Enhanced Binding Energy of One-Dimensional Excitons in Quantum Wires

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Photoluminescence spectra are systematically studied on a series of 5 nm scale, T-shaped, GaAs quantum wires (QWRs). Potential profiles and quantized energies of QWRs are precisely determined as a function of structure parameters. By comparing the experimental quantized energy with the calculation without the Coulomb interaction, the binding energy E_b of one-dimensional (1D) excitons is quantitatively evaluated. Upon increasing the 1D confinement, E_b in QWRs is found to be enhanced to 27 $\pm$ 3 meV, which is 6–7 times larger than the bulk value.

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The Coulomb interaction of electrons confined in quantum structures realizes novel correlated states in condensed-matter physics. Since quantum confinement forces electrons to interact strongly, the binding energy E_b of two-dimensional (2D) excitons in quantum wells (QWs) has been shown to increase, while reducing the well thickness, up to 4 times as large as the bulk value (4.2 meV in GaAs) [1–5], leading to the room temperature excitonic absorption [6]. In quantum wires (QWRs), the excitonic effect is potentially further enhanced by laterally compressing wave functions into the nanometer-scale regime [7]. The recent epitaxial growth technology makes it possible to fabricate high-quality 5 nm scale GaAs QWRs [8], enabling one to study the enhanced Coulomb interaction in one-dimensional (1D) systems.

In this work, the enhanced E_b of 1D excitons has been systematically studied in a series of 5 nm scale GaAs QWRs. As a function of structure parameters, the quantized energy of QWRs is precisely determined via photoluminescence (PL). By comparing the measured quantized energy with the calculation, E_b of 1D excitons is quantitatively evaluated. We have found that by increasing the 1D confinement E_b is enhanced to 27 $\pm$ 3 meV, which is 6–7 times larger than the bulk value.

Employing the modified molecular beam epitaxy process, the cleaved-edge overgrowth method [9], we have fabricated two series (S1 and S2) of T-shaped QWRs (T-QWRs) [10,11], whose cross section is schematically shown in the inset of Fig. 1. In the first growth, multi-QWs (QW1) with thickness d_1 were formed on a (001) GaAs substrate. After cleavage, a GaAs QW layer (QW2) with thickness d_2 was deposited in the second growth onto a cleaved (110) surface of the multi-QW structure. Samples in series S1 have $Al_{0.3}Ga_{0.7}As$ barriers and the QW1 thickness $d_1 = 5.4$ nm, while those in series S2 have AlAs barriers and $d_1 = 5.3$ nm [12]. Since QW1 in each series was grown simultaneously, the QW1 thickness d_1 is constant, whereas the QW2 thickness d_2 is changed around d_1.

PL spectra of these samples were measured via the (110) surface in the backward scattering geometry. We studied PL from S1 at 4 K using a He-Ne laser and PL from S2 at 8 K using an Ar^+ laser as an excitation source. Typical PL spectra of series S1 and S2 are shown in Figs. 1(a) and 1(b), respectively, for three samples having the different thickness d_2 of QW2. Three PL peaks are clearly seen by the spatially resolved PL measurement, and found to come from QWR, QW1, and

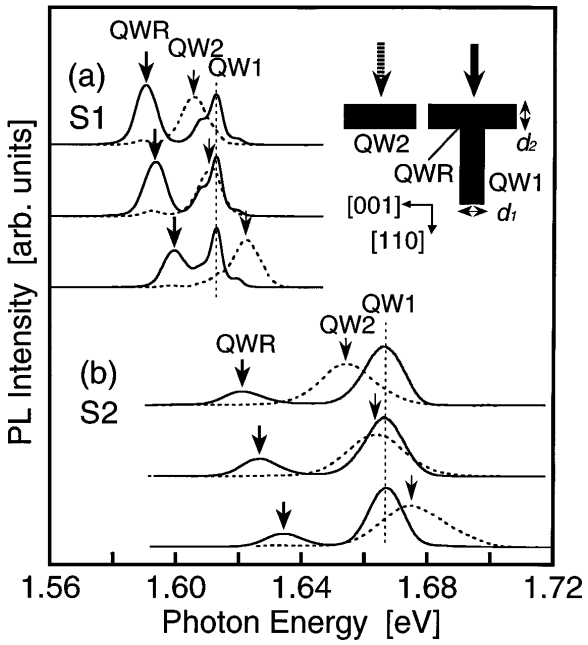

FIG. 1. Typical spatially resolved photoluminescence spectra from QWs (QW1 and QW2) and QWRs of two series of samples with various thicknesses d_2 of QW2. The data for series S1 are given in (a) and those of series S2 are given in (b). PL spectra shown by dashed curves were measured by exciting the QW2 region, while those shown by solid curves were measured by exciting the T-QWR region. A basic structure of T-QWRs is shown as the inset, where QW1, QW2, and QWR are also defined.

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QW2, as described in the separate papers [8,13]. In such a measurement, PL spectra shown by dashed curves in Fig. 1 were measured by exciting the QW2 region, while those shown by solid curves were measured by exciting the T-QWR region. As a result, we can precisely determine potential profiles and quantized energies of QWRs. All the PL peaks of S1 are sharp with a linewidth of 5-8 meV, indicating the good uniformity of the structures. Although the PL peaks of S2 are broader due to the tighter confinement, PL from QWRs is still sharp with a linewidth of 14 meV.

We determined the energies of PL peaks from 20 samples in series S1 and 11 samples in series S2. They are plotted in Fig. 2 against the QW2 thickness d_2, in which PL energies of T-QWR are shown by solid circles, those of QW1 by blank squares, and those of QW2 by blank circles. Here, the QW2 thickness d_2 is not nominal thickness, but is determined by comparing the observed PL peak of QW2 with the theoretical value; for this purpose, we calculated the energy level of QW2 under the effective mass approximation, and the result is shown by broken curves in Fig. 2. Here, we used the following conventional parameters: the electron effective mass of $0.067m_0$, the hole effective mass in the (001) QWs of $0.38m_0$, the hole effective mass in the (110) QWs of $0.71m_0$, the GaAs band gap with exciton binding energy of 1.519 eV, and the conduction (valence) band offset of 0.243 (0.131) eV for $Al_{0.3}Ga_{0.7}As$ and 1.036 (0.558) eV for AlAs, respectively [14]. It has been empirically shown that the calculation of energy levels using this

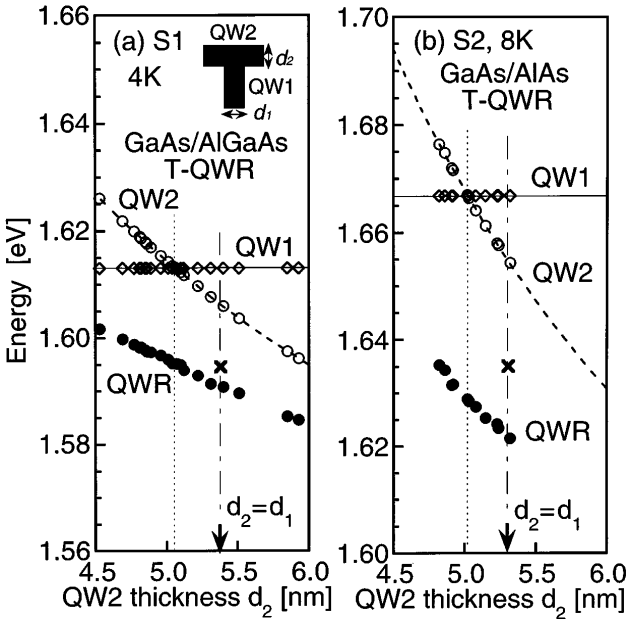

FIG. 2. The energies of PL peaks are shown by solid circles for QWRs, by blank squares for QW1, and by blank circles for QW2 in S1 (a) and S2 (b) as a function of the QW2 thickness d_2. The broken curves show the energy level of QW2 calculated under the effective mass approximation. The dotted lines indicate the energy level when the levels of QW2 and QW1 are identical. The dash-dotted lines indicate the identical thickness d_2 with d_1.

set of parameters and its comparison with PL energy allow us to estimate d_2 accurately with an error less than one monolayer as long as d_2 is in the range of 4-9 nm. Later, we will discuss the possibility and effects of selecting other sets of material parameters [15,16]. In this way, we have precisely determined the dependence of the energy level of T-QWRs on the QW2 thickness d_2.

We should first point out in Fig. 2 that the energy level of T-QWRs increases with reducing d_2. Note that the energy level of T-QWRs approaches the energy level of QW1 as d_2 decreases, while it approaches that of QW2 when d_2 increases. This tendency is reasonable, because the T-QWR state converges into QW1 or QW2 in the limit of thin QW2 ($d_2 \ll d_1$) or thick QW2 ($d_2 \gg d_1$), respectively.

The slope of the energy level of T-QWRs and that of QW2 against d_2 give the energy broadening induced by the monolayer (ML) fluctuation of the QW2 thickness d_2. The slope of T-QWRs is smaller than that of QW2 with the same thickness d_2, showing that T-QWRs are less affected by d_2 than QW2. Fitting the data points with a linear relation, we obtain the broadening energy of T-QWRs at $d_2 = d_1$ to be 3.4 and 7.0 meV/ML (1 ML = 0.283 nm) for S1 and S2, respectively. These two values give the reasonable estimation that the observed PL linewidth of T-QWRs corresponds to about 2 ML fluctuation of d_2.

In Fig. 2, one can easily evaluate the spacing between PL energy of T-QWRs and that of QW1 and QW2. We denote this spacing as the effective lateral confinement energy of excitons ($E^*_{1D\text{-}2D}$) [13]. Note that $E^*_{1D\text{-}2D}$ is the energy difference between the 1D exciton state in QWRs and the 2D exciton state in neighboring QWs and is, therefore, a key parameter to represent the stability and the 1D nature of the excitons confined in T-QWRs. It is clear from Fig. 2 that $E^*_{1D\text{-}2D}$ reaches a maximum when the energy levels of QW1 and QW2 coincide, reaching as large as 18 meV for S1 and 38 meV for S2.

In the following, we analyze these data quantitatively to estimate the binding energy E_b of 1D excitons in T-QWRs. For simplicity, hereafter we examine T-QWRs formed by two QWs with identical well thickness ($d_2 = d_1$), which we call balanced T-QWRs. Note that balanced T-QWRs are best suited to confine electrons efficiently. For the tighter confinement of holes, however, it is better to set d_2 (the QW2 thickness) slightly smaller than d_1 (the QW1 thickness); that is, to compensate the anisotropy of the hole effective mass, which is heavier in QW2 than in QW1. These facts simplify the evaluation of the energy of balanced T-QWRs, as we discuss below.

To calculate the energy levels of T-QWRs, we evaluate first the lateral confinement energy $E_{1D\text{-}2D}$ of electrons, the energy difference between T-QWRs and QWs, by neglecting the Coulomb interaction and by using the single band effective mass approximation [17]. The lateral confinement energy of holes is estimated to be small (1-2 meV) [17] in the balanced T-QWRs due to the

heavy and anisotropic hole mass. Thus, we can obtain the energy level of T-QWRs by simply subtracting $E_{1D\text{-}2D}$ of electrons from the energy level of QW2. The energies thus calculated neglecting the Coulomb interaction are denoted by two crosses in Fig. 2. Note that the measured PL energy is lower than the calculated energy by 4 meV in S1 and by 14 meV in S2. These discrepancies should be mostly attributed to the additional enhancement of the Coulomb effect of 1D excitons due to the lateral confinement.

In evaluation of the enhancement of E_b in balanced T-QWRs, we should estimate that experimental errors caused by the broadening of PL spectra are about ± 2 meV and theoretical errors caused by the neglected contribution of holes are -1 ± 1 meV . Therefore, we conclude that the additional enhancement of E_b due to the lateral confinement is 3 ± 3 meV for S1 and 13 ± 3 meV for S2. If we assume E_b in 5 nm thick GaAs QWs to be 14 meV as reported [4], the binding energy E_b of 1D excitons in balanced T-QWRs is concluded to be 17 ± 3 meV for S1 and 27 ± 3 meV for S2 [18]. Note that E_b of S2 is about 6–7 times larger than the bulk value. We believe that this is the first clear demonstration of the enhanced Coulomb interaction in semiconductor QWRs with the binding energy far exceeding that of QWs.

Here, we examine the validity of our calculation to evaluate $E_{1D\text{-}2D}$ of electrons on the basis of the present material parameters. It is possible to select other sets of material parameters and/or more sophisticated models, including band nonparabolicity and other effects, for the estimation of well thickness with 1 ML accuracy and for the calculation of $E_{1D\text{-}2D}$. However, the evaluation of $E_{1D\text{-}2D}$ of electrons is not sensitive to those parameter variations. For example, $E_{1D\text{-}2D}$ of electrons in S2 is changed only by less than 1 meV, when the electron mass changes from $0.067m_0$ to $0.070m_0$, the conduction band offset varies from 1.036 to 1.116 eV, or the well thickness is altered from 5.30 to 5.44 nm. Note that effects of the valence-band parameters such as masses, offsets, and band nonparabolicity are counted in the estimation of $E_{1D\text{-}2D}$ of electrons only implicitly through the well thickness, as we have already discussed. Hence, the calculation of $E_{1D\text{-}2D}$ of electrons is sufficiently accurate and the comparison with experiment can be done with a ± 3 meV accuracy.

We should note that the additional enhancement of E_b due to lateral confinement is significant (13 meV) in S2, whereas it is rather small (3 meV) in S1. Since S1 and S2 have almost the same well thickness ($d_1 = d_2 =$ 5.3–5.4 nm), the difference in the enhancement of E_b is mainly due to the barrier height difference, indicating the importance of very tight confinement.

To explain this situation more clearly, we plot in Fig. 3 the lateral confinement energies as a function of the PL energy of QW2. Two solid circles represent the effective lateral confinement energies $E^*_{1D\text{-}2D}$ measured in our balanced T-QWRs of series S1 and S2. For comparison, we have shown by solid or broken lines in Fig. 3 the theoretical lateral confinement energy $E_{1D\text{-}2D}$ of electrons, which is calculated by Pfeiffer *et al.* for the barrier height of 245, 355, 500 meV, and infinity [17,19]. We also show by crosses the theoretical values of $E_{1D\text{-}2D}$ of electrons calculated for our samples. Note in Fig. 3 that the calculated $E_{1D\text{-}2D}$ of electrons increases linearly when the barrier height is infinite ($V_0 = \infty$), but it rises only sublinearly with the QW2 energy when V_0 is finite. The experimental data indicated by solid circles, however, show a superlinear dependence, which indicates that the Coulomb interaction becomes increasingly important.

In the case of infinite barriers, both the energy level of balanced T-QWRs and that of QW2 are scaled by d_2^{-2}. As a result, $E_{1D\text{-}2D}$ of electrons is scaled by the energy level of QW2, which gives the linear dependence as shown in Fig. 3. For finite barriers, a portion of the wave function penetration into barriers becomes larger for QWR than for QW2 in the strong confinement regime, so that the sublinear dependence is obtained. In the weak confinement regime, however, all the energy levels are approximately scaled by the energy level of QW2, since the difference of the wave function penetration into barriers is negligible.

Note in Fig. 3 that our theoretical values for $E_{1D\text{-}2D}$ of electrons represented by two crosses are very close to the solid line. This means that our balanced T-QWR samples are still in the regime where the scaling law should hold well. Thus, the contribution of the confinement energy without the Coulomb effect to $E^*_{1D\text{-}2D}$ should be proportional to the energy level of QW2. Hence, the observed superlinear dependence of $E^*_{1D\text{-}2D}$ is clear evidence of the substantial enhancement of the Coulomb interaction in 1D excitons when they are tightly confined in QWR structures.

We wish to stress that our work is accurate enough to draw a reliable conclusion on the enhancement of E_b.

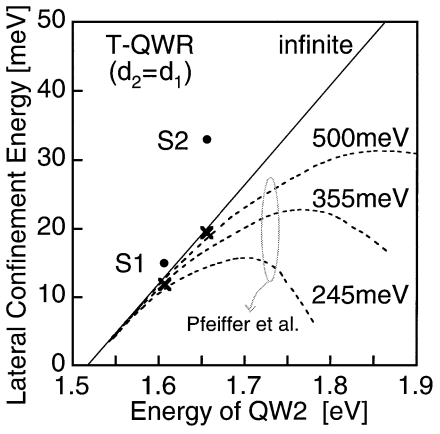

FIG. 3. The lateral confinement energy in T-QWRs formed by two QWs with the identical well thickness is shown as a function of the energy of QW2. The effective lateral confinement energy of excitons (E^*_{1D-2D}) shown by solid circles is compared with the lateral confinement energy $E_{1D\text{-}2D}$ of electrons (the broken or solid lines) for 245, 355, 500 meV, and infinite barriers, calculated by Pfeiffer *et al.* (Refs. [17,19]).

Firstly, $E^{*}_{1D\text{-}2D}$ is determined directly from the measured energies of T-QWRs, QW1, and QW2, and plotted in Fig. 3 as a function of measured PL energy of QW2. Secondly, the evaluation of $E_{1D\text{-}2D}$ of electrons is almost independent of the calculation model, since the scaling law of $E_{1D\text{-}2D}$ of electrons holds well in our T-QWR samples. Therefore, the enhancement of E_b is reliably determined as the difference between $E^{*}_{1D\text{-}2D}$ and $E_{1D\text{-}2D}$ of electrons.

Finally, we comment on previous reports in which the binding energies E_b in several different QWRs were discussed [20–24]. One must note first that E_b of excitons in large QWRs was reported to be almost the same as that of 2D excitons in QWs [20–22]; this is consistent with our results on series S1 of T-QWRs with an $Al_{0.3}Ga_{0.7}As$ barrier. In Refs. [23,24], the enhancement of E_b was determined even when QWRs are larger than our S1 samples. In these reports, however, the reduced diamagnetic shifts of PL were analyzed on the basis of the hydrogen-like exciton model assuming an anisotropic reduced mass. Such an analysis has attributed the squeezing of an exciton wave function to the enhanced Coulomb effect despite the importance of lateral confinement, and has resulted in the overestimation of E_b. We emphasize here that tight 1D confinement comparable with S2 is required to demonstrate the noticeable enhancement of E_b for 1D excitons in semiconductor QWR structures.

In summary, PL spectra have been studied on two series of high-quality 5 nm scale T-QWRs. It is demonstrated that the binding energy E_b of 1D excitons is enhanced by increasing the 1D confinement. Although the enhancement of E_b is small in T-QWRs with $Al_{0.3}Ga_{0.7}As$ barriers, it has been found that the binding energy E_b of excitons in T-QWRs with AlAs barriers is substantially enhanced to as large as 27 ± 3 meV [18]. The evaluated E_b of 1D excitons is far beyond the theoretical limit for 2D excitons in GaAs QWs (17 meV), and becomes 6-7 times larger than the bulk value.

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[18] Since we have directly plotted the PL peak energy in Fig. 2, the corrections caused by Stokes shifts are not included. To precisely evaluate the effective lateral confinement energy, and hence the exciton binding energy, the difference of the Stokes shifts for T-QWRs and adjacent QWs should be taken into account. According to results of our PL excitation spectroscopy so far obtained on similar structures, the Stokes shifts of T-QWRs tend to be smaller than those of adjacent QWs typically by 2 ± 1 meV [H. Akiyama, T. Someya, and H. Sakaki, Phys. Rev. B **53**, 4229 (1996)]. Therefore, the present estimation of 1D exciton binding energy should most likely be further increased by this amount.
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