Nonresonant tunneling in single asymmetric pairs of vertically stacked InP quantum dots

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Single pairs of vertically stacked asymmetric pairs of InP quantum dots embedded in GaInP barriers have been investigated as a function of interdot spacer thickness. Time integrated and time-resolved photoluminescence measurements have been performed, with the former showing a change in the intensity ratio between the two dots and the latter an increasing difference in the photoluminescence decay time of the two dots when reducing the spacer thickness. Hence, we suggest transitions from vanishing tunnel coupling to electron tunneling and, finally, to electron and hole tunneling for decreasing barrier widths. The different times are estimated from the measurement data, and the changes are described by a rate equation model. The results clearly show the nonresonant character of the tunneling process as a result of the different ground state energies (approximately 40 meV) of the unequally sized dots.

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In recent years, a lot of progress has been made in the growth and understanding of quantum dots (QDs),1 for example, single QDs have been shown to be capable of emitting single photons,2 indistinguishable photons,3 and entangled photons.4,5 However, for more complex devices, the energetically higher-lying levels of the small dot usually not aligned, and resonant tunneling is prevented. Different for the two dots. Therefore, the electronic levels are concentrated on asymmetric quantum dot pairs (AQDPs). In these reports, the main objective was to determine interdot tunneling times,10–12 which give some useful insights into the principal interdot tunneling mechanisms; however, as ensembles of QDs were investigated, inhomogeneous broadening due to QD size fluctuations prevents the observation of single pair effects. Recently, however, single vertical InAs/GaAs AQDPs have been investigated, and clear anticrossings were observed when the electron and hole levels were tuned into resonance.13,14

As QD pairs inherently exhibit size differences between the individual dots, it is important to study the effects of this asymmetry. Such asymmetry may be advantageous in future applications, such as tunneling devices or novel laser devices.15 In addition, such pairs allow for the study of basic physical properties, such as interdot tunneling mechanisms, as the interdot distance, a precisely adjustable parameter, and the degree of asymmetry can be easily controlled during growth. Schematics of the sample structure and the resulting band structure are presented in Fig. 1. The QD size asymmetry primarily enables unidirectional tunneling of carriers from one dot to the other, as the ground state energy is different for the two dots. Therefore, the electronic levels are usually not aligned, and resonant tunneling is prevented. Nevertheless, it is still possible for carriers to tunnel out of the energetically higher-lying levels of the small dot (SD) into the lower-lying states of the large dot (LD) via phonon emission.16,17

The samples were grown by metal-organic vapor phase epitaxy on (100) GaAs substrates oriented by 6° toward the [111]A direction. The lower dots were grown using the Stranski-Krastanow growth mode by depositing 2.1 monolayers of InP on a 430 nm GaAs1.51In0.49P barrier layer and were overgrown by a GaInP spacer layer, with different thicknesses of 2.5, 5, 10, and 20 nm depending on the sample. The upper QD layer was then grown using 75% of the material 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 layer of dots. Finally, a 30 nm GaInP cap was deposited on top for photoluminescence (PL) measurements (see Ref. 18 for details). Due to strain effects, the top dots grow above the bottom dots in the vast majority of cases.19 Finally, the samples were structured into mesas of 200 nm diameter by electron-beam lithography and dry etching in order to address single QD pairs.

In order to perform PL experiments at 4 K, the sample was mounted in a He-flow cryostat and excited by a frequency doubled Ti:sapphire laser operating at 410 nm (3.02 eV) with a repetition rate of 76.2 MHz. The light was focused on the sample using a 50× microscope objective down to a spot diameter of approximately 1 μm. The PL from the sample was then collected by the same objective and dispersed by a 0.75 m spectrometer. Afterwards the PL signal was detected either by two avalanche photodiodes (APDs) in a Hanbury-Brown and Twiss type setup when carrying out autocorrelation measure-
ments, or (3) an APD with a high temporal resolution when performing time-correlated single-photon-counting measurements. An ultimate temporal resolution of 35 ps could be achieved using the latter setup.

In Fig. 2, we present the PL emitted from typical AQDPs with different spacer widths. We found that the QDs emit approximately 30 meV higher in energy than one would expect for single dots grown under these conditions; this difference is most likely due to the altered strain situation. For each spacer thickness, at least five pairs were carefully examined, and the representative results presented in this paper could be repeatedly observed. Distinct differences between the samples with different spacer layers have been found. For a 20 nm spacer layer [Fig. 2(a)], an independent behavior of both dots is apparent. Both dots exhibit similar intensities and emission of biexcitonic luminescence. The weakness or absence of an exciton related transition has been almost invariably observed and is thought to be due to the strong influence of an energetically lower-lying exciton dark state in such InP/GaInP QDs, which will be discussed elsewhere.

At high powers, an additional recombination line from the p shell is observed in the LD spectrum. The spectra change dramatically when reducing the barrier width. For example, for a 10 nm spacer layer at the lowest measure power density [Fig. 2(b)] the emission of the LD is much more intense than that of the SD (by a factor of 15). In general, at low power densities, the LD dominates the spectrum, with the associated emission consisting primarily of three transitions (1.8346, 1.8352, and 1.8363 eV). With increasing excitation power, the LD lower energy line (1.8346 eV) gains intensity compared to the LD higher energy line (1.8363 eV). Meanwhile, the emission from the SD is split into a doublet (1.8798 and 18809 eV) as the intensity of the SD low-energy transition (1.8798 eV) increases with increasing power compared to the SD high-energy line (1.8809 eV). The splitting of the lines also observed from other similar QD pairs ranges between 0.7 and 2.5 meV, values that are clearly smaller than the typical QD exciton-biexciton splitting (4.5–5.5 meV) for this material system. Therefore, these lines may result from charged exciton transitions. From these observations, we suggest the following scenario. The 10 nm barrier width is sufficiently small to allow electrons to tunnel from the SD to the LD, while hole tunneling, on the other hand, is still prevented due to the larger hole effective mass \( m_e \). An estimation of the interdot transfer times will be given later in this work. The electron tunneling leaves holes or positively charged excitons in the SD and leads to electrons or negatively charged excitons in the LD. The high-energy lines are, therefore, probably due to negatively (positively) charged transitions in the LD (SD). Finally, at high powers, as the LD ground state becomes saturated, Pauli blocking suppresses electron tunneling and the neutral transitions from both dots can finally be observed. An alternative explanation for the appearance of double peaks may be the quantum confined Stark effect arising from the electric field, that builds up, when an electron tunnels from the SD to the LD. However, for a more rigorous peak assignment, a more detailed theoretical investigation is necessary.

Yet another regime is observed for very small barriers (2.5 nm spacer), as shown in Fig. 2(c). Now single lines are visible once again. For small and medium excitation powers, only the recombination lines of the exciton (X) and the biexciton (XX) from the LD are visible, and it is only at very high excitation powers that emission from the SD emerges. The observation of emission lines originating solely from neutral excitonic transitions in the LD at moderate excitation powers can be explained by assuming that the hole tunneling time is now also faster than the excitonic radiative recombination time. In other words, both electrons and holes can now tunnel (estimation of hole/electron tunneling time ratio is presented later). Therefore, emission from the SD is not observed. It is only at very high excitation powers, when the LD ground state is already saturated and interdot tunneling is blocked due to state filling effects, that emission from the SD may be observed. For a 5 nm spacer layer, a similar behavior was apparent (not shown here), with the exception that stronger PL emission from the SD was observed for a given power density, as the tunneling rates are lower in this case.

To lend further weight to the proposed tunneling mechanisms outlined above, the intensity ratio \( I_{SD}/I_{LD} \) was analyzed and is plotted in Fig. 3(a). For the 20 nm spacer, the intensity ratio stays nearly constant over the investigated power range, implying that no tunneling is present. As ex-
expected, the situation changes for the 10 nm spacer and the intensity ratio increases by a factor of approximately 2 over the 2 orders of magnitude change in power. However, an even stronger increase of the intensity ratio was found for the smallest barriers, with the value increasing by more than 1 order of magnitude over the investigated power range. To model the underlying processes, a simple rate equation model was used,\textsuperscript{12}

\begin{equation}
\begin{align*}
\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)],
\end{align*}
\end{equation}

where \( n_{SD}(t) \) and \( n_{LD}(t) \) represent the occupation of the lowest states of the SD and the LD, respectively, \( G_{SD} \) and \( G_{LD} \) are the respective generation rates, \( \tau_{SD} \) and \( \tau_{LD} \) are the respective radiative recombination times, and \( \tau_{T} \) is the interdot tunneling time. Equal generation rates are used and the radiative recombination times are fixed to a value of 600 ps, the average value obtained from single dot PL measurements. We chose strongly different tunneling times of 100 ps, 1.5 ns, and 10 ns to qualitatively monitor the differences for a regime of strong tunneling, medium tunneling, and weak tunneling, respectively. The resulting curves of the intensity ratio for the steady state case \( n_{SD}/n_{SD} \) are shown in Fig. 3(b). As the absolute values depend on the particular dot, the results are not expected to reproduce the exact measurement data; however, a qualitative explanation for the behavior of the intensity ratio for different tunneling times can be obtained. Please note that the tunneling times that were used to solve the rate equation model have not been extracted from fits to the data, and are different from the transfer times calculated later from the time-resolved PL measurements. The results indicate that for a long tunneling time (10 ns) the intensity ratio stays nearly constant, for a medium tunneling time (1.5 ns) the ratio doubles, and for a relatively fast tunneling time (100 ps) the intensity ratio increases strongly (by 1 order of magnitude). Also, the shape of the curve (first a constant ratio then an increase) could be simulated. For the 10 nm spacer, it is probable that only the increase in the medium power range was measured over the 2 orders of magnitude excitation power range that was investigated.

Time-resolved measurements are a powerful tool that can help us to further understand the dynamics of the tunneling processes. The results of such measurements are shown in Fig. 4 together with a simulation. For the 20 nm spacing [Fig. 4(a)], the LD and SD transients are very similar. In contrast, for the 10 nm spacer [Fig. 4(b)], although both dots still reach their maximum at a similar time, the decay of the SD PL signal is now faster (290 ps) than that of the LD (390 ps). We propose that this behavior indicates the presence of an extra decay channel for the SD, namely, carrier tunneling into the LD. As expected, the differences are even more pronounced for the 2.5 nm spacer (LD: 570 ps, SD: 140 ps) [Fig. 4(c)]. In this case, the emission from the SD also reaches a maximum earlier than that of the LD. Once again, this can be explained by the influence of a delayed carrier filling of the LD by the SD. The difference of the decay time is much more pronounced for such small barriers as a result of the increased tunneling rate.

This temporal behavior was modeled using the rate equation outlined above, with the radiative recombination times \( \tau_{LD} \) and \( \tau_{SD} \) set to 600 ps and the generation rates \( G_{LD} = G_{SD} = G(t) \) described using a monoeponential decay function with a time constant of 5 ps to represent the carrier capture into the dots under pulsed excitation conditions [Fig. 4(d)]. All time constants, including the tunneling times, are the same as those used in the simulation of the intensity ratio. Please note that the results from the simulation have been convoluted with the instrument response function.

For long tunneling times (10 ns, dashed lines), the transients are very similar to those observed for the 20 nm spacer sample. For medium tunneling times (1.5 ns, dotted lines), a
TABLE I. Measured decay times $\tau_d$ and estimated transfer times $\tau_{TF}$ for the different barrier widths estimated from the spacer thickness as described in the text.

<table>
<thead>
<tr>
<th>Spacer</th>
<th>2.5 nm</th>
<th>5 nm</th>
<th>10 nm</th>
<th>20 nm</th>
</tr>
</thead>
<tbody>
<tr>
<td>Barrier</td>
<td>1.5 nm</td>
<td>3.5 nm</td>
<td>8 nm</td>
<td>18 nm</td>
</tr>
<tr>
<td>$\tau_d$ (ps)</td>
<td>280</td>
<td>400</td>
<td>500</td>
<td>570</td>
</tr>
<tr>
<td>$\tau_{TF}$ (ps)</td>
<td>525</td>
<td>1200</td>
<td>3000</td>
<td>11400</td>
</tr>
</tbody>
</table>

significant difference between the decay time of the two dots is apparent, in line with the difference observed in the measurements. Finally, for fast tunneling times (100 ps), the maximum of the emission from the LD is clearly delayed, and in addition, there is a strongly pronounced difference between the decay times, as experimentally observed in the PL dynamics. Therefore, both simulations are in good agreement with the associated experiments and thereby support the proposed tunneling scenarios.

By fitting the initial decay of the time-resolved PL measurements, it is possible to estimate the interdot transfer time of carriers from the SD to the LD. This time results from tunneling in the presence of Pauli-blocking effects, as tunneling can only occur if the final state is not populated. The transfer times $\tau_{TF}$ can be estimated from the measured decay time $\tau_d$ when the radiative recombination time $\tau_r$ of the SD is known, using the following relation:

$$\frac{1}{\tau_d} = \frac{1}{\tau_{TF}} + \frac{1}{\tau_r}.$$  

The results of these estimates are given in Table I together with the average of the associated measured decay times for different AQDPs.

We would like to emphasize at this point that the measured width of the spacer layer is not exactly equal to the actual tunnel barrier [Fig. 5, insets]. For larger spacers, the barrier is reduced by the dot height, which is approximately 2 nm. It is only for the smallest barriers presented here that the barrier is similar to the actual spacer thickness, as a bending of the upper wetting layer at the position of the bottom dot occurs, as has been observed for similarly grown stacked QDs in the transmission electron microscopy (TEM) measurements of Ref. 23. Please note that we can only give an estimate for the quantitative amount of bending here as no TEM images of these particular dots are available at present. Where necessary, the barrier width has been corrected prior to calculating the transfer times. The times obtained by Eq. (2) are in reasonable agreement with those extracted from QD ensemble measurements.10,12,16

As expected, one observes an exponential decrease of the estimated transfer time with decreasing barrier width, as shown in Fig. 5. In theory, tunneling rates should approach infinity for infinitesimal spacer widths; however, we observe an offset of approximately 425 ps. This offset is due to the aforementioned Pauli-blocking effects. The transfer times given in Table I represent the faster component of the tunneling process, thus implying that the electron transfer times have been estimated.

As it is not possible for us to distinguish between tunneling and Pauli blocking, it is difficult to calculate hole transfer times or hole tunneling times. However, it is possible to estimate the ratio of hole tunneling time $\tau_{T,h}$ to electron tunneling time $\tau_{T,e}$ by using the Wentzel-Kramer-Brillouin approximation,

$$\frac{\tau_{T,h}}{\tau_{T,e}} = \exp[2d\sqrt{\frac{2\hbar}{\pi}}(\frac{m^*_e}{m^*_h}\Delta V_h - \frac{m^*_e}{m^*_h}\Delta V_r)],$$

where $d$ is the tunneling barrier, $m^*_e$ and $m^*_h$ are the effective masses for the electron or heavy hole, and $\Delta V_h$ and $\Delta V_r$ are the respective band offsets. With $m^*_e = 0.105m_0$, $m^*_h = 0.48m_0$, $\Delta V_r = 70$ meV, and $\Delta V_h = 30$ meV, we estimate that the hole tunneling time is only a factor of 1.7 longer than the electron tunneling time for the 1.5 nm tunnel barrier; however, it is already a factor of 16 longer for the 8 nm tunnel barrier. This estimation clearly shows that hole tunneling is very improbable for large barriers, as expected. In contrast, holes can tunnel quite rapidly for very small barriers. This calculation, therefore, supports the assumption, deduced from the spectra, that electron tunneling occurs for medium barriers (10 nm) and that electron plus hole tunneling occurs for small barriers (<5 nm).

The slow transfer times highlight the nonresonant character of the tunneling process as one would expect values of a few picoseconds in the resonant case.25 Previously published reports argue that nonresonant tunneling occurs via LO-phonon emission for QDs as well as for asymmetric quantum well structures that have been studied in more detail to date. By coincidence, for the 10 and 2.5 nm spacer layers, the difference of the ground state emission energy of the two dots that we present here closely fits the energy of the LO phonon in GaInP, which is 46 meV. We do, however, also observe the effects mentioned in this paper for smaller energy differences. Therefore, we propose that phonons other
than the LO phonon may also be responsible for the energy loss in such a nonresonant tunneling process.

In summary, we have investigated the carrier tunneling in asymmetric quantum dots as a function of interdot barrier distance in detail. We have found transitions from regimes of no tunneling, to electron tunneling, and finally, to electron and hole tunneling with ever decreasing barrier width and have observed characteristic spectra that clearly identify each regime. The intensity ratio and the time-resolved PL measurements have been simulated by a simple rate equation model, and good agreement between simulation and experiment is obtained. In addition, we have extracted transfer times by fitting the time-resolved PL data and have found that the observed transfer times are approximately 525 ps for the smallest investigated spacer (2.5 nm). As a result of the size asymmetry of the QD pairs, the tunneling process is of nonresonant nature, and is proposed to be phonon assisted.

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