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Photoluminescence Up-conversion in GaAs/AlGaAs Heterostructures

Hyeonsik M. Cheong

Department of Physics, Sogang University, Seoul 121-742, Korea
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Abstract

Photoluminescence up-conversion in semiconductor heterostructures is a phenomenon in which luminescence occurs at energies higher than that of the excitation photons. It has been observed in many semiconductor heterostructure systems, including InP/AnALAs, CdTe/CdMgTe, GaAs/ordered-(Al)GaInP, GaAs/AlGaAs, and InAs/GaAs. In this work, GaAs/AlGaAs heterostructures are used as a model system to study the mechanism of the up-conversion process. This system is ideal for testing different models because the band offsets are quite well documented. Different heterostructures are designed to study the effect of disorder on the up-converted luminescence efficiency. In order to study the roles of different types of carriers, the effect of doping was investigated. It was found that the up-converted luminescence is significantly enhanced by p-type doping of the higher-band-gap material.

1. Introduction

Photoluminescence (PL) up-conversion in semiconductor heterostructures is a phenomenon where luminescence occurs at energies higher than that of the excitation photons. It has been observed in many hetero-structures, including InP/AlInAs [1], CdTe/CdMgTe [2], and GaAs/ordered-(Al)GaInP [3-8]. The GaAs/ordered-(Al)GaInP heterojunctions (HJs), in particular, have attracted much attention due in part to very large energy gains (up to 700 meV), leading to emission in the technologically important red-green region of the spectrum. Here, luminescence from spontaneously-ordered (Al)GaInP (~1.9-2.2 eV) was observed when these HJs were excited with photons with an energy just higher than the band gap of GaAs (1.519 eV). Up-converted PL (UPL) requires that (1) there exists a mechanism that "up-converts" electrons and/or holes from the low-band-gap material to the high-band-gap material and (2) such up-converted carriers do not relax back to the low-band-gap material before they radiatively recombine. It has been demonstrated that the photocarriers created in the lower band-gap-energy semiconductor are successively excited into the higher-band-gap-energy material and then radiatively recombine, giving luminescence at the energy of the higher-band-gap material. However, this material system is not suitable for studying the detailed physical mechanism of UPL due to the inadequate knowledge of the band alignment between GaAs and ordered-(Al)GaInP. As an alternative, the GaAs/AlGaAs heterostructure system [9] is suggested as a prototypical system to study the physics of the UPL phenomenon. The advantage here is that the band alignment between GaAs and AlGaAs is well established. Most studies to date used nominally undoped samples and the data were analyzed under the assumption that the background doping was negligible. On the other hand, if the sample is explicitly doped, the different roles of electrons and holes in the up-conversion process could be illustrated. In this paper, we report the effect of doping on the up-converted photoluminescence in

GaAs/AlGaAs heterostructures.

2. Experimental

A series of samples with different heterojunctions were designed as shown schematically in Fig. 1. The samples have the same structure, except for doping, consisting of a GaAs substrate, a nominally undoped 0.5-1.0 μm GaAs buffer layer, a 300-Å $Al_{0.3}Ga_{0.7}As$ barrier layer, a 1000-Å $Al_{0.2}Ga_{0.8}As$ layer, another 300-Å $Al_{0.3}Ga_{0.7}As$ barrier layer, and a 100-Å GaAs capping layer. The $Al_{0.3}Ga_{0.7}As$ layers serve as barriers that prevent relaxation of up-converted carriers from the $Al_{0.2}Ga_{0.8}As$ layer to the GaAs layers. Carriers are introduced by delta-doping either the $Al_{0.3}Ga_{0.7}As$ barrier layer (remote doping) or the $Al_{0.2}Ga_{0.8}As$ layer (center doping). The samples were grown by low-pressure (75 torr) metal-organic chemical vapor deposition (MOCVD) at 750℃. The delta doping was carried out by interrupting growth and then exposing the sample surface for 20 s to arsine and disilane for *n*-type Si doping and to arsine and diethylzinc for *p*-type Zn doping. The doping densities, estimated from secondary-ion mass spectroscopy measurements, are approximately 1×10^{18} cm^{-3} and 1×10^{17} cm^{-3} for Si and Zn, respectively. UPL measurements were performed at 10 K with a cw titanium:sapphire laser at 8034 Å (1.543 eV) as the excitation. An RG780 long-pass filter was placed in the laser beam path in order to prevent the short-wavelength tail of the titanium:sapphire luminescence from exciting AlGaAs PL directly. The UPL signal was dispersed by a 0.6-m triple spectrometer and detected with a liquid-nitrogen-cooled back-illuminated charge-coupled-device (CCD) array.

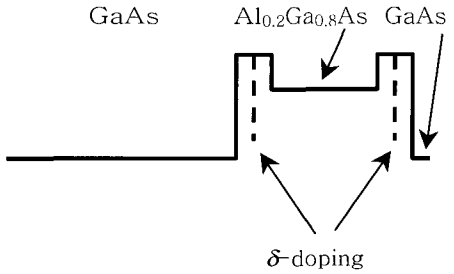

Fig. 1. Schematic band edge diagrams of GaAs/AlGaAs heterostructures used in this work. Only the conduction band edge for a remote delta-doped sample is shown for simplicity.

3. Results and Discussion

Figure 2 shows luminescence spectra of the four samples. For each sample, a UPL signal is observed about 200 meV above the excitation energy, indicated by an arrow. Much stronger GaAs PL (scaled down by a factor of 10000) is seen at the GaAs exciton energy at 1.515 eV. Figure 2 compares the UPL spectra of the samples grown on undoped substrates with different doping. There is some variation in UPL energy due to the growth-to-growth variation of Al concentration. There is a definite correlation between the doping and the UPL intensity: for the same structure, *p*-type doping gives stronger UPL than *n*-type doping [(c) vs. (b) or (e) vs. (d)]; and for the same type of doping, remote doping results in stronger UPL than center-doping [(b) vs. (d) or (c) vs. (e)]. In order to show this correlation more clearly, we calculated the integrated UPL intensity for different sample structures and summarized it in Table 1. The above-mentioned correlation is clearly seen. The results given in Table 1 can be summarized as follows: (1) *n*-type doping of the AlGaAs layer modestly reduces the UPL intensity and *p*-type doping increases it significantly, relative to the case of undoped AlGaAs; (2) remotely doped samples show stronger UPL than center-doped samples; and (3) samples grown on *n*-type substrates give a lot stronger UPL than those grown on undoped or *p*-type substrates. Similar substrate dependence was observed in the PL intensity of the GaAs buffer layers of these samples: samples grown on *n*-type substrates show stronger GaAs buffer PL than those grown on *p*-type substrate. Therefore, the dependence of the UPL

Table 1. Integrated up-converted PL intensity (in arbitrary units) for different doping of the AlGaAs layer and the substrate.

AlGaAs doping	Substrate type	UPL Integrated Intensity	
		Remote Doping	Center Doping
	n	455.0	196.1
p	undoped	291.4	155.1
	p	271.8	19.6
undoped	undoped	122.3	
	n	103.9	8.57
n	undoped	95.3	17.1
	p	32.7	0.6

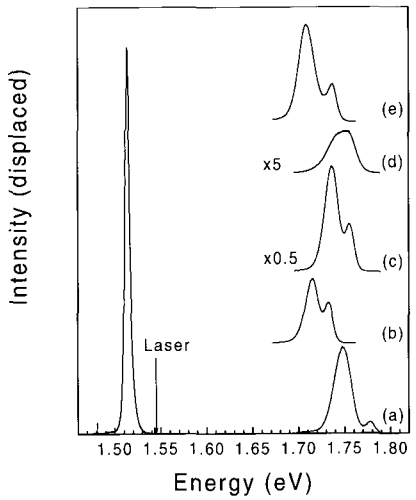

Fig. 2. Up-converted PL spectra of the samples grown on undoped GaAs substrates. The AlGaAs layers are (a) undoped, (b) *n*-type remotely doped, (c) *p*-type remotely doped, (d) *n*-type center-doped, and (e) *p*-type center-doped. The excitation laser energy (1.543 eV) is indicated by an arrow. The photoluminescence spectrum from the GaAs buffer layer for the undoped sample, scaled down by a factor of 10000, is also shown for comparison.

intensity on substrate doping type is not directly related to the up-conversion process.

The fact that remote doping results in much stronger UPL intensity than center doping can be easily understood because introduction of dopants in the $Al_{0.2}Ga_{0.8}As$ layer decreases the radiative recombination efficiency of the up-converted carriers. On the other hand, there are not many free carriers in the layer because most carriers freeze out at 10 K since the carriers and the dopants are not spatially separated. In the remotely doped structures, however, the carriers do not freeze out because the dopants and the carriers are spatially separated by the heterojunction, and there is no dopant in the $Al_{0.2}Ga_{0.8}As$ layer to reduce the radiative recombination efficiency. There is a negative effect in this case as well: the excess carriers in the $Al_{0.2}Ga_{0.8}As$ layer result in band-bending. Since the $Al_{0.2}Ga_{0.8}As$ layer is 1000-A thick, the band-bending spatially separates electrons and holes, reducing the radiative recombination efficiency. Therefore, there is a competition between the positive effect of excess carriers and the negative effect of the band-bending. The band-bending thus reduces the radiative recombination efficiency for both p- and n-type doped samples. In the case of the n-type doped samples, this negative effect is greater than the positive effect of introducing excess carriers, and that is why the UPL intensity of the n-type doped samples is lower than that of the undoped sample. For the p-type doped samples, the positive effect of excess carriers is so large that it overcomes the negative effect of band bending.

If the excitation efficiencies of electrons and holes are similar, then the effect of doping on the UPL intensity should be similar. However, our results show clearly that the electrons are excited more efficiently than the holes. In other words, the rate-limiting factor in the up-conversion process is the excitation efficiency of holes. In the cold-Auger excitation model, this is explained as being due to the heavier hole mass because the Auger excitation rate is smaller for heavier particles. [10] A similar calculation has not been reported for other models for UPL.

Our result has implications in the interpretation of the intensity-dependence data for UPL. It has been a common practice to measure the dependence of the

UPL intensity on the excitation intensity in order to support or refute various models for the dominant mechanism for the up-conversion process. [1,2,7,9] In these analyses, it has been assumed that the excitation efficiency is the same for electrons and holes, and furthermore, the background doping was ignored except for the explicitly doped case. [1] However, if the excitation efficiency is dissimilar for electrons and holes, the excitation power dependence would be affected. For example, in the non-excitonic Auger excitation model, an excited electron-hole pair is created through hhe and eeh Auger processes: 3 electrons and 3 holes are needed to create one excited pair. Therefore, if the excitation efficiency were the same for electrons and holes, the UPL intensity would have a cubic dependence on the excitation intensity. On the other hand, if the excitation efficiency is much higher for the electrons than for the holes, then the UPL intensity would have a quadratic dependence on the excitation intensity because then the hhe Auger process would be the rate-limiting process. Similar arguments can be applied to other models for the up-conversion process.

In summary, we have found that the UPL intensity in GaAs/AlGaAs heterostructures is enhanced when the AlGaAs layer is remotely doped with holes. It indicates that the rate-limiting process in the up-conversion process is the excitation of holes. Our result shows that one has to be careful in interpreting the excitation power dependence of the UPL intensity because dissimilar excitation efficiencies for electrons and holes and possible background doping of the samples can significantly modify the excitation power dependence.

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