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Effect of exciton oscillator strength on upconversion photoluminescence in GaAs/AlAs multiple quantum wells

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We report upconversion photoluminescence (UCPL) in GaAs/AlAs multiple quantum wells. UCPL from the AlAs barrier is caused by the resonant excitation of the excitons in the GaAs well. When the quantum well has sufficient miniband width, UCPL is hardly observed because of the small exciton oscillator strength. The excitation-energy and excitation-density dependences of UCPL intensity show the exciton resonant profile and a linear increase, respectively. These results demonstrate that the observed UCPL caused by the saturated two-step excitation process requires a large number of excitons. © *2014 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4901080]

Upconversion photoluminescence (UCPL), which is photo-emission with photon energies higher than those of the excitation source, has been intensively studied from the perspectives of material physics and applications.[1–19] Several excitation mechanisms cause UCPL, including the two-photon absorption process,[13,14,18] Auger process,[2–4] and resonant tunneling.[1,6,10,15,16] Recently, an important application of the upconversion process has been in the photovoltaic cells, in which low-energy photons are absorbed through upconversion.[20–22] In particular, the upconversion process is essential to avoid the fabrication of complicated devices and to make structures simpler.

Therefore, if the high-efficiency upconversion process is realized, it is possible to improve the device performance from various aspects. In this work, we focused on the exciton oscillator strength and investigated UCPL in GaAs/AlAs multiple quantum wells (MQWs) with varying the barrier thickness. UCPL from AlAs, which is the barrier, was observed under the condition of resonant excitation of the exciton state in MQWs without miniband. On the other hand, UCPL was hardly observed in the samples with miniband because of weak exciton oscillator strength. The UCPL mechanism will be discussed on the basis of the results of excitation-energy and excitation-power dependences.

The samples used in this work were four $(GaAs)_{30}/(AlAs)_n$ MQWs grown on a (001) GaAs substrate by molecular-beam epitaxy, where subscripts denote the constitution layer thicknesses by a monolayer unit of 0.283 nm. The thickness of the AlAs barrier layer n was changed from 30 to 2. Hereafter, we will call the samples as "$n = X$ sample." The calculated miniband widths of electron and heavy hole (HH) subbands based on the effective mass approximation are listed in Table I. In this calculation, the values of the effective mass and band offset are taken from Refs. 23 and 24, respectively. The MQW period was 30 for all samples except for the $n = 4$ sample, which has 20 quantum wells. To measure the UCPL spectra, a mode-locked Ti:sapphire pulse laser, delivering an approximately 120-fs pulse with a repetition rate of 80 MHz, was used as the excitation source. The emitted light was dispersed in a 35-cm monochromator with a resolution of 0.1 nm and detected by a charge-coupled device. To evaluate exciton energies in the samples, we measured the conventional photoluminescence (PL) spectra excited by a semiconductor laser with the 1.850-eV line.

Figure 1 shows the PL spectra in the samples. The PL peaks originate from the HH excitons confined in GaAs well layers. The exciton line widths in the samples are almost equal; samples are of almost the same quality. The UCPL spectra were measured under these exciton resonant excitation conditions.

Figure 2 depicts UCPL spectra observed under the condition of HH exciton excitation in the samples. UCPL spectra were clearly observed in the $n = 10$ and $n = 30$ samples at only around 2.36 eV, as indicated by the dashed and solid curves, respectively. UCPL spectra were hardly observed in the $n = 2$ and $n = 4$ samples. The peak energy is close to the transition energy from the L valley of the conduction band to the Γ point of the valence band in AlAs, which is 2.46 eV.[25] Therefore, this UCPL arises from the L-Γ transition in AlAs. The difference of PL energy from L-Γ transition energy is attributed to alloying of GaAs-AlAs at the interface.

To reveal the reason behind the absence of UCPL in the $n = 2$ and $n = 4$ samples, the exciton PL peak intensity in Fig. 1 and the UCPL peak intensity in Fig. 2 were plotted as functions of barrier thickness n in Fig. 3 and shown as open and closed circles, respectively. In this plot, though the well

TABLE I. Calculated miniband widths of electron and HH subbands.

n	Electron (meV)	HH (meV)
30	0	0
10	3.5	0
4	11	0.3
2	49	2.6

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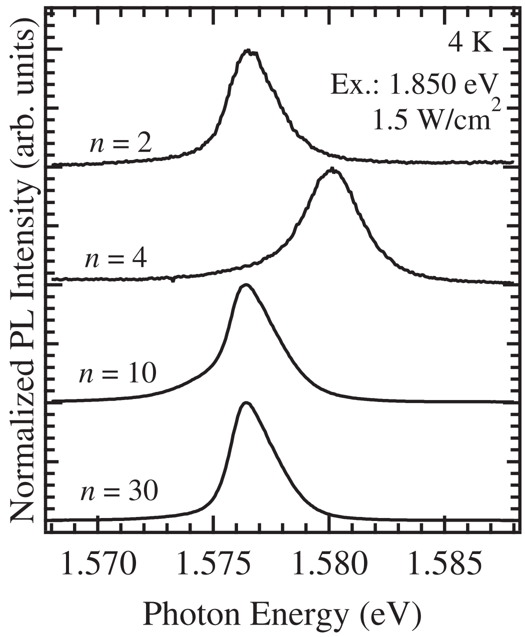

FIG. 1. Exciton PL spectra in the samples.

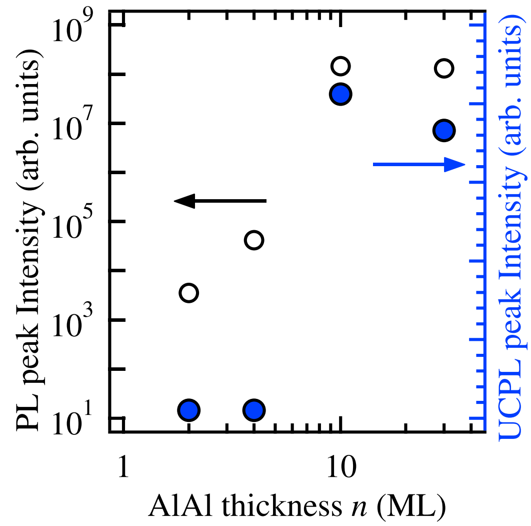

FIG. 3. Exciton PL and UCPL peak intensities plotted as a function of n and shown as open and closed circles, respectively. The left axis is in logarithmic scale.

number in the $n=4$ sample is different from that in other samples, it is not a problem in this scale. It should be noted that the exciton PL intensity was plotted in the logarithmic scale. Although the UCPL efficiency is very low (less than 1%), this comparison indicates that the upconversion process is somehow related to the exciton oscillator strength. This is a key point in this work.

To clarify the upconversion mechanism, we measured the excitation energy dependence with different excitation densities in the $n=30$ sample. The UCPL peak intensity is plotted as a function of excitation energy in Fig. 4. The dotted line indicates the HH exciton energy. This result clearly indicates that UCPL is caused by the resonant excitation of the HH exciton, which was observed in all excitation densities. Mainly, there are three possible mechanisms that explain the upconversion process; (i) two-photon absorption, (ii) Auger process, and (iii) two-step excitation. When the two-photon absorption causes upconversion, the excitation-energy dependence should depend on the absorption band of AlAs. Considering the transition energy at the Γ point of 3.13 eV, two-photon absorption is ruled out. Therefore, the two-step excitation or Auger process are possible mechanisms.

Subsequently, we focused on the excitation-density dependence of the UCPL intensity in Fig. 4. The peak intensity of UCPL in the $n=30$ sample was plotted as a function of excitation density in Fig. 5. The dependence of the UCPL intensity on the excitation density was evaluated through least-squares fitting. This result indicates that the UCPL intensity depends on the excitation density almost linearly. The linear dependence rules out the possibilities of the disorder-mediated UCPL.[26] There are several reports suggesting that the linear dependence of UCPL is caused by limited number

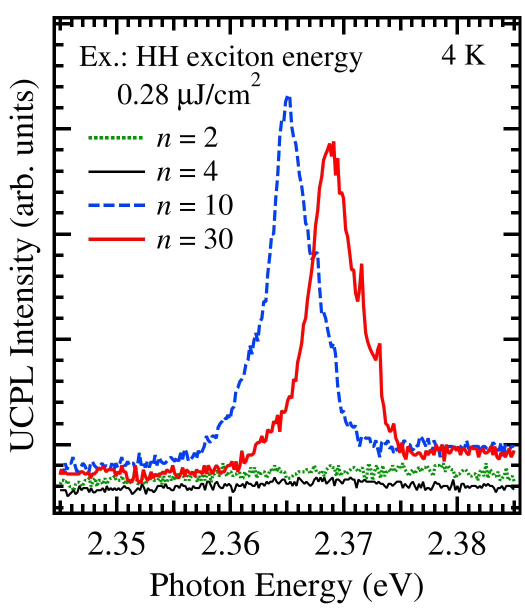

FIG. 2. UCPL spectra observed under the condition of HH exciton excitation.

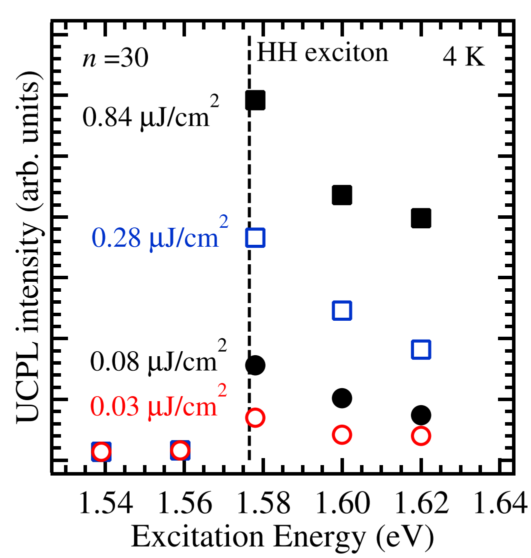

FIG. 4. Excitation-energy dependence of the UCPL intensity observed at different excitation densities. The dotted line indicates the HH exciton energy.

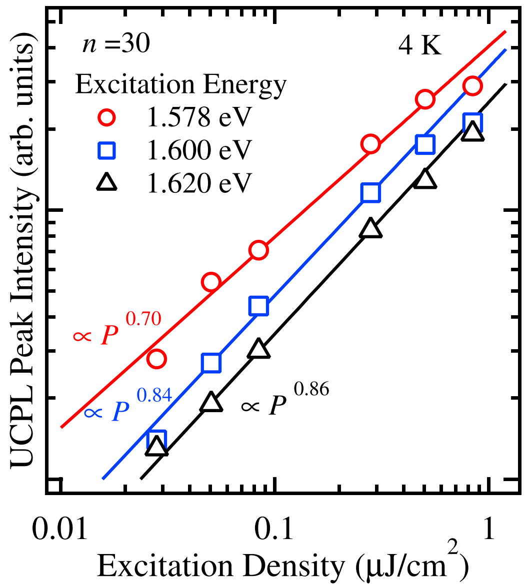

FIG. 5. Excitation-density dependence of the UCPL intensity observed at different excitation energies. Solid lines indicate the results of least-squares fitting.

of trap states,[27] thermal distribution,[8,28–30] Auger process,[31] and saturation effect.[32,33]

Among these mechanisms leading to linear dependence, we focused on the saturation effect. That is, while the excitons in the GaAs wells are excited by the two-step excitation process and the excited electrons and holes move to AlAs barriers, the saturation effect modifies the excitation-density dependence to linear. This saturation of the exciton state is similar to the excitation-density dependence of the four-wave-mixing signal intensity.[34] Although this upconversion process is saturated, a large number of excitons is required to cause this UCPL. Therefore, samples with miniband width, that is, those with weak oscillator strength, hardly exhibit UCPL. This result indicates that the achievement of high density exciton is a key factor to induce UCPL.

We have investigated the UCPL mechanism appearing in GaAs/AlAs MQWs under the resonant exciton excitation condition. UCPL was observed in MQWs with large exciton oscillator strength, and it shows an exciton resonant profile in excitation-energy dependence and linear dependence on excitation density. The observed UCPL originates from the saturated two-step excitation process. Our results demonstrate that the realization of exciton reservoir leads to high-efficiency UCPL.

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