Large excited state Stokes shift in crescent-shaped AlGaAs/GaAs quantum wires

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The Stokes shifts of the ground and the excited states in a crescent-shaped AlGaAs/GaAs quantum wire (QWR) are investigated using photoluminescence (PL) and PL excitation spectroscopy. The first excited electron to heavy-hole transition showed a Stokes shift (∼17 meV) considerably larger than that of ground state-related transitions (∼4 meV). This is a quite different phenomenon than that observed in two dimensional quantum well structures, and can be explained by the spatial separation of wave functions with different confinement energies in crescent-shaped QWRs.

It is well known that the heterointerface quality has a severe influence on the performance of optical and electronic devices involving semiconductor heterostructures. The heterointerface inhomogeneities of semiconductor quantum structures can usually be detected as a peak energy difference between photoluminescence (PL) and PL excitation (PLE) or optical absorption spectra, i.e., the Stokes shift.1 On the other hand, the information about the excited states of semiconductor quantum structures is very important for devices utilizing excited state transitions, for example the recently developed quantum cascade laser.2 In two dimensional quantum wells (QWLs), the Stokes shifts for quantum states with different confinement energies mainly depend on the variation of confinement energy with well thickness since the wave functions of the ground and the excited states extend over the same heterointerfaces. However, in the cases of quantum wire (QWR) and quantum dot (QD) structures,3 which are objects currently under intensive investigation, the situation could be much different. Most of the QWRs and QDs fabricated by the conventional growth techniques have an irregular cross-sectional shape4,5 which will cause spatial separation of wave functions for the ground and the excited states. This fact means that the wave functions of the ground and the excited states in QWRs and QDs could be exposed to heterointerfaces with different structural quality. In this letter, we show that the spatially separated wave function distribution can result in a large difference in the Stokes shift between the ground and the excited states in crescent-shaped AlGaAs/GaAs QWRs.

The sample used here is a 4.5-nm-thick single QWR grown on a 4.8 μm pitch V-grooved GaAs substrate by metalorganic vapor phase epitaxy (MOVPE) at 630 °C. The GaAs wire layer was grown by flow rate modulation epitaxy (FME), a modified MOVPE growth technique, which was found to have superior low temperature growth selectivity and layer thickness controllability compared with the conventional MOVPE selective growth.5,6 The sample structure has been described in a previous publication.7 Figure 1(a) shows the high resolution transmission electron microscopy (HRTEM) image of a vertically stacked multiple QWR sample which was grown under the same conditions as the sample investigated here.7 Hence, the first QWR in Fig. 1(a) is considered to have approximately the same cross-sectional dimensions as the present sample. From Fig. 1(a), the upper surface of the GaAs wire layer consists of three distinct crystalline facets: one (001) and two (311)A. The (001) heterointerface is almost atomically flat, indicating excellent thickness controllability of the FME technique, whereas the heterointerfaces of the (311)A facets showed thickness fluctuations on the order of 2–3 (311)A monolayers (MLs). We also calculated the electronic state of the QWR by numerically solving the two dimensional single band Schrödinger equation using finite element method. Figure 1(b) shows the calculated electron density probability of the first two electron states. The ground state electron (n = 1) wave function mainly distributes over the (001) facet, while the wave function of the second electron state (n = 2) has a large distribution probability over the (311)A facets. In other words, the wave functions for the ground and the excited states separate spatially in crescent-shaped QWR and consequently will be affected by the heterointerface quality of the (001) and the (311)A facets, respectively.

PL and PLE measurements were used to evaluate the heterointerface quality of the grown QWR sample.6,8,9 Figure 2 shows two PL spectra measured at 12 K with different excitation power densities. In both these cases, luminescence from QWR at about 1.6466 eV dominates the spectra and those from (111)A side wall QW (∼1.83 eV) and vertical QW (VQW) (∼1.87 eV) are very weak. At low excitation power density, only a single peak corresponding to the ground state electron to heavy-hole (1e−1hh) recombination was observed. When the excitation power density is increased to about 30 W/cm², emission peaks from excited states become observable. As shown in the inset of Fig. 2, the QWR spectrum at the energy range of 1.58–1.74 eV can be separated into three Gaussian-shaped peaks. We attribute the peaks at 1.6591 and 1.6775 eV to the recombination between the ground state electron and the first light-hole state (1e−1lh) and to that between the second electron and the second heavy-hole state (2e−2hh), respectively. We also performed polarization dependent PL measurements by which the relative intensity of the 1.6591 eV peak to the 1.6466 and the 1.6775 eV peaks in the case of parallel (to the wire axis) polarization was found to be much weaker than

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the case of perpendicular polarization. This result also confirms that the 1.6591 and the 1.6775 eV peaks are mainly of light-hole and heavy-hole character, respectively.

The excited state structures were further investigated by PLE measurements. Figure 3 shows the PLE spectra measured at 20 K, where a PL spectrum (thin solid line) measured with Ti:sapphire laser (wavelength=710 nm) was also given for reference. With the laser beam polarized parallel to the wire axis (bold solid line), we can observe very sharp PLE peaks related to heavy-hole states, characteristic of the sawtooth-shaped density of states of QWR structures. The three strong PLE peaks observed in the measured energy range can be attributed to transitions between electrons and heavy-holes with the same quantum numbers, that is, the peak at 1.6521 eV to $1^e-1hh$, the peak at 1.6986 eV to $2^e-2hh$ and the peak at 1.7200 eV to $3^e-3hh$. On the other hand, well-resolved PLE structures related to light-holes are observed when the laser beam was polarized perpendicular to the wire axis (dashed line). The peaks at 1.6668 and 1.6764 eV are attributed to the transitions due to the ground electron state with the first and the second light-hole states ($1^e-1lh$ and $1^e-2lh$), respectively from their strong polarization dependence. A weak shoulder at about 1.6594 eV was also observed at both polarizations which could be attributed to the transition between the ground electron state and the second heavy-hole states ($1^e-2hh$) since its energy separation from the $1^e-1hh$ peak (7.3 meV) is quite near to the calculated value of about 9.7 meV. We have not confirmed whether the observation of transitions with different quantum numbers in this work ($\Delta n=1$) is a characteristic property of crescent-shaped QWRs or due to some extrinsic effects, for example the asymmetry of wire shape.

We next discuss the Stokes shifts using the spectra given in Figs. 2 and 3. From Fig. 3, the Stokes shift of the $1^e-1hh$ peak is only about 4 meV, a value two times smaller than that of QWRs fabricated by MOVPE continuous growth under similar conditions. The $1^e-1hh$ transition also showed a Stokes shift exactly the same as the $1^e-1hh$ transition (≈4 meV) since both of the PLE and the high excitation PL spec-
break into periodic step array with lower index facets along the [011] direction to reduce the surface energy during epitaxial growth. The height and the period of the step array are reported to be about 32 and 10 Å in molecular beam epitaxy. Recently, Reinhardt et al. made an atomic force microscopy observation of the surface of GaAs QWRs grown on V-grooved substrates by low pressure MOVPE. They found that the (001) facet at the V-groove bottom is segmented into many sections with typical length of several hundred nanometers along the wire axis direction and within these sections monolayer step terraces were observed. On the contrary, strong step bunching was observed on the (311)A facets with a mean periodicity of about 50 nm and a height variation up to 5 nm. Therefore, the large Stokes shift of the 2e−2hh transition can be attributed to the spatial separation of the 1e−1hh and the 2e−2hh wave functions which are distributed mainly on the (001) and the (311)A facets, respectively, as shown in Fig. 1(b).

In summary, we have studied the Stokes shift of different energy states of a crescent-shaped AlGaAs/GaAs QWR grown by FME using PLE and high excitation PL measurements. The 2e−2hh excited state transition showed a Stokes shift (17 meV) considerably larger than that of ground state-related transitions (4 meV for 1e−1hh, 1e−1lh). The large difference in Stokes shift can be explained by the spatial separation of wave functions of the ground and the excited states in crescent-shaped QWR structures.

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1. See, for example, G. Bastard, Wave Mechanics Applied to Semiconductor Heterostructures (Les Editions de Physique, Les Ulis, 1988).
11. Thickness fluctuation of one (001) ML is calculated to cause an energy variation of about 10 and 12.5 meV for the 1e−1hh and 2e−2hh peaks, respectively. Therefore, the large difference in Stokes shift between the 1e−1hh and the 2e−2hh states cannot be explained only by the confinement energy variation with thickness fluctuation as in the case of QWL structure.