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著者	Tsuruoka T., Ohizumi Y., Ushioda S., Ohno Y., Ohno H.
journal or publication title	Applied Physics Letters
volume	73
number	11
page range	1544-1546
year	1998
URL	http://hdl.handle.net/10097/51781

doi: 10.1063/1.122200

Light emission spectra of AlGaAs/GaAs multiquantum wells induced by scanning tunneling microscope

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(Received 24 April 1998; accepted for publication 13 July 1998)

We have investigated the scanning-tunneling-microscope light emission (STM-LE) spectra of p-$Al_{0.4}Ga_{0.6}As$/p-GaAs multiquantum wells. The injection current level was kept as low as 0.1–0.5 nA to ensure that the sample is not damaged by the tunneling current. This is the current level ordinarily used for taking STM images. The peak energy of the emission shifts to the high energy side with decreasing well widths. A corresponding peak shift behavior was also observed in the photoluminescence (PL) spectra for the same samples. From comparisons of the STM-LE and the PL spectra, we find that although there is a difference in the excitation process, the final recombination process is identical in both cases. © *1998 American Institute of Physics.*
[S0003-6951(98)01837-3]

One of the most recent applications of the scanning-tunneling microscope (STM) is to use the tip as an injection source of charge carriers (electrons or holes) to induce light emission from semiconductor surfaces and quantum structures. For direct band gap semiconductors like GaAs, the carriers injected locally from the tip recombine radiatively in the sample. Thus, emitted photons are expected to provide information about local electronic properties of the material. In this regard STM light emission (STM-LE) spectroscopy is similar to cathodoluminescence (CL) using the scanning electron microscope. However, the STM has important advantages such as better spatial resolution and lower energies of injected carriers compared to CL. STM-LE can investigate luminescence properties simultaneously with the topography of the sample surface with a resolution of a nanometer scale.

Measurements of STM-LE from various semiconductor surfaces and structures have been reported by several groups in recent years. Berndt *et al.*[1] measured the STM-LE spectra and its integrated intensity maps of a cleaved CdS (1120) surface, and showed that this technique can be used as a sensitive and high spatial resolution probe for defect levels. Alvarado and co-workers[2–4] reported the STM-LE intensity maps of cleaved AlGaAs/GaAs heterostructures, and estimated the carrier diffusion length by measuring the decay profile of the luminescence intensity from quantum wells. Samuelson *et al.*[5] measured the STM-LE spectra of single quantum wires made of GaAs/AlGaAs heterostructures grown on V grooves using a pre-etched corrugated substrate. They found a difference in the spectral features at two points separated by about 100 nm on the sample surface. The same group has also reported the STM-LE spectra of InP quantum dots, and observed the Stark shift of the emission peak by changing the tip–sample bias.[6,7]

In most of the previous studies the intensity maps and the spectra of STM-LE were measured by setting the tunneling current and the bias voltage much higher than for usual measurements of surface topography. Under these conditions it is difficult to take STM images simultaneously, and the sample surface may be modified by a high tunneling current. To understand the physical origin of the luminescence induced by STM, it is necessary to measure the emitted light using a low tunneling current level so that one can simultaneously take STM images without modifying the sample surface. Recently, Murashita[8] has measured the STM-LE spectra of a cleaved AlGaAs/GaAs quantum well structure with a tunneling current less than 1 nA, by using a novel conductive transparent tip that can inject carriers and collect light simultaneously. In this letter we report on light emission studies of AlGaAs/GaAs multiple quantum wells with different well widths using a low tunneling current level of STM.

The three samples discussed in this letter have twenty periods of $Al_{0.4}Ga_{0.6}As$/GaAs quantum wells that were grown on p-type GaAs (100) substrates by molecular-beam epitaxy (MBE). The three samples have an identical thickness of the $Al_{0.4}Ga_{0.6}As$ layers (20.4 nm) and differ in the thickness of the GaAs well layers, being 10.2, 5.1, and 2 nm, respectively. The sample surface was terminated by an $Al_{0.4}Ga_{0.6}As$ layer. All the layers were Be doped to a level of about 1×10^{19} cm^{-3}. The growth rates were 0.50 and 0.83 monolayer/s for GaAs and $Al_{0.4}Ga_{0.6}As$, respectively. After growth the samples were cooled below room temperature, and were exposed to a molecular beam of As_4 for 30 min, in order to form a protective overlayer of As. This procedure allows us to transfer the sample from the MBE chamber to the STM chamber through atmosphere without any contamination of the sample surface.[9] After introducing the sample into the STM chamber, the As layer was desorbed by heating to 370 °C for 30 min.

The STM-LE measurements were performed under ultrahigh vacuum conditions with a base pressure better than

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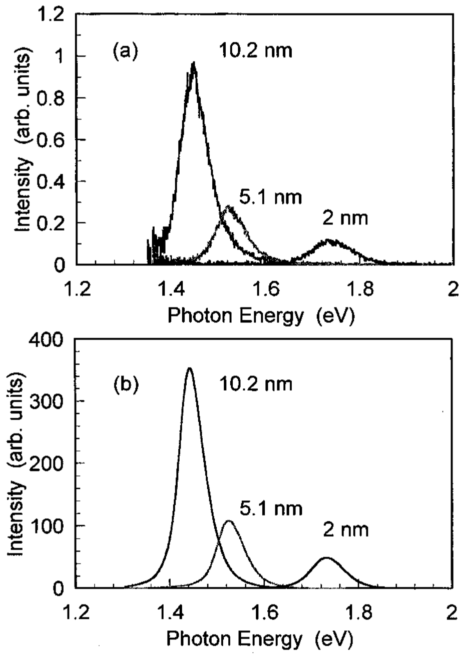

FIG. 1. Comparison of STM-LE (a) and PL (b) spectra of $Al_{0.4}Ga_{0.6}As/GaAs$ multi QW's. The STM-LE spectra were taken with a tunneling current of 0.5 nA and a sample bias voltage of +3 V. The energy resolution for the STM-LE and PL are about 50 and 12 meV, respectively.

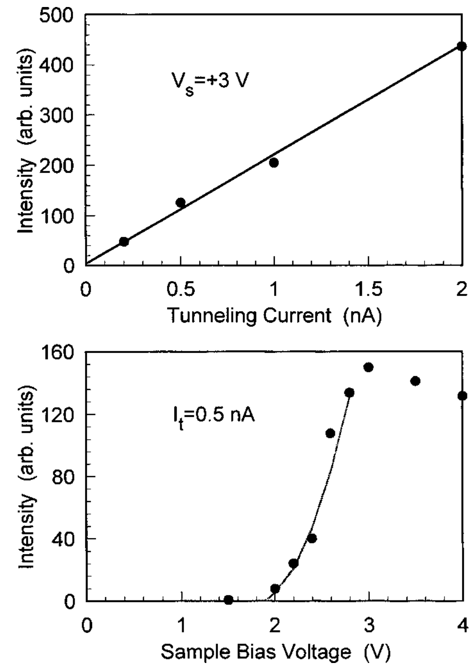

FIG. 2. Dependence of spectrally integrated intensity on the tunneling current and sample bias voltage. (a) Intensity as a function of tunneling current, recorded at a constant sample bias voltage of +3 V. (b) Intensity as a function of sample bias voltage, recorded at a constant tunneling current of 0.5 nA. The solid lines are the guide to the eye.

3×10^{-9} Pa at room temperature. The STM was operated in a constant-tunneling-current mode. Probe tips made of PtIr were used. Light emission measurements were started after confirming that the STM image of the sample surface shows a (2×4) reconstruction structure. The emitted light from the sample was collected by a lens mounted in vacuum. It was then led into a spectrograph through a viewing port and a second lens, and finally detected by an optical multichannel photodetector (1024 channels). The tunneling current was typically in the range from 0.2 to 2 nA. We note that the STM-LE spectra could be recorded using a tunneling current of 0.1 nA. The STM-LE spectra shown in this letter are corrected for the energy-dependent sensitivity of the photodetection system.

Figure 1(a) shows the STM-LE spectra of the three samples, taken with a tunneling current of 0.5 nA and a sample bias voltage of +3 V. All spectra consists of a single emission peak. The peak energy of the emission shifts to the high energy side from 1.44 to 1.73 eV with decreasing well widths. A corresponding peak shift was also observed in the photoluminescence (PL) spectra shown in Fig. 1(b). The PL spectra were measured in air for the same samples, using the 514.5 nm line of an argon ion laser as the excitation source with an incident power of 5 mW.

To identify the observed emission peaks, we calculated the energy levels of a simple quantum well (QW) model in which the band bending effect caused by charge transfer from AlGaAs to GaAs layers is neglected.[10] From this calculation the emission peaks could be assigned to transitions between the $n=1$ single-quantum-well electron and hole states of the respective samples. Figure 1 shows that the spectral profile and the relative intensity are basically identical in both the STM-LE and the PL spectra. Thus we see that the final process in light emission is identical in both STM-LE and PL; i.e., electron–hole pair recombination in the well. However, there is a difference in the initial part of the excitation process. No emission was observed for negative sample bias ranging up to −4 V. This means that photon emission occurred in a tunneling mode rather than a field-emission mode of STM. We conclude that in STM-LE of *p*-type samples minority carrier injection is necessary, while in PL both minority and majority carriers are automatically created in the sample by the incident laser.

Figure 2 shows the dependence of the spectrally integrated intensity on tunneling current and sample bias voltage for the sample with the well width of 10.2 nm. Similar results were obtained for the other two samples. The integrated emission intensity is proportional to the tunneling current as seen in Fig. 2(a). Since no shift and broadening of the emission peak is observed when the tunneling current is increased, there is no band filling in the lowest energy states of the GaAs wells within the measured current range. In the bias voltage dependence shown in Fig. 2(b) there is a threshold below +2 V, which closely corresponds to the band gap energy of $Al_{0.4}Ga_{0.6}As$. Just above the threshold the luminescence intensity has a V^2 dependence where V is the bias voltage, similar to the case of internal photoemission[11] and injection of electrons from a metal into a semiconductor in a ballistic electron emission microscope.[12] This quadratic dependence was observed also for the STM-LE of heterojunctions of $Al_xGa_{1-x}As$,[4] InP,[13] GaN,[14] and gold-passivated GaAs.[15] The intensity beyond the threshold increases initially, but starts to decrease slightly at sample bias voltages above +3 V. Calculations and inverse-photoemission measurements show that the density of states (DOS) of GaAs has a local minimum at 4 eV above the top of the valence band.[16] We believe that the decrease observed in the integrated intensity is related to this dip in the conduction band DOS.

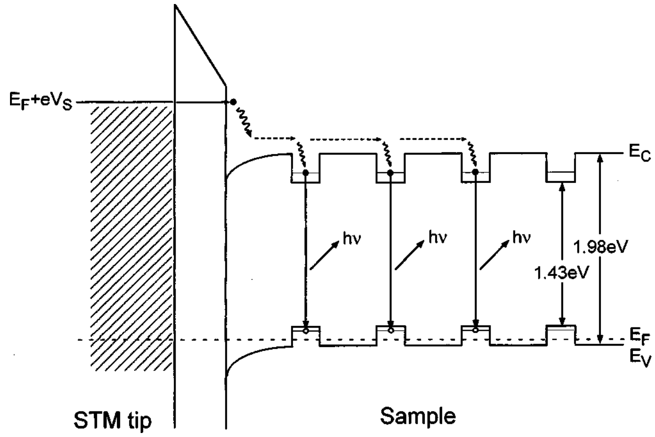

FIG. 3. A schematic diagram of light emission by injection of minority carriers from the STM tip into the $Al_{0.4}Ga_{0.6}As$/GaAs multi QW structure. The horizontal lines across the quantum wells show the energy levels of the electron and hole ground states.

The process of the observed light emission can be schematically illustrated in Fig. 3. For heavily *p*-doped samples, the Fermi level lies very close to or below the top of the valence band. Thus, in the case of multi QW structure shown in Fig. 3, most of the holes are confined in the GaAs quantum wells. When the sample bias voltage becomes larger than the band gap energy of the top $Al_{0.4}Ga_{0.6}As$ layer, electrons are injected from the tip into the conduction band of the sample. Some of them are trapped by contaminants or defects at the surface, and others diffusing into the sample fall in the GaAs wells and relax to the electron ground state. Finally luminescence occurs through recombination of the electrons with holes in the hole ground state of the wells. Since most of the injected electrons thermalize within a range of 100 nm from the surface,[17] we believe that luminescence arises only from several wells close to the sample surface.

The well width is expected to have a fluctuation on the order of a monolayer from place to place on the surface. Thus one would expect to see small variations in the peak position as the STM tip is moved to different locations. However, the expected peak energy variation due to well width fluctuations is smaller than 26 meV corresponding to the thermal energy broadening at room temperature. Thus we did not observe such peak position shifts that depend on the location of the tip.

Also we did not see the energy splitting due to the presence of light and heavy holes which is again smaller than the thermal energy broadening. STM-LE measurements at low temperatures should allow us to obtain the spectra of unbroadened luminescence with higher spectral resolution and thus permit a more detailed study of quantum structures.

In summary we have measured the STM-LE spectra of $Al_{0.4}Ga_{0.6}As$/GaAs multi QW's while keeping the tunneling current level as low as 0.1–0.5 nA. This current level ensures that one can measure the topography and the light emission spectra simultaneously without modifying the sample surface. Shift of the emission peak that depends on well widths was observed also in the PL spectra, and agrees with the calculation for the transition energies of the QW structures. The comparison of the STM-LE and the PL spectra demonstrates that the final light emission processes in both cases involves electron–hole pair annihilation, although the initial excitation processes are different in the two methods.

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