Photoluminescence of InAs quantum dots in n-i-p-i GaAs superlattice

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Large blueshift and linewidth increase in photoluminescence (PL) spectra of InAs quantum dots (QD’s) in n-i-p-i GaAs superlattice were observed. By increasing the excitation intensity from 0.5 to 32 W/cm², the PL peak position blueshifted 18 meV, and the linewidth increased by 20 meV. Such large changes are due to the state-filling effects of the QD’s resulted from the separation of photogenerated electrons and holes caused by the doping potential.

The optical properties of InAs self-assembled quantum dots (QD’s) have been of great interest in recent years. The interband and intraband optical transitions of these QD’s are widely studied, both for the interest in fundamental physical properties and for the development of QD lasers and photodetectors. It is important to investigate electron-hole separation effects in QD’s and design QD structures that can separate and store photogenerated carriers for memory-device applications. However, few works were done in this field. Recently, Schoenfeld et al.¹ have designed a QD device that can separate and store photogenerated electrons and holes. Their structure consisted of two GaAs quantum wells of different thickness, separated by a thin AlAs barrier. An InAs QD layer is inserted in the thick quantum well. The strain these InAs QD’s create induces QD’s within the thin GaAs quantum well that are coupled to the InAs QD’s in the thick well. In their structure, photogenerated electrons and holes are spatially separated into the InAs QD’s and strain-induced QD’s, respectively.

In this work, we study a different QD structure designed to spatially separate and store photogenerated electrons and holes. In this structure, InAs self-assembled QD’s were grown in the n-type and p-type regions of a n-i-p-i GaAs superlattice. Photogenerated electrons and holes in GaAs barriers are expected to be swept by the intrinsic electric field to the n-doped regions and the p-doped regions, respectively, and trapped by the InAs QD’s there. As the photogenerated electrons and holes fill up the lower QD states, photoluminescence (PL) properties of these InAs QD’s different from conventional QD’s inserted in intrinsic GaAs layers are expected. In our PL experiments, large blueshift of the PL peak and increase of the PL linewidth with increasing excitation intensity were observed for the QD’s in the n-i-p-i GaAs superlattice, whereas for similar InAs QD’s in intrinsic GaAs layers, only a small blueshift of the PL peak and increase of the linewidth were observed. The large blueshift in the n-i-p-i QD’s is a clear indication of the state filling of the QD’s due to electron-hole separation.

The samples were grown by molecular-beam epitaxy on a (001) N⁺ GaAs substrate. After deposition of a 1-µm semi-insulating GaAs type buffer layer, 10 periods of n-type GaAs (20 nm)/InAs QD’s (2 ML)/n-type GaAs (20 nm)/p-type GaAs (20 nm)/InAs QD’s (2 ML)/p-type GaAs (20 nm) were grown. The n-type regions were Si doped and the p-type regions were Be doped; both n and p doping concentrations are 1×10¹⁸ cm⁻³. The growth temperature for the whole sample was 520 °C. Here we refer to the QD’s in the n-type GaAs layers and in the p-type GaAs layers n-n QD’s and p-p QD’s, respectively. For comparison, a reference sample with 20 periods of 2 ML InAs QD’s separated by 40 nm of intrinsic GaAs (with the same type of substrate and GaAs buffer layer) was also grown at the same growth condition. For convenience of analysis, we refer to the QD’s of the reference sample i-i QD’s as there are in the intrinsic GaAs layer.

The PL measurements were performed at 15 K with a Fourier transform spectrometer equipped with an In,Ga₁₋ₓAs detector. The spectra were obtained under excitation from the 514.5-nm line of an argon-ion laser with intensity varied from 0.5 to 32 W/cm². Figure 1(a) shows the PL spectra of the sample under excitation power density of 0.5 and 32 W/cm². The shift of PL peak energy as well as the increase of full width at half maximum (FWHM) of the sample under high-excitation power density is quite significant. For comparison, the corresponding PL spectra of the i-i QD sample are plotted in Fig. 1(b). It is seen that the increase of FWHM and shift of peak energy of this sample under high excitation is much smaller than the n-i-p-i sample. It is also seen that under low-excitation intensity of 0.5 W/cm² the PL peak energy of the n-i-p-i sample is about 30 meV higher than that of the i-i sample. The shift of peak energy and the increase of FWHM of the two samples are plotted as a function of pump power density in Figs. 2(a) and 2(b), respectively. From 0.5 to 32 W/cm², the PL peak position of the n-i-p-i sample has blueshifted 18 meV, and the FWHM increased by 20 meV. The peak position and the FWHM of the i-i QD sample, on the other hand, have changed by only 3 meV [solid squares in Figs. 2(a) and 2(b)]. This is similar to the PL of conventional i-i QD’s reported by others.²
We now discuss the possible causes of the unusually large blueshift and the FWHM increase in the \textit{n-i-p-i} sample. The band diagram of this sample is shown in Fig. 3. For the sake of convenience, the QD layers, which inevitably have size distribution, can be regarded as having many energy levels with small spacing in between. First, let us consider the PL of conventional \textit{i-i} QD's in GaAs. In these samples, the photogenerated electrons and holes in the lower QD energy states are quickly recombined, and the electrons and holes in the higher states can quickly thermal-relax to the lower states. As a result, most of the PL comes from the recombination from the lower states and a few from the higher states. In the \textit{n-i-p-i} QD case, the lower electron energy states of the \textit{n-n} QD's are already filled by electrons from the dopants. Upon photoexcitation, most of the photogenerated electrons are swept by the built-in electric field to the \textit{n}-doped regions and fill in the higher-energy states of the \textit{n-n} QD's. The small amount of the photogenerated holes captured by the \textit{n-n} QD's then recombines with the electrons, some at lower states and some at higher states. Similarly, most of the photogenerated holes are captured by the \textit{p-p} QD's and a small amount of them are recombined with the few captured electrons. Therefore, the overall PL peak energy is expected to be higher than that of the conventional \textit{i-i} QD's, and the PL intensity is lower than that of the \textit{i-i} QD's. This is consistent with the experimental results where the PL intensity of the \textit{n-i-p-i} QD sample is about $10^{-1}$ of that of the \textit{i-i} QD's, and the peak position of the \textit{n-i-p-i} QD's is higher than that of the \textit{i-i} QD's. In the meantime, most of the photogenerated electrons are still in the \textit{n-n} QD's and the holes are in the \textit{p-p} QD's, which are separated from the electrons by about 40 nm of GaAs barrier layer. Indirect recombination between these electrons and holes, which would produce PL at lower photoenergies, is unlikely because of the large spatial separation. At higher photoexcitation intensity more electrons fill the upper energy states of the \textit{n-n} QD's and recombine with the holes, resulting in the blueshift of the PL peak and the increase of FWHM. These changes of PL spectra with the increase of excitation intensity finally saturate because the photogenerated electron-hole separation reduces, and finally cancels, the built-in electric field.

Now we compare our PL results of the \textit{n-i-p-i} sample with similar works\textsuperscript{3–8} where many-body effects (or charging effects) in QD's were investigated. In these studies, the QD's were filled with electrons by doping. In the PL experiments, with increasing doping concentration, contrary phenomena were observed. In Ref. 7, the PL showed a blueshift, whereas in Ref. 4, the PL showed a redshift. The blueshift was suspected to result from state-filling effects and the redshift was attributed to band-gap renormalization. In the \textit{n-i-p-i} QD sample, due to the electron-hole separation, both \textit{n-n} QD's and \textit{p-p} QD's are charged during photoexcitation. It is a typical many-body system with the number of electrons (or holes) occupying the \textit{n-n} QD's and the \textit{p-p} QD's controlled by the excitation intensity. It is seen that the role of increasing the excitation intensity for the \textit{n-i-p-i} sample is similar to the role of increasing the doping concentration in Refs. 4 and 7. Our PL results are consistent with that of Ref. 7.

In summary, we have grown InAs QD's in an \textit{n-i-p-i} GaAs superlattice designed to spatially separate and store
photoexcited electrons and holes, and have investigated their PL properties. With increasing the excitation power density, large blueshift of PL peak position and increase of PL line-width is observed. This is due to the enhanced state-filling effects, which results from electron-hole separation in the $n$-$i$-$p$-$i$ structure.

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