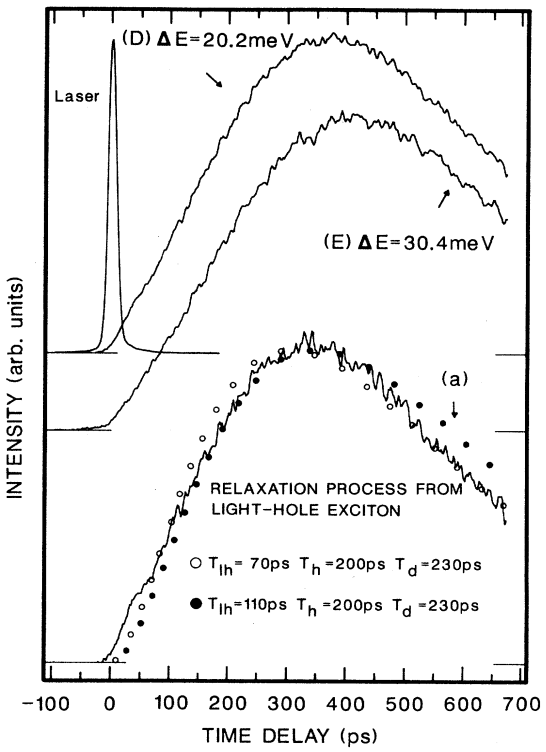

FIG. 7. Time response (D) of the luminescence of sample I at 1.8 K from dye-laser excitation of the light-hole exciton band. The excitation photon energy is marked by the arrow D in Fig. 1. The time response of (E) and that of (a) indicate the luminescence contributed to the subband and to the light-hole exciton, respectively. Open and solid circles represent calculated results from the four-level rate equation.

The degenerate valence band in the superlattice structure is divided into the heavy-hole band ($J = \frac{3}{2}$, $m_z = \pm\frac{3}{2}$) and the light-hole band ($J = \frac{3}{2}$, $m_z = \pm\frac{1}{2}$) by the change of point-group symmetry. Therefore, the excitons have the same angular momentum. During the relaxation process (T_{lh}) at $\Delta E = 20.2$ meV, the light-hole exciton relaxes with an energy of 14.2 meV along with an angular-momentum relaxation and forms the heavy-hole exciton. Our experiment suggests that the light-hole exciton interacts strongly with the heavy-hole exciton. This strong interaction comes from the exchange interaction of the excitons by enhancement of the overlap of the wave functions of the electron and the hole.

IV. CONCLUSION

The relaxation process of the 2D exciton in a GaAs superlattice structure has been studied under weak excitation conditions by picosecond spectroscopy. The extremely slow energy relaxation was observed only in the high-quality superlattice at a lattice temperature of 1.8 K. The relaxation process depends on the photon energy of the dye laser, and three kinds of relaxation processes, the heavy-hole exciton, the light-hole exciton, and the electron-hole pair, are classified according to the difference in initially photogenerated carriers. The relaxation rate in the 2D exciton band is 45 ps/meV, the generation rate of the 2D exciton from the electron-hole pair is 1/(190 ps), and the relaxation time between the light-hole exciton and the heavy-hole exciton is 90 ps.

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