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 $1/(430\, \text{ps})$ 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. 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$_{0.29}$Ga$_{0.71}$As MQW in an electric field. It has been pointed out that these phenomena are possibly correlated with the yield-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. The main part consists of 100 alternation 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 $p^+$-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 ($\chi$), 1.88 eV ($\Gamma$), and 1.519 eV ($\Gamma$), respectively. 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 synchroskan 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 photocconductivity 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 photocconductivity 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, $\Delta E$, is proportional to the square of the electric field $F$ as follows:

$$\Delta E = -2.135 \times 10^{-3} (m_e^* + m_h^*) \frac{e^2 F^2 L^4}{\hbar^2},$$  

(1)

where $e$ is the charge of electron, the $z$ axis is along the superlattice direction, $L_z$ is the well layer thickness, and $m_e^* (= 0.665 m_0)$ and $m_h^* (= 0.45 m_0)$ 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 $\Delta E$ by an order of magnitude. In fact, Eq. (1) explains the energy shift of the excitons in a strikingly complete manner, as 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 photo-generated carriers 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 $F_i \sim 5.0 \times 10^3$ 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 $F_i \sim 5.0 \times 10^3$ 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 photocconductivity 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, photocconductivity spectra under the externally applied voltage are shown. The lowest-energy peak corresponds to heavy excitons.

**FIG. 2.** Luminescence intensity of heavy excitons (O) 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 $5.0 \times 10^3$, $9.8 \times 10^3$, $2.5 \times 10^4$, and $2.8 \times 10^4$ V/cm, respectively.
The field gain is almost equal to the exciton binding energy in 120-A quantum wells (10 meV) derived from the magneto-optical measurements. The field gain compensates for the binding energy. Therefore, it is quite reasonable for excitons to dissociate at a field of $F_i \approx 5.0 \times 10^3 \text{ V/cm}$. Curves of photocurrent and luminescence intensity show a plateau between $-5.0 \times 10^3$ and $-2.0 \times 10^4 \text{ V/cm}$. With the further increase in the electric field up to $-2.8 \times 10^4 \text{ V/cm}$, the photocurrent increases and the exciton luminescence decreases.

The transient response of the exciton luminescence is shown in Fig. 3. At the field of $F_i \approx 5.0 \times 10^3 \text{ V/cm}$, the temporal profile of the exciton luminescence changes drastically. Below $F_i$, the exciton luminescence exponentially decays with a time constant of $1.52 \text{ ns}$. Above $F_i$, 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 $F_i \approx 5.0 \times 10^3 \text{ V/cm}$. 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 $1/(430 \text{ ps}) = 2.3 \times 10^7 \text{ s}^{-1}$. At a field of $-2.8 \times 10^4 \text{ V/cm}$, 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 $F_i$, the dissociation rate of excitons, $w_1$, is given by the following formula which also describes the ionization rate of hydrogen atoms:

$$w_1 = \frac{16R^2/\pi eF_{ag}(\exp(-4R/3eF_{ag}) - 1)}{2 \pi \hbar^2/2m^*_{\text{hh}}L^2} \exp(-2\sqrt{2m^*_{\text{hh}}(U-E)/eF_{ag}}),$$

where $R(=4.2 \text{ meV})$ is the Rydberg energy and $a_{\text{B}}(=136 \text{ Å})$ is the exciton Bohr radius. The dissociation rate reaches $2.8 \times 10^{13} \text{ s}^{-1}$ at an electric field of $F_i \approx 5.0 \times 10^3 \text{ V/cm}$. 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 $A_{0.25 \text{GaAs}}$ 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

$$w_2 = \frac{\pi \hbar^2/2m^*_{\text{hh}}L^2}{2 \pi \hbar^2/2m^*_{\text{hh}}L^2} \exp(-2\sqrt{2m^*_{\text{hh}}(U-E)/eF_{ag}}),$$

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 $d_{\text{B}} (=58 \text{ Å})$ is the barrier thickness. The factor $2m^*_{\text{hh}}L^2/\pi \hbar^2$ is the classical period of the electron (heavy-hole) motion in the quantum well, because the $z$ component of the velocity of electrons (heavy holes) in the lowest sublevel is $\hbar/k/m^*_{\text{hh}}L$. 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 $85\text{ meV}$ is correct, the conduction-band discontinuity $E_c$ is $307 \text{ meV}$ and the valence-band discontinuity $E_v$ is $54 \text{ meV}$. Then, the tunneling rate of electrons is $3.9 \times 10^9 \text{ s}^{-1}$ and that of holes is $2.7 \times 10^8 \text{ s}^{-1}$. On the other hand, the tunneling rate of electrons is $1.8 \times 10^9 \text{ s}^{-1}$ and that of holes is $4.3 \times 10^8 \text{ s}^{-1}$. If the band-gap discontinuity split of $57\text{ meV}$ ($E_c = 206 \text{ meV}$, $E_v = 155 \text{ meV}$) is correct. 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 $5.0 \times 10^3$ and $2.8 \times 10^4 \text{ V/cm}$. In fact, unlike the earlier experiments, 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 $F_i \approx 5.0 \times 10^3 \text{ V/cm}$, because the field gain can compensate for the binding energy of excitons. It is not easy to properly 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 $-5.6 \times 10^3$ and $-2.8 \times 10^4 \text{ V/cm}$ may be due to nonequilibrium excitons made of stripped electrons and holes.
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_{0.25}Ga_{0.75}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_{0.25}Ga_{0.75}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 $1/(430 \text{ ps})$, 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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