Optical Study of the Anisotropic Confinement of Excitons in GaAs/AlGaAs Quantum Dots

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Anisotropic confinement of excitons in a single GaAs/Al$_{0.405}$Ga$_{0.595}$As quantum dot (QD) is studied using micro-photoluminescence (micro-PL) measurements. In this study, a low-density GaAs QD sample is prepared by Modified Droplet Epitaxy (MDE). The QDs are anisotropic in shape and relatively large. This low density sample is combined with micro-PL spectroscopy to obtain single QD spectra. From the single GaAs QD micro-PL spectrum-excitation density dependence, an initial blue shift of approximately 100 µeV is seen for the main peak with increasing excitation density, and as the excitation density is further increased, an overall red shift of approximately 200 µeV is seen. A similar red shift is seen for the multi-excitonic lines. The blue shift can be attributed to a repulsive interaction related to the Pauli Exclusion Principle governing the behavior of the electrons and holes which comprise the excitons, and is a manifestation of the one dimensional nature of the linearly-aligned excitons in these QDs. The red shift can be attributed to an attractive Van der Waals interaction between confined excitons, which increases as the exciton density increases. The anisotropic confinement of excitons is probed by the magnetic field, excitation density, and photoluminescence (PL) polarization dependence of the single QD micro-PL spectrum. It is found that the anisotropic confinement, as well as the large QD size, are responsible for this lack of balance between repulsive forces due to the Pauli Exclusion Principle and van der Waals attractive forces.

Keywords: Molecular beam epitaxy; Self-assembly; Gallium arsenide; Nano-particles, quantum dots, and supra-molecules; Quantum effects; micro-Photoluminescence; Anisotropic confinement; Excitons

I. INTRODUCTION

Single GaAs/Al$_{0.405}$Ga$_{0.595}$As quantum dots (QDs) with anisotropic shapes made by Modified Droplet Epitaxy (MDE) are studied using micro-photoluminescence (micro-PL). Here, the shape of the QD under investigation departs from that of the previously reported, pyramidal-shaped GaAs/AlGaAs QDs, to that of an anisotropic, quantum wire-like shape. The difference in shape is expected to give a different confinement potential and different carrier correlation effects. Furthermore, MDE makes possible the growth of nanostructures using lattice-matched materials. Thus, strain, which is inevitably introduced in the conventional Stranski-Krastanov QD growth mode, does not affect the confinement potential. Therefore, the confinement potential of these GaAs QDs are defined predominantly by the shape of the nanostructure.

The shifts of the very sharp luminescence lines from a single GaAs anisotropic QD with increasing excitation density are seen. These sharp lines correspond to the recombination of excitons localized in an anisotropic QD. The evolution of the fine-structured photoluminescence (PL) spectra with increasing exciton density is interpreted in terms of many-body effects which lead to an increase of the exciton self-energy and to the formation of excitonic complexes.

II. EXPERIMENTAL

A. GaAs/Al$_{0.405}$Ga$_{0.595}$As QD sample

The sample used here was grown by MDE using a Riber-32P molecular beam epitaxy (MBE) system with elemental sources and an EPI (Veeco)-valved As cracking source, which enables the rapid irradiation of an accurately controlled As$_4$ flux. After native oxide desorption, a 300 nm-thick GaAs buffer layer and a 500 nm-thick Al$_{0.405}$Ga$_{0.595}$As barrier are grown on a GaAs (001) wafer at 580°C. The substrate temperature is then reduced to 330°C. The As supply is stopped, and a Ga supply of 1 ML is grown, according to enhanced epitaxy to avoid the two-dimensional regrowth of the QDs. The substrate temperature is lowered to 150°C, and these droplets are crystallized by the irradiation of a high As$_4$ flux. QDs with an average height of 32 (±7) nm, an average base size of 48 (±8) nm in the [011] direction, 55 (±9) nm in the [011] direction, and a density of approximately 2×10$^8$ cm$^{-2}$ is grown, according to atomic force microscopy (AFM) measurements. The size of this QD is relatively large. The substrate temperature is raised to 200°C, and a 10 nm-thick Al$_{0.405}$Ga$_{0.595}$As barrier layer is grown over the QDs using migration enhanced epitaxy to avoid the two-dimensional regrowth of the QDs. The temperature is raised to 580°C, and a 90 nm-thick Al$_{0.405}$Ga$_{0.595}$As barrier and a 10 nm-thick GaAs capping layer are grown by standard MBE. The sample is consecutively annealed at 680°C for one hour.
FIG. 1: Single anisotropic QD micro-PL spectrum excitation density dependence; the estimated values of the carrier populations in the dot for each excitation density are given in parenthesis next to the excitation density value.

No two-dimensional underlying GaAs layer exists, because any existing GaAs monolayer washes out during the annealing process, as calculated by Sanguinetti, et. al.[3]. A relatively large-sized, anisotropic QD was formed instead of a pyramidal QD, judging from AFM studies of the sample. The slight difference in the time between droplet formation and crystallization most probably played an important role in the definition of the QD shape. Also, the formation of relatively large Ga droplets, a tendency seen in very low density droplet formation conditions, contributed to the increased anisotropic diffusion of Ga from the Ga droplet during crystallization. This is an issue which requires further inspection.

III. RESULTS AND DISCUSSION

A. Zeeman splitting

The Zeeman splitting of the discrete energy levels in a single anisotropic QD, deduced from the $\sigma^+$- and $\sigma^-$-polarized spectral peaks’ positions at 5T, is negligible, and it can be said that the confinement by the anisotropic confinement potential is stronger than the confinement induced by the magnetic field. Pyramidal GaAs/AlGaAs QDs made by MDE have been shown to have a Zeeman splitting of 0.3meV at 5T[5]. This reflects the strong anisotropy of the anisotropic QD.

B. Excitation density dependence

An example of the single anisotropic QD micro-PL spectrum-excitation density dependence is shown in figure 1. The spectra are vertically offset for clarity. The estimated values of the carrier populations in the dot for each excitation density are evaluated using the carrier-capturing cross section value determined for similar GaAs QDs made by MDE[4]. These spectra are taken at 10 K, with 0.1 meV monochromator resolution.

Fine structured PL spectra-evolution with increasing excitation density is seen, due to the exchange interaction energies of the carriers[6]. These are multi-excitonic lines which are characteristic of the discrete energy levels of the QDs, and shows that the structure is essentially QD-like in nature. The appearance of the biexciton peak (peak B) at 88 W/cm$^2$ is in general agreement with the estimated carrier population at that excitation density (2). However, a distinct difference is seen in the peak positions with increasing excitation density, a phenomenon not seen in pyramidal QDs. In the pyramidal QDs, no shift of the spectral lines are seen with increasing excitation density[7]; in the anisotropic QDs, however, a clear shift of the spectral lines are seen with increasing excitation density. To clarify this shift, the amount of shift relative to the peak position as it initially appears is plotted against the excitation density in figure 2. The separate
peaks are labeled in figure 1.

An initial blue shift of approximately 100 μeV is seen for the main peak (labeled A) only. With higher increasing excitation density, however, the multi-excitonic lines (labeled B, C, D, E, and F) show an overall red shift of approximately 200 μeV; the main peak also shows such a red shift eventually. The initial blue shift most probably results from the mutual exciton-exciton interactions, leading to a renormalization of the exciton self-energy[8, 9]. Here, the sample is still in an electronically highly excited state, but with the electron-hole pairs in the form of a dense gas of excitons. However, the long-range direct Coulomb correlations of a given exciton with other neutral bound pairs, which give rise to the screening and correspond to the usual van der Waals attraction in an exciton gas, are very small effects that even in three dimensions are dominated by the short-range exchange effects. These short range exchange effects, or the repulsion due to Pauli’s exclusion principle, dominates over the screening which is responsible for the red shift[10]. In bulk semiconductors, these two effects cancel each other out exactly, and a constant exciton recombination energy at increasing exciton density is expected[11]. In this case, the initial blue shift is seen due to the repulsive inter-particle interaction related to the Pauli exclusion principle governing the electrons and holes forming the exciton, which is increased by the decreased dimensionality of the system; this is further enhanced by the one dimensional nature of the linearly-aligned excitons in these anisotropic QDs[9, 12]. The existence of a permanent dipole between an electron and a hole in their ground state, which is caused by the anisotropic confinement potential inherent to the shape of the anisotropic QD, cannot be ignored. Thus, while there are, on average, a few excitons confined in the anisotropic QD, a blue shift of the main peak is seen. Band filling is not included in this interpretation of the blue shift, due to its higher saturation density for excitonic absorption in low-dimensional systems[10]. As the average exciton number increases, the van der Waals attractive forces become stronger due to the weak confinement of this relatively large anisotropic QD, resulting in the redshift of the multi-excitonic lines. It is considered that the many exciton system does not form into a plasma state in this excitation density range.

To summarize, these shifts are ascribed to the lack of balance between repulsive forces due to Pauli’s exclusion principle in an exciton gas, caused by the anisotropic shape of the confinement potential, and van der Waals attractive forces, caused by the large size of the anisotropic QD.

C. PL polarization

The polarization of the PL is studied using a linear polarizer placed in front of the monochromator slit. This simple setup is used, because the polarization of the excitation light is not selected. The spectral dependence on the observed linear polarization is shown in figure 3. The intensities are shown in the logarithmic scale, in order to emphasize the peaks with small intensities. The angle of the linear polarizer is determined by rotating the polarizer and taking the angle which produces the largest observable change as 90°, and then taking the spectra while rotating the polarizer by 30°.

From figure 3, it can be seen that the spectral peak shifts originate from a combination of two perpendicularly polarized PL spectra. The PL spectra from this anisotropic QD consists of two perpendicular spectral components which most likely correspond in direction to the [011] and [01T] directions of the sample surface. The pyramidal QDs, measured at the same time with the same setup, show no such feature; thus this feature is characteristic of anisotropic QDs.

Figure 4 shows the spectral peak energy dependence on the observed linear polarization. From this it is clear that the peak labeled “B” consists of two discrete peaks, which are mostly overlapped, resulting in a broader peak.

These results show the anisotropy of the confinement potential, which gives a different spectra for different con-
finement directions. This is a result of the lifting of the degeneracy in the confinement directions.

IV. CONCLUSION

The anisotropic confinement potential of the QD is probed by single QD micro-PL. It is found that the anisotropy of the confinement potential, as well as its relatively large size, are responsible for the lack of balance between repulsive forces due to Pauli’s exclusion principle in an exciton gas and van der Waals attractive forces. Furthermore, it can be said that the degeneracy of the confinement directions are lifted, and its effects are apparent in the difference in the degree of the polarization of the PL, reflecting the anisotropy of the QD shape.

It has been reported that, for similarly shaped CdSe/ZnS nanorods, the one-dimensional exciton ground state changes from the optically forbidden (“dark”) state to the optically active state as the size is reduced[13]. Thus, future work includes the determination of the degree of linear polarization of the PL, as well as the QD size dependence of the PL decay times.

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