Non-resonant tunneling in single pairs of vertically stacked asymmetric InP/GaInP quantum dots

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Available online 18 September 2007

Abstract

In this work, single vertically stacked asymmetric InP/GaInP quantum dot (QD) pairs that are separated by different barrier widths have been investigated. We have found that for large (20 nm) inter-dot distances no tunneling is possible, that for medium spacer widths (10 nm) electrons can tunnel from the large dot to the small dot, and that finally, for very small (5 nm) barriers both electrons and holes can tunnel. We have simulated our results using a rate-equation model and have found a good agreement between simulation and experiment.

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PACS: 73.40.Gk; 78.67.Hc

Keywords: Asymmetric quantum dot pairs; Non-resonant tunneling

Coupled quantum dots (QDs) are interesting candidates for future devices, such as, quantum gates for quantum computers. Studies performed to date have mainly concentrated on the coupling mechanisms and optical properties\cite{1,2,3} of nearly symmetric QD pairs. However, as QD pairs inherently exhibit size differences between the individual dots, it is also important to study the effects of this asymmetry. The QD size asymmetry primarily enables uni-directional tunneling of carriers, probably via phonon emission, from one dot to the other, as the ground state energy is different for the two dots. Up to now only a few reports have concentrated on asymmetric QD pairs (AQDP), for example, tunneling times have been determined from ensemble measurements\cite{4,5} and on a single dot level anticrossings have been observed by tuning the electron and hole levels in single InAs/GaAs AQDP\cite{6} into resonance using a static electric field.

The samples were grown by metal-organic vapor phase epitaxy on (1 0 0) GaAs substrates. The self-assembled lower InP dots were grown on a 430 nm Ga\textsubscript{0.51}In\textsubscript{0.49}P barrier layer and overgrown by a GaInP spacer layer, with different thicknesses of 5, 10, and 20 nm depending on the sample. The upper InP QD layer was then grown using only 75\% of the material quantity that was used to produce the lower QD layer. In this way, we could, on average, produce smaller upper dots that consequently emit at higher energies than the lower dots. Finally, a 30 nm GaInP cap was deposited for photoluminescence (PL) measurements. See Ref.\cite{7} for details. Due to strain effects the upper dots grow above the lower dots in the vast majority of cases\cite{8}. The samples were then finally structured into mesas with a diameter of 200 nm by electron-beam lithography and dry etching, in order to address single QD pairs via micro-PL.

In order to perform PL experiments at 4 K, the sample was mounted in a He-flow cryostat and excited by a frequency doubled pulsed Ti–Sapphire laser operating at 410 nm (3.02 eV) and at a repetition rate of 76.2 MHz. The light from the sample was dispersed in a 0.75 m spectrometer and either detected using a liquid nitrogen-cooled charge-coupled device camera or sent to a Hanbury-Brown and Twiss type setup in order to perform autocorrelation measurements (see Ref.\cite{9} for details).

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doi:10.1016/j.physe.2007.09.015
In Fig. 1 we present the results from power-dependent PL measurements. For the sample with a 20 nm spacer between the dots (Fig. 1(a)) the ratio between the intensity from the small dot (SD) and the intensity from the energetically lower-lying large dot (LD) remains approximately constant for all of the investigated excitation power densities (PD). In contrast for the 10 nm spacer (Fig. 1(b)) and the 5 nm spacer (Fig. 1(c)), the PL emission from the SD appears only at elevated PD. Furthermore, additional lines are observed in the case of the 10 nm spacer. In addition, at elevated PD, emission from the first excited state (P) is seen.

These findings can be explained by inter-dot carrier tunneling effects. For an inter-dot distance of 20 nm no tunneling is possible, as the barrier thickness is too large. Therefore, the emission from the AQDP is from two independent dots. We note that for these dots there is mainly emission from the neutral biexciton (XX) and only weak emission from the neutral exciton. This has been corroborated by the superlinear and the linear rise of the intensity of the biexciton and the exciton, respectively, with PD in single dot measurements [9]. This may be attributed to the influence of the exciton dark state which will be the subject of an upcoming publication.

On the other hand, for the 10 nm spacer the barrier is small enough to allow electrons to tunnel from the SD to the LD, while hole tunneling is prevented due to the larger hole effective mass. For small PD excitons primarily radiatively recombine in the LD as the electrons can tunnel out of the SD. It is therefore only at elevated PD, when tunneling is suppressed by Pauli blocking in the LD, that emission from the SD emerges. The appearance of double peaks may be explained by the quantum confined Stark effect arising from the electric field, that builds up because of the charge in the neighbouring QD, when an electron has tunneled from the SD to the LD [10]. An alternative explanation would be the appearance of charged excitons caused by electron tunneling; however, in this case one would normally expect the Coulomb related energy shift to be somewhat larger than the one observed here.

Ultimately, in the case of the 5 nm spacer the barrier is now small enough to allow both electrons and holes to tunnel from the SD to the LD and consequently no electric field can develop. Therefore, a single line from the LD is observed once again at small PD. Emission from SD is only observed at high PD when tunneling is suppressed due to Pauli blocking in LD, as evidenced by the simultaneous observation of LD p-shell emission.

The intensity ratio (intensity SD/intensity LD) has been analyzed and is shown in Fig. 2(a). The ratio has been normalized to 1 for the highest PD considering that the two dots have different carrier capture rates. One observes that the ratio is constant for the 20 nm spacer, while it increases distinctly for the 10 nm spacer and increases even further for the 5 nm spacer. This behavior has been modeled using a simple rate-equation model following that given in Ref. [5]:

\[
\frac{dn_{SD}(t)}{dt} = -\frac{n_{SD}(t)}{\tau_{SD}} - \frac{n_{SD}(t)[1 - n_{LD}(t)]}{\tau_{T}} + G_{SD}[1 - n_{SD}(t)],
\]

\[
\frac{dn_{LD}(t)}{dt} = -\frac{n_{LD}(t)}{\tau_{LD}} + \frac{n_{SD}(t)[1 - n_{LD}(t)]}{\tau_{T}} + G_{LD}[1 - n_{LD}(t)]
\]

(1)

with \(n_{SD}(t)\) and \(n_{LD}(t)\) representing the occupation of the lowest states of the SD and the LD, respectively, \(G_{SD}\) and \(G_{LD}\) are the respective exciton generation rates, \(\tau_{SD}\) and \(\tau_{LD}\) are the respective radiative recombination times, and \(\tau_{T}\) is the inter-dot tunneling time. We used equal generation rates and a radiative recombination time of 600 ps for both dots, that is the average decay time obtained from single dot PL measurements at 4 K. The tunneling times were set to 100 ps, 1.5 ns and 10 ns to qualitatively monitor the influence of this time on the intensity ratio. The results are shown in Fig. 2(b) and a

![Fig. 1. Power-dependent PL spectra of an AQDP for (a) a 20 nm spacer, (b) a 10 nm spacer, (c) a 5 nm spacer.](image)
good agreement with the experimentally measured values is obtained.

To confirm the zero-dimensional nature of the system we also performed autocorrelation measurements and the results are shown in Fig. 3 for the three AQDP samples that have been investigated. We found a decrease in multiphoton-pulses by a factor of two or more (up to 10) compared to a Poissonian source with the same average intensity. This indicates that the QDs are capable of emitting triggered single photons.

In conclusion, we have shown that in AQDPs the barrier width strongly influences the tunneling dynamics of the system. We have found a transition from a regime with no carrier tunneling for large (20 nm) inter-dot distance to a regime with electrons tunneling for intermediate (10 nm) inter-dot separations and finally to a regime where both electrons and holes can tunnel for small (5 nm) barrier width. The intensity ratio between LD and SD has been analyzed and the different behavior for varying inter-dot separations could be explained using a rate-equation model. Finally, we have shown that the AQDP are comprised of two single QDs that can both provide triggered single-photon emission.

References