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Tunneling dynamics of photogenerated carriers in semiconductor superlattices

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The tunneling dynamics of photogenerated carriers in GaAs-$Al_{0.29}Ga_{0.71}As$ multiple-quantum-well structures is studied using picosecond spectroscopy with an electric field perpendicular to the well layers. Drastic changes, such as an increase in the photocurrent, a decrease in the exciton luminescence, and a change in the exciton lifetime, take place simultaneously for electric fields of $\sim 5.0\times 10^3$ V/cm. These changes are ascribed to the onset of exciton dissociation and electron tunneling through the $Al_{0.29}Ga_{0.71}As$ barrier layers. The electron tunneling rate is determined to be 1/(430 ps) and is compared with calculations.

Recently, the dynamics of carriers in semiconductor superlattices has attracted growing interest. There are two directions of motion of the carriers in semiconductor superlattices. One is motion in the plane of heterostructures and the other is motion across the heterostructures. So far, the former has been studied extensively in two-dimensional carrier systems by means of transport experiments. However, the study of the latter has been relatively scarce and limited to current-voltage characteristics since the first observation of resonant tunneling of electrons.[1] Current-voltage characteristics could be used to clarify the energy levels in superlattices but not the dynamical aspects of tunneling. Therefore, new techniques have been required.

In this work, picosecond spectroscopy has been used to study the motion of photogenerated carriers in the direction of the superlattice. We probed the exciton luminescence in GaAs-$Al_{0.29}Ga_{0.71}As$ multiple-quantum-well structures (MQW) in an electric field applied in the direction of the superlattice. Exciton luminescence quenching and the change in the exciton lifetime have already been observed in a GaAs-$Al_xGa_{1-x}As$ MQW in an electric field.[2-4] It has been pointed out that these phenomena are possibly correlated with the field-induced separation of electrons from holes in a well or the tunneling of electrons or holes across the barrier potential. In the preceding study, definitive evidence of the tunneling was not produced. In addition, the electric field in the MQW was not accurately determined, so that the experimental data could not be compared with the calculation. This is because the electric field is screened, to some extent, by the excess photogenerated carriers. We have overcome this difficulty. We have determined the electric field in the MQW by the peak shift of the heavy-exciton luminescence. In addition, we found that the exciton lifetime is surely dominated by the tunneling of electrons across the $Al_{0.29}Ga_{0.71}As$ barrier above a certain critical electric field because the tunneling rate of electrons becomes faster than the decay rate of the exciton population, due to the other processes including radiative recombination. Then, the tunneling rate can be estimated by transient measurement of the exciton luminescence. The tunneling rate thus obtained is compared with the calculated one on the basis of the precise knowledge of the electric field in the MQW.

The sample is a *p-i-n* diode structure grown by molecular-beam epitaxy.[5] The main part consists of 100 alternate periods of undoped 120-Å GaAs well layers and undoped 58-Å $Al_{0.29}Ga_{0.71}As$ barrier layers. The MQW are surrounded by the *p*-type and *n*-type $Al_{0.54}Ga_{0.46}As$ cladding layers 1 μm thick. The MQW and cladding layers are sandwiched between the p^+-type GaAs cap layer (0.5 μm thick) and the *n*-type GaAs substrate. Electrical Ohmic contacts are made on both the p^+-GaAs cap layer and *n*-GaAs substrate. With the sample in a perpendicular electric field at 4.2 K, we made measurements of the photoconductivity, the photocurrent-voltage characteristics, the exciton luminescence, the exciton-luminescence–voltage characteristics, and the picosecond transient response of the exciton luminescence.

For all measurements except photoconductivity, a dye laser synchronously pumped by a cw mode-locked Nd^{3+}:YAG laser (Quantronix 416) (where YAG is yttrium aluminum garnet) is used as the excitation source. The dye laser gives 1–2-ps light pulses with a repetition rate of 82 MHz. The laser dyes Rhodamine 6G and DCM are used to generate 605-nm (2.05-eV) and 678-nm (1.83-eV) light pulses, respectively. The band-gap energies of $Al_{0.54}Ga_{0.46}As$, $Al_{0.29}Ga_{0.71}As$, and GaAs are 2.15 eV (X), 1.88 eV (Γ), and 1.519 eV (Γ), respectively.[6] Therefore, neither 605- nor 678-nm light excites the $Al_{0.54}Ga_{0.46}As$ cladding layers. The 605-nm light excites both the well layers and the barrier layers, while the 678-nm light excites only the well layers. No significant difference was found in the results, including the time-resolved result, whether the barrier layers were excited or not. Therefore, only the results for the 605-nm light excitation are presented in this paper. The laser beam is loosely focused on a *p-i-n* diode sample immersed in liquid helium. To reduce the electrical screening by photogenerated carriers, the excitation laser intensity is reduced to being as weak as possible for the picosecond time-resolved study. The excitation density is about 10 mW/cm^2. The luminescence spectra are recorded by using a 50-cm monochromator. The exciton luminescence is temporally analyzed by using a synchroscan streak camera with 140-μm slit width. Then the time resolution is 42 ps.

The energy shift of the heavy exciton can be used to

determine the electric field in the MQW as described below. We measured the photoconductivity spectra of the sample in the electric field by using the incandescent lamp. The result is shown in the inset of Fig. 1. In the photoconductivity spectra, the heavy excitons in the quantum wells produce a peak. The heavy-exciton peak shifts toward lower energy with the increase in the applied electric voltage. The shifts do not vary with further reduction of the excitation light level. The built-in voltage V_{bi} was determined to be 1.8 V from the curve of the current-voltage characteristics. The sample has no buffer layers. Therefore, the field is simply calculated to be $F=(V_{ext}+V_{bi})/d$, where V_{ext} is the externally applied voltage and $d=1.78\ \mu$m is the total thickness of the MQW. Following the calculation of Bastard for an infinite well, the energy shift of the lowest transition between the electron and hole sublevels, ΔE, is proportional to the square of the electric field F as follows:[7]

$$\Delta E=-2.135\times10^{-3}(m_e^*+m_{hh}^*)e^2F^2L_z^4/\hbar^2\ , \qquad (1)$$

where e is the charge of electron, the z axis is along the superlattice direction, L_z is the well layer thickness, and m_e^* ($=0.665m_0$) and m_{hh}^* ($=0.45m_0$) are effective masses of the electron and the heavy hole, respectively. The change of the exciton binding energy is neglected because it is smaller than ΔE by an order of magnitude.[8] In fact, Eq. (1) explains the energy shift of the excitons in a strikingly complete manner, as is shown in Fig. 1. Conversely, the electric field in the MQW can be determined from the peak shift of excitons.

Under the picosecond laser excitation, the exciton-luminescence energy shifts toward lower energy with an increase in the externally applied voltage as is shown in the inset of Fig. 2. Then, however, the peak shift is not so large as expected. We attribute this disagreement to the screening of the electric field by the high density of photogenerated carriers[3] because we observe that the peak shift is reduced with an increase in the excitation intensity. Therefore, we do not estimate the electric field from the applied voltage. Instead, we determine the electric field from the peak shift on the basis of Eq. (1). In Fig. 2, both the exciton-luminescence intensity and the photocurrent across the superlattices are plotted as a function of electric field thus estimated. A sudden change is observed at a field of $F_t\sim5.0\times10^3$ V/cm. At this value, the exciton-luminescence intensity decreases and the photocurrent increases. These facts indicate that the excitons dissociate at this field and that electrons or holes tunnel through the potential barriers and contribute to the photocurrent. At the field of $F_t\sim5.0\times10^3$ V/cm, the field gain for the electrons or holes amounts to 9 meV when they move by a superlattice period

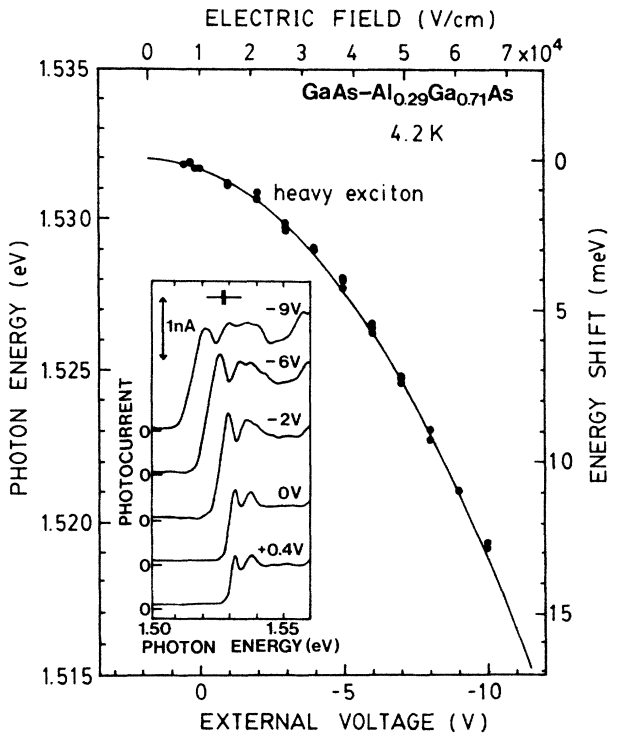

FIG. 1. Peak shift of the heavy excitons observed in photoconductivity spectra as a function of the applied electric voltage V_{ext}. Experimental data are shown by solid circles. The built-in voltage V_{bi} is +1.8 V. Electric field in the MQW is calculated to be $(V_{ext}+V_{bi})/d$. The solid line is the calculated one on the basis of Eq. (1). In the inset, photoconductivity spectra under the externally applied voltage are shown. The lowest-energy peak corresponds to heavy excitons.

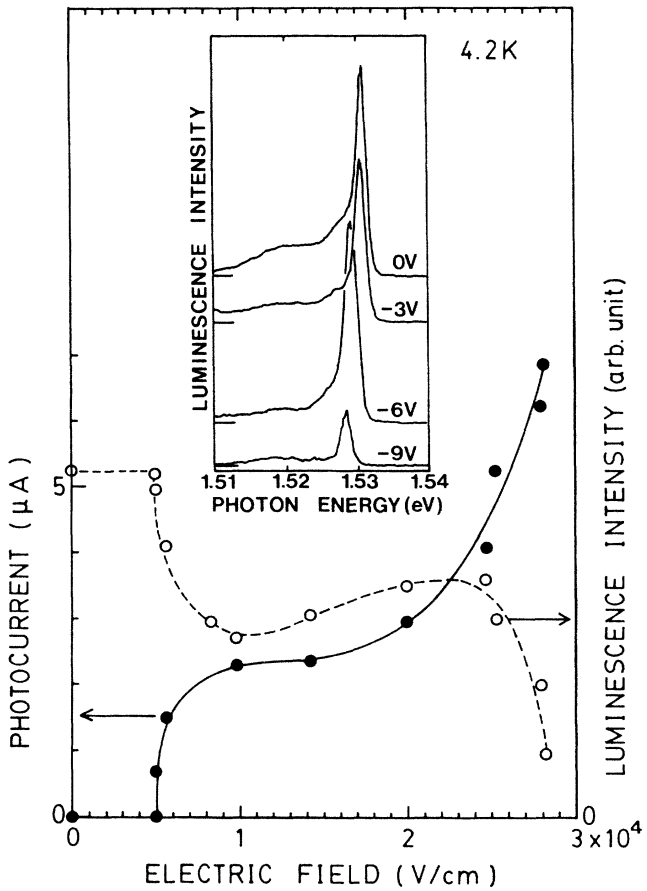

FIG. 2. Luminescence itensity of heavy excitons (○) and photocurrent (●) as a function of the electric field in the MQW. Solid and dashed lines are guides for the eyes. The inset shows heavy-exciton luminescence spectra under externally applied voltage. The peak shifts of heavy excitons indicate that applied voltages 0, −3, −6, and −9 V correspond to electric fields 5.0×10^3, 9.8×10^3, 2.5×10^4, and 2.8×10^4 V/cm, respectively.

(=178 Å). The field gain is almost equal to the exciton binding energy in 120-Å quantum wells (10 meV) derived from the magneto-optical measurements.[9] The field gain compensates for the binding energy. Therefore, it is quite reasonable for excitons to dissociate at a field of $F_t \sim 5.0\times10^3$ V/cm. Curves of photocurrent and luminescence intensity show a plateau between $\sim 1.0\times10^4$ and $\sim 2.0\times10^4$ V/cm. With the further increase in the electric field up to $\sim 2.8\times10^4$ V/cm, the photocurrent increases and the exciton luminescence decreases.

The transient response of the exciton luminescence is shown in Fig. 3. At the field of $F_t \sim 5.0\times10^3$ V/cm, the temporal profile of the exciton luminescence changes drastically. Below F_t, the exciton luminescence exponentially decays with a time constant of 1.52 ns. Above F_t, the exciton-luminescence decay seems to consist of two components. The fast decay has a time constant of 430 ps. The slow one has a long time constant which has not been determined in this experiment. We determine that the change of the temporal profile comes from the tunneling of the electrons or holes because the luminescence decrease and the photocurrent increase take place at the common electric field of $F_t \sim 5.0\times10^3$ V/cm. Then, the fast decay of exciton luminescence is dominated by the tunneling of electrons or holes across the potential barriers. Therefore the tunneling rate can be estimated to be $1/(430\text{ ps}) = 2.3\times10^9\ \text{s}^{-1}$. At a field of $\sim 2.8\times10^4$ V/cm, the slow-decay component of the exciton luminescence vanishes completely and the exciton luminescence exponentially decays with a time constant of 430 ps.

It is not easy to estimate the dissociation rate of excitons in the MQW. To estimate it, we must calculate the rates of at least two processes, the dissociation of excitons, and the tunneling of electrons or holes across the barrier potential. If the excitons are not in the quantum wells and are in the electric field F, the dissociation rate of excitons, w_1, is given by the following formula which also describes the ionization rate of hydrogen atoms.[10]

$$w_1 = (16R^2/\hbar eFa_B)\exp(-4R/3eFa_B) \ , \tag{2}$$

where R (=4.2 meV) is the Rydberg energy and a_B (=136 Å) is the exciton Bohr radius. The dissociation rate reaches $2.8\times10^{13}\ \text{s}^{-1}$ at an electric field of $F_t \sim 5.0\times10^3$ V/cm. The rate is much faster than the observed rate.

Compared with the above-mentioned dissociation rate, the tunneling rate of electrons or holes across the barrier potential is expected to be slow because the $Al_{0.29}Ga_{0.71}As$ barrier potential is much higher than the Coulomb barrier. The experimental results correspond exactly with this expectation. In the Wentzel-Kramers-Brillouin approximation, the tunneling rate of electrons or heavy holes across the barrier potential w_2 is estimated to be[11]

$$w_2 = (\pi\hbar/2m^*_{e(\text{hh})}L_z^2)\exp[-(2/\hbar)\sqrt{2m^*_{e(\text{hh})}(U-E)}\,d_b] \ , \tag{3}$$

if the Coulomb interaction between the electrons and heavy holes is neglected. Here, E is the band discontinuity, U is the confinement energy of the lowest sublevel, and d_b (=58 Å) is the barrier thickness. The factor $2m^*_{e(\text{hh})}L_z^2/\pi\hbar$ is the classical period of the electron (heavy-hole) motion in the quantum well,[11] because the z component of the velocity of electrons (heavy holes) in the lowest sublevel is $\hbar k/m^*_{e(\text{hh})} = \pi\hbar/m^*_{e(\text{hh})}L_z$. Equation (3) is the expression in the case of zero applied electric field. However, this equation approximately holds under the condition $eFd \ll U-E$, even when the electric field is applied.

The rate calculated on the basis of Eq. (3) is not altered much when the confinement energy U is neglected because U is smaller than the band discontinuity E by an order of magnitude. Therefore U is neglected for simplicity. If the band-gap discontinuity split of 85:15 is correct,[12] the conduction-band discontinuity E_c is 307 meV and the valence-band discontinuity E_v is 54 meV. Then, the tunneling rate of electrons is $3.9\times10^9\ \text{s}^{-1}$ and that of holes is $2.7\times10^8\ \text{s}^{-1}$. On the other hand, the tunneling rate of electrons is $1.8\times10^{10}\ \text{s}^{-1}$ and that of holes is $4.3\times10^5\ \text{s}^{-1}$, if the band-gap discontinuity split of 57:43 (E_c=206 meV, E_v=155 meV) is correct.[13] In both the cases, tunneling of electrons dominates the field dissociation rate of excitons across the barrier potential. The calculated tunneling rates are faster than the experimental value. However, disagreement is within an order of magnitude, although the calculation is the simplest one.

The simple calculation well explains the experimental characteristics that the tunneling rate is not affected by the electric field between 5.0×10^3 and 2.8×10^4 V/cm. In fact, unlike the earlier experiments,[2,3] the condition $eFd \ll U-E$ holds in our experiment. Nevertheless, excitons are stripped of electrons and electrons tunnel across barrier potentials critically at the field $F_t \sim 5.0\times10^3$ V/cm, because the field gain can compensate for the binding energy of excitons. It is not easy to consider the tunneling of electrons through both the Coulomb barrier and the barrier potential. In fact, there is no available theoretical study of this problem. The slow-decay component observed between $\sim 5.6\times10^3$ and $\sim 2.8\times10^4$ V/cm may be due to nongeminate excitons made of stripped electrons and holes. With

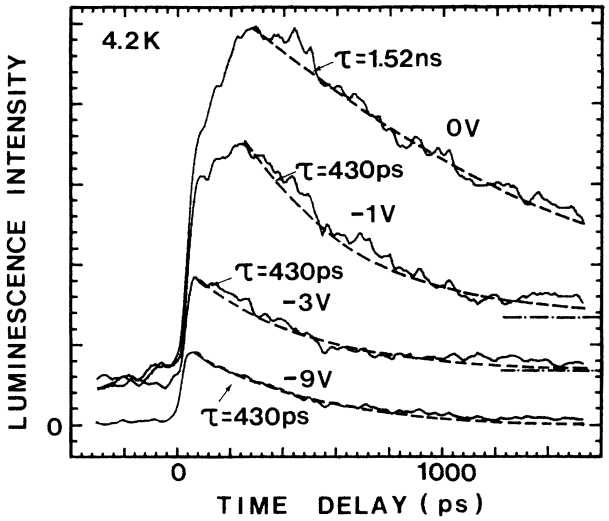

FIG. 3. Temporal profile of the heavy-exciton luminescence intensity in the perpendicular electric field. The estimated electric fields in the MQW are 5.0×10^3, 5.6×10^3, 9.8×10^3, and 2.8×10^4 V/cm for the applied voltages of 0, −1, −3, and −9 V, respectively. Dashed lines show exponential decay with respective time constant τ. Dash-dotted lines show long-lived component. The background observed before 0 ps comes from the stray of 6-ns-lived luminescence which is peculiar to the synchroscan streak camera.

the increase in the electric field, the slow-decay component decreases and the photocurrent increases. We may attribute this change to the onset of successive tunneling of electrons through many barriers. However, these processes remain to be clarified by future study.

In summary, the tunneling dynamics of photogenerated carriers in GaAs-$Al_{0.29}Ga_{0.71}As$ multiple-quantum-well structures have been studied in an electric field perpendicular to the well layers. Drastic changes, such as an increase in the photocurrent, a decrease in the exciton luminescence, and a change of the exciton lifetime take place simultaneously at the critical electric field of $\sim 5.0\times 10^3$ V/cm. The changes are ascribed to the dissociation of excitons and the tunneling of electrons through the $Al_{0.29}Ga_{0.71}As$ potential barriers. These processes take place when the field gain can compensate for the exciton binding energy. The tunneling rate of the electrons is determined to be 1/(430 ps), slower than the simple estimation.

Note added. A recent work by Polland *et al.* [Phys. Rev. Lett. **55**, 2610 (1985)] covers similar ground treated in the present work. Contrary to the present results, they have observed that the lifetime of excitons is prolonged with an increase in the electric field. The contradiction probably comes from a difference in the quantum-well structures, especially with respect to the thickness of barriers.

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[1]L. L. Chang, L. Esaki, and R. Tsu, Appl. Phys. Lett. **24**, 593 (1974).

[2]E. E. Mendez, G. Bastard, L. L. Chang, L. Esaki, H. Morkoc, and R. Fisher, Phys. Rev. B **26**, 7101 (1982).

[3]J. A. Kash, E. E. Mendez, and H. Morkoc, Appl. Phys. Lett. **46**, 173 (1985).

[4]Y. Horikoshi, A. Fischer, and K. Ploog, Phys. Rev. B **31**, 7859 (1985).

[5]H. Iwamura, T. Saku, and H. Okamoto, Jpn. J. Appl. Phys. **24**, 104 (1985).

[6]H. C. Casey, Jr. and M. B. Panish, *Heterostructure Lasers* (Academic, New York, 1978), Part A, Chap. 4.

[7]G. Bastard, E. E. Mendez, L. L. Chang, and L. Esaki, Phys. Rev. B **28**, 3241 (1983).

[8]D. A. B. Miller, D. S. Chemla, T. C. Damen, A. C. Gossard, W. Wiegmann, T. H. Wood, and C. A. Burrus, Phys. Rev. Lett. **53**, 2173 (1984); Phys. Rev. B **32**, 1043 (1985).

[9]S. Tarucha, H. Okamoto, Y. Iwasa, and N. Miura, Solid State Commun. **52**, 815 (1984).

[10]L. D. Landau and E. M. Lifshitz, *Quantum Mechanics (Non-relativistic Theory)*, 3rd ed. (Pergamon, New York, 1977), Chap. 10.

[11]D. Bohm, *Quantum Theory* (Prentice-Hall, Englewood Cliffs, 1951), Chap. 12.

[12]R. Dingle, in *Festkörperprobleme,* edited by H. J. Queisser, Advances in Solid State Physics, Vol. XV (Pergamon, New York, 1975), p. 21.

[13]R. C. Miller, D. A. Kleinman, and A. C. Gossard, Phys. Rev. B **29**, 7085 (1984).