Observation of resonant tunneling through single self-assembled InAs quantum dots using electrophotoluminescence spectroscopy

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The resonant tunneling through single InAs quantum dots embedded in an n-GaAs/i-$Al_{0.38}Ga_{0.62}As$/n-GaAs diode has been studied by using microscopic electrophotoluminescence spectroscopy. Many sharp luminescence lines which originated from single quantum dots were observed by injecting resonant electrons from the emitter to the dots. Bias dependence of a single luminescence line was investigated. The peak intensity showed triangular dependence which was similar to the current–voltage characteristics of electron resonant tunneling from three dimension to zero dimension. When the bias voltage was increased, the peak energy slightly shifted to a lower energy indicating the existence of Stark effect, and the linewidth slightly increased. The higher the luminescence energy was, the broader the linewidth was. This result agrees with the calculated resonant level width. The lifetime of resonant states was estimated to be 2.4–27 ps for luminescence linewidth of 250–22 μeV. [S0021-8979(00)04009-3]

I. INTRODUCTION

Resonant tunneling phenomenon in double barrier semiconductor heterostructures has received much interest because of its importance for both fundamental physics and ultra-high-speed electronics, since it was proposed by Tsu *et al.*[1–4] These studies have treated resonant tunneling via two-dimensional bound states which are formed by conventional epitaxial methods. Recently, it has become possible to fabricate high-quality quantum dots (QDs) with zero-dimensional bound states by using self-assembled phenomenon in strained layer epitaxy.[5–10] Resonant tunneling through the zero-dimensional bound states of such self- assembled QDs has also been attracting much interest from a physics point of view. There are some reports discussing the electric characteristics of resonant tunneling diodes in which self-assembled InAs QDs are sandwiched by AlGaAs or AlAs barriers.[11–14] The current peaks due to three- to zero-dimensional (3D–0D) tunneling[11] and spin splitting under magnetic field[12–14] have been observed in the current-voltage ($I-V$) characteristics.

In order to investigate carrier transport behavior in the QDs of the diode, we employed a novel approach, microscopic electrophotoluminescence (μ-EPL) spectroscopy, as a way to detect resonant tunneling through single QDs. It measures microscopic photoluminescence (μ-PL) under bias voltage. This article is organized as follows. Section II describes the device structure and experimental details of the μ-EPL spectroscopy. The experimental results are discussed in Sec. III. The bias dependence of EPL spectrum at wide energy range is shown. Finally, focusing on a single luminescence line, the bias dependence of the peak intensity, peak energy, and linewidth are discussed.

II. DEVICE STRUCTURE AND EXPERIMENTAL DETAILS

The device used in this study is a single-barrier n-GaAs/i-$Al_{0.38}Ga_{0.62}As$/n-GaAs tunneling diode, where InAs self-assembled QDs were embedded at the center of the barrier. The epitaxial layer structure is shown in Table I. It was grown by MBE on a (100)-oriented n^+-GaAs substrate ($n^+ = 5\times10^{18}$ cm^{-3}). The growth rates were 0.8 and 0.04 ML/s for GaAs and InAs, respectively. The arsenic-beam equivalent pressure was 1.4×10^{-5} Torr. The growth temperature was 530 °C for the resonant tunneling structure between bottom and top spacer layers and 630 °C for the other layers. After the deposition of 1.8 ML InAs, a growth interruption of about 40 s was included to form self-assembled InAs QDs by the Stranski–Krastanov mode. The formation was confirmed by *in situ* refractive high-energy electron diffraction (RHEED) observation. The RHEED pattern changed from streaky to spotty during the growth interruption.

To characterize the QDs, atomic force microscopy (AFM) and PL were used. AFM imaging was performed on the sample whose growth was terminated after depositing the InAs layer. It allowed us to estimate the height and diameter of QDs as 1.5–3.0 and 20–40 nm, respectively, and the density as $1.0-1.6\times10^{11}$ cm^{-2}. A sample for PL spectroscopy was grown under the same condition as our QD tunneling device but has a thicker $Al_{0.35}Ga_{0.65}As$ barrier (100 nm). The macroscopic PL spectrum is shown in Fig. 1. The spectrum exhibits two structures; a broad line from 1.4 to 1.7 eV with a maximum at 1.6 eV and relatively narrow peak at 1.8 eV. The former and latter are attributed to InAs QDs and wetting layer, respectively. Because the QD emission line overlaps GaAs band gap energy and has a maximum about 100 meV above, it would be predicted that many QDs come into resonance by applying bias voltage.

TABLE I. Epitaxial layer structure.

Layer name	Composition	Doping (cm^{-3})	Thickness (nm)
Collector contact	n^+-$In_{0.6}Ga_{0.4}As$	1×10^{19}	30
	n^+-$In_xGa_{1-x}As$	1×10^{19}	50
	n^+-GaAs	2×10^{19}	20
Collector	*n*-GaAs	1×10^{17}	200
Spacer	*i*-GaAs	—	10
Barrier	*i*-$Al_{0.38}Ga_{0.62}As$	—	7
Quantum dots	*i*-GaAs	—	1.8 ML
Barrier	*i*-$Al_{0.38}Ga_{0.62}As$	—	7
Spacer	*i*-GaAs	—	10
Emitter	*n*-GaAs	1×10^{17}	200
Emitter contact	n^+-GaAs	5×10^{18}	400
Substrate	n^+-GaAs	5×10^{18}	

The device was fabricated by conventional photolithography, wet etching, metal evaporation, and lift-off processes. The Ti/Au collector electrode was evaporated on the top n^+-InGaAs to form a nonalloyed ohmic contact having a window for luminescence measurement. The device area is 100×100 μm^2.

The diode was mounted on a cryostat with a window for luminescence measurement. The EPL spectroscopy is based on the measurement of the photoluminescence from the *n*-*i*-*n* diode under bias voltage. Minority holes were excited by the 514.5 nm line of an Ar^+ laser. The laser light was focused on the device surface by an objective lens. The μ-EPL signal was collected by the objective lens, dispersed with a 1 m double monochromator, and detected with a liquid-nitrogen-cooled Si charge coupled device. The measurement was performed at 6 K. The energy resolution of the spectrometer was estimated to be 20–30 μeV.

III. RESULTS AND DISCUSSION

The EPL spectra were measured in a wide energy region from 1.50 to 1.75 eV, focusing the excitation laser with 20 μm diameter on the sample surface at an excitation power density of $\approx$3 W/cm^2. Such quasimicroscopic spectroscopy is useful for investigating the spectral distribution due to inhomogeneity of QDs. Figure 2 shows the EPL spectra at various bias voltages (V_{bias}) of 0–1.0 V, where electrons flow from substrate to the top collector layer. No luminescence was obtained at $V_{bias}=0$ V except GaAs band edge emission at 1.51 eV. When V_{bias} was larger than 0.1 V, on the other hand, many sharp luminescence lines with linewidths of 22–250 μeV were observed, indicating carriers were injected into QDs by applying bias voltage. Even though no structure corresponding to the resonant tunneling was observed in the *I*–*V* characteristics of the diode because of the large number of QDs ($\approx10^7$), we could obtain clear luminescence signal corresponding to the resonant tunneling of electrons via single InAs QDs. This is because the issue of inhomogeneous broadening in observing the peak of *I*–*V* characteristics can be overcome by using the EPL technique with high energy resolution. This demonstrates the advantage of the present EPL technique.

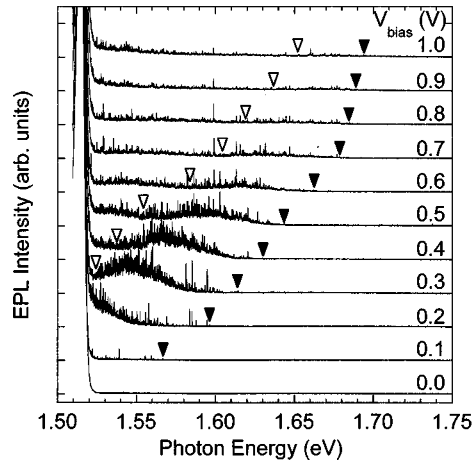

FIG. 2. Bias voltage dependence of EPL spectrum in wide energy range from 1.50 to 1.75 eV. The luminescence group between the white and black arrows corresponds to QDs resonating with electron accumulation layer of emitter.

Figure 3 shows the schematic band diagram of the sample under bias voltage. At zero bias, only a small number of electrons and holes photoexcited in the thin AlGaAs barrier can drop into the InAs QDs, resulting in little luminescence from the QDs. Under bias voltage, on the other hand, holes excited in the collector layer drift to the AlGaAs barrier due to electric field and tunnel into the QDs. Electrons

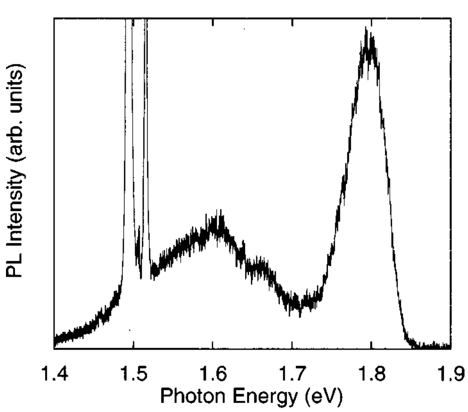

FIG. 1. PL spectrum of InAs QDs buried in a 100 nm AlGaAs barrier. The luminescence with energy of 1.4–1.7 eV originates from QDs.

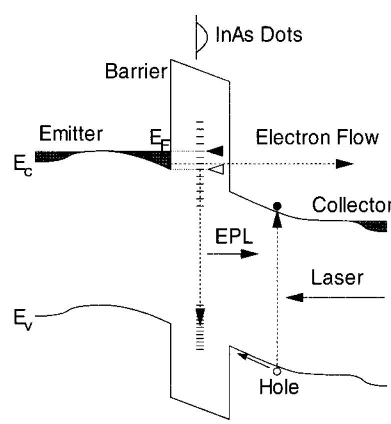

FIG. 3. Schematic band diagram of *n*-GaAs/*i*-AlGaAs/*n*-GaAs tunneling diode under bias voltage. InAs QDs are embedded in the *i*-AlGaAs tunnel barrier. The EPL mechanism is also shown.

resonantly tunnel into the QDs from the emitter accumulation layer. The electrons and holes in the QDs recombine and radiate photons.

It should be pointed out that a group of luminescence lines between the black and white arrows shown in Fig. 2 shifts to a higher-energy region with increasing V_{bias}, although individual luminescence lines show small redshift as will be described later. The black and white arrows are there as a guide for the highest and lowest energies of the group of the EPL lines. The blueshift of the group of luminescence lines can be explained by referring to the energy band diagram of the *n-i-n* diode with InAs QDs shown in Fig. 3. QDs on resonance with electrons in the emitter accumulation layer change from large QDs with low-energy quantum level to small QDs with high-energy quantum level with increasing V_{bias}. The black arrows in Fig. 2 correspond to the luminescence lines originating from the QDs whose electronic states align with electrons with the highest energy in the emitter. At zero temperature, the position of the black arrow corresponds to the emitter Fermi energy (E_F) as shown in Fig. 3. The white arrows correspond to the luminescence lines originating from the QDs, which align with the emitter conduction band edge E_C. Each QD comes into resonance successively by changing V_{bias}. This means that it is possible to select QDs on resonance by choosing V_{bias}. The Fermi energy of emitter triangular potential is estimated to be about 90 meV at V_{bias}=0.4 V from the energy separation between the black and white arrows. When V_{bias} is larger than 0.7 V, the position of the black arrow in Fig. 2 does not change beyond 1.68–1.70 eV in spite of the increase in V_{bias}. This probably reflects the fact that there are little InAs QDs with energy higher than the position of the black arrows.

Many luminescence lines were also observed in the energy regime lower than the white arrows as shown in Fig. 2. The QDs corresponding to these luminescence lines were at off-resonance because their quantum levels were pulled down below the emitter E_C by applying large V_{bias}. Even though the details are not clear, some electrons might be injected into the QDs by phonon-assisted tunneling.[15] Another possibility is that electrons tunneled into higher excited states of QDs then relaxed to ground states of QDs. These luminescence lines suggest the existence of the sequential tunneling process. Further study will be necessary to clarify this point.

In order to analyze resonant tunneling through a single QD, the observation area was minimized to 2 μm by focusing the excitation laser. Figure 4 shows the V_{bias} dependence of μ-EPL spectra at a narrow energy range of 1.645–1.660 eV. Several isolated luminescence lines corresponding to single QDs that were on resonance were obtained at V_{bias} of 0.4–0.9 V. This indicates that it is possible to study resonant tunneling through a QD by observing a single EPL line. Focusing on a single luminescence line at an energy of 1.6527 eV, the V_{bias} dependences of the peak intensity, peak energy, and linewidth are summarized in Fig. 5.

The EPL intensity gradually decreased after showing a maximum at 610 mV as shown in Fig. 5(a). The behavior can be understood if we consider the 3D–0D electron tunneling. The luminescence intensity is dependent on the number of carriers injected into the QDs per unit time. Because there are many quantum levels in the valence band of a QD, holes can tunnel into the QD and rapidly relax to the ground state. This means that the number of holes in the QD ground state would be regarded as independent of bias voltage. On the other hand, the number of electrons injected into the QD strongly depend on bias voltage because each quantum level of electrons is separated with sufficient energy due to light electron mass. Therefore, the bias dependence of EPL intensity would be expected to be determined by the bias depen-

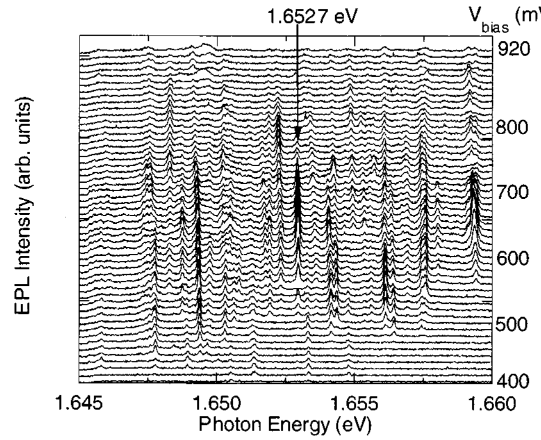

FIG. 4. Bias voltage dependence of μ-EPL spectrum in narrow energy range from 1.645 to 1.660 eV. Several isolated luminescence lines corresponding to single QDs at resonance are observed.

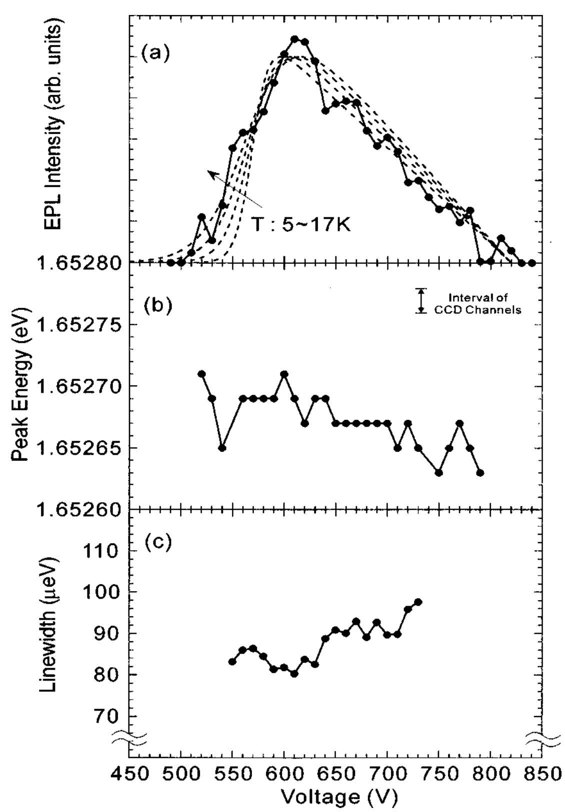

FIG. 5. Bias voltage dependences of (a) peak intensity, (b) peak energy, and (c) linewidth of a single EPL line of 1.6527 eV. The calculated I–V characteristics of 3D–0D resonant tunneling are also shown by broken line in (a).

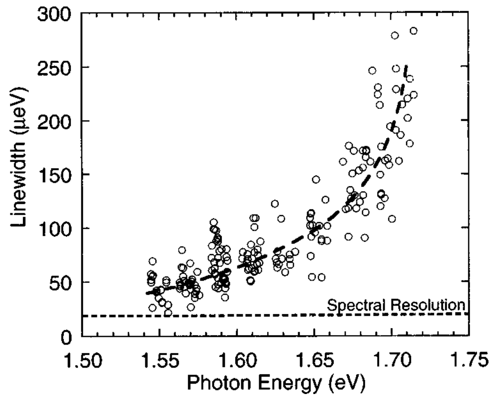

FIG. 6. Linewidth of various EPL lines vs EPL peak energy. The width is broader for the EPL line with higher energy.

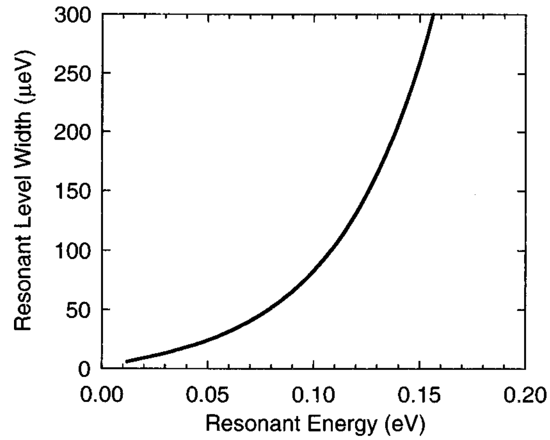

FIG. 7. Resonant energy dependence of resonant level width calculated for one- dimensional resonant tunneling structures. Resonant energy of the horizontal axis is the difference between band edge of emitter and resonant level.

dence of resonant tunneling current of electrons. The *I–V* characteristics in resonant tunneling via single QD are expressed as follows assuming 3D–0D tunneling:[11,16]

$$I \propto \frac{V_{\mathrm{EC}} - V}{1 + \exp\left[\frac{q\alpha}{kT}(V_{\mathrm{EF}} - V)\right]} \quad \text{for } V \leqslant V_{\mathrm{EC}}$$

$$I = 0 \quad \text{for } V > V_{\mathrm{EC}}, \tag{1}$$

where V_{EC} and V_{EF} are the bias voltages when the E_C and E_F of the emitter align with the quantum level of the QD, respectively, α is the leverage factor, T the absolute temperature, k the Boltzmann constant, and q the electron charge. The *I–V* characteristics calculated by Eq. (1) are shown by a broken line in Fig. 5(a) for various T from 5 to 17 K with a step of 4 K. The values used in the calculation were $V_{\mathrm{EC}} = 0.57$ V and $V_{\mathrm{EF}} = 0.82$ V. α was estimated to be 0.08–0.06 at V_{bias} of 450–850 mV by solving Poisson's equation. The V_{bias} dependence of the EPL intensity is in good agreement with the calculated *I–V* characteristics at 13–17 K, indicating that EPL intensity reflects resonant tunneling current in the 3D–0D system as discussed above. Even though these temperatures are a little higher than the thermocouple temperature of 6 K, it is reasonable if we consider that the sample was mounted on a cold finger of a cryostat.

The peak energy slightly shifted to a lower energy with increasing V_{bias} as shown in Fig. 5(b), suggesting the existence of the quantum-confined Stark effect. The redshift confirms that the luminescence originate from electron–hole recombination in the dots. The amount of redshift was as small as about 50 μeV because the carriers were confined in a very small QD. Present results agree with the analysis which was recently reported by Raymond *et al.*[17]

The V_{bias} dependence of EPL linewidth (Γ_{EPL}) is shown in Fig. 5(c). The Γ_{EPL} increases slightly from 80 to 100 μeV with the increase in V_{bias}. In order to elucidate the dominant factor contributing to Γ_{EPL}, the linewidths of EPL lines of various luminescence energies were measured. The results are shown in Fig. 6. The broken line is a guide for the eyes. Γ_{EPL} is broader for the EPL line with higher energy. This suggests the contribution of resonant coupling in Γ_{EPL}. The widths of resonant levels (Γ_{res}) were calculated for one-dimensional GaAs/AlGaAs/InAs/AlGaAs/GaAs double barrier resonant tunneling structures with different well width, assuming a flatband condition and 300 meV conduction-band offset at GaAs/AlGaAs interface. The calculated results are shown in Fig. 7 as a function of resonant energy. Here, the resonant energy is the difference between the conduction band edge of emitter and resonant level. Γ_{res} is broader for higher resonant level because the effective barrier height is lower for higher resonant level, leading to enhanced coupling between the quantum well and contact layers. We can see good agreement between Figs. 6 and 7, suggesting that the resonant coupling between QD and emitter/collector layers plays an important role in EPL linewidth broadening and its dependence on the collector voltage. If we assume Γ_{EPL} represents the inverse lifetime of the resonant state $(=\hbar/\Gamma_{\mathrm{res}})$, the lifetime of the resonant state is estimated to be 2.4–27 ps for Γ_{EPL} of 250–22 μeV. If we take into account the effect of exciton formation, the lifetime will be longer. The dispersion of linewidth in Fig. 6 is not small. This cannot be explained solely by the resonant width. Even though the reason has not been clear yet, exciton migration between neighboring QDs and the extinction due to transition to the localized states associated with QD surface or defects[18] might be responsible for the linewidth dispersion. Further study is necessary to clarify this point.

IV. SUMMARY

The resonant tunneling through a single InAs self-assembled QD was studied by using μ-EPL spectroscopy. A group of sharp luminescence lines originating from QDs at resonance was observed. The 3D–0D tunneling and Stark effect were confirmed by investigating the bias dependence of the intensity and energy of the peak of a single QD. The linewidth slightly increased with increase in bias voltage, and EPL linewidths were broader for the lines with higher energy. These behaviors are explained by the energy width of resonant level. The lifetime of resonant level was estimated to be 2.4–27 ps for EPL lines at 250–22 μeV.

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