ANOMALOUS REAL SPACE CHARGE TRANSFER THROUGH THICK BARRIERS IN GaAs/Al<sub>x</sub>Ga<sub>1-x</sub>As ASYMMETRIC DOUBLE QUANTUM WELLS: Al<sub>x</sub>Ga<sub>1-x</sub>As AS A PERCOLATING BARRIER


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Anomalously large real space charge transfer through thick barriers in GaAs asymmetric double quantum wells is studied. This inter-well exciton transfer is very large when the barrier is the Al<sub>x</sub>Ga<sub>1-x</sub>As alloy, but disappears when the barrier is an equivalent GaAs/AlAs digital alloy. These results combined with observed x and barrier thickness dependence suggest that the inhomogeneities in the barrier may be responsible for this transfer. This picture is supported by the quantum mechanical calculation in three dimensions. Copyright © 1996 Published by Elsevier Science Ltd

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Fig. 1. (a) PLE spectra obtained at 14 K for the GaAs/AlGaAs/GaAs (75 Å/300 Å/100 Å) ADQW, with \( x = 0.3 \) (top), and \( x = 0.5 \) (bottom). (b) PLE spectra at 14 K for the GaAs/GaAs digital alloy/GaAs ABQW (75 Å/300 Å/1000 Å) [top], and In\(_{0.15}\)Ga\(_{0.87}\)As/GaAs/GaAs [100 Å/300 Å/100 Å] ADQW (bottom). Next to each figure, the schematics of the sample structure are shown, with the dotted arrow indicating strong transfer, and the dotted arrow crossed out indicating much smaller transfer. SWHH denotes the heavy hole of the shallower well (In\(_{0.15}\)Ga\(_{0.87}\)As).

samples. The sample parameters are: GaAs/Al\(_{0.3}\)Ga\(_{0.7}\)As/GaAs (75 Å/300 Å/100 Å) for the top, and GaAs/Al\(_{0.5}\)Ga\(_{0.5}\)As/GaAs (75 Å/300 Å/100 Å) for the bottom. For \( x = 0.3 \), the NW peaks are pronounced, indicating a strong transfer from the NW to the WW. The height of the NW heavy-hole (HH) peak is significantly larger than the background due to the continuum excitation, and comparable to the WW light-hole (LH) peak. The experiments were performed at low excitation density, so that intensity dependent nonlinear effects can be ruled out.

When there exists strong transfer from the NW to the WW (Fig. 1a, top), the amount of electron-hole pairs from the NW that eventually end up in the WW can be estimated in the following simple way: when the laser photon energy is at the NWHH, both the NW exciton and the WW continuum are excited. Since the WW luminescence is enhanced by a factor of 2 when the NWHH is resonantly excited, it can be estimated that nearly half of the electron-hole pairs in the WW originate from the NW. From this and the PLE spectra with the low energy tail of the NW luminescence as a window, we can easily estimate that up to 30% of the resonantly excited NW excitons eventually end up in the WW. We can also divide the area under the NWHH peak by that of the WWLH peak [11], and come up with \( \approx 30\% \) of the transfer efficiency \( T \). With this eventual \( T \) and life time of excitons (100 ps–1 ns; [12]), we can estimate the transfer coefficient per single trial \( t \) as \( t \sim T/N \), where \( N \) is the number of round trips of holes during the exciton life time. Holes were used rather than electrons, since in general, transfer of holes is slower. \( t \) estimated in this way is of the order of \( 10^{-3} - 10^{-4} \), which is at least 10 orders of magnitude greater than the tunneling coefficient over the one-dimensional mean field barrier.

In Fig. 1(b), a more striking example of the complete failure of the mean field approach is shown. PLE spectra of a GaAs ADQW with a digital alloy barrier (top), and a shallow In\(_{0.15}\)Ga\(_{0.87}\)As/GaAs ADQW are shown. The digital alloy sample was chosen so that the effective alloy concentration is 0.28, and \( d \) was kept at 300 Å. For the digital alloy barrier, which is roughly equivalent to 100 Å of AlAs, the mean field theory would predict a larger transfer coefficient than for the 300 Å Al\(_{0.3}\)Ga\(_{0.7}\)As alloy barrier. Likewise, the exciton transfer of the InGaAs/GaAs ADQW should be much larger than the GaAs ADQW with \( x = 0.3 \) since the wells are much shallower. The absence of NW peaks in the PLE spectra of the digital alloy ADQW, or the weak shallower well (SW) HH peak in the In\(_{0.15}\)Ga\(_{0.87}\)As/GaAs ADQW tells exactly the opposite story. The absence of the exciton transfer when the barrier is GaAs or AlAs/GaAs digital alloy strongly suggests that this anomalous transfer is a result of the alloy nature of the barrier. Al\(_{0.5}\)Ga\(_{0.5}\), being a substitutional alloy, has intrinsic spatial fluctuation of atomic arrangement. Therefore, we contend that a large spatial variation of the alloy potential barrier is responsible for this puzzling phenomenon. Since the order parameter or the size of the fluctuations in alloys are often strong functions of \( x \), this picture is consistent with the observed sharp decrease between \( x = 0.3 \) and \( x = 0.5 \).

To investigate the \( x \)-dependence of the transfer more systematically, we studied many GaAs/Al\(_{1-x}\)Ga\(_{x}\)As ADQW samples with varying \( x \). In Fig. 2, \( T \) is plotted against \( x \), where a sharp decrease is observed around
Fig. 2. Transfer efficiency $T$ at 14 K defined as the ratio of the areas under the NWHH to the WWHH peaks in the PLE spectra, plotted against $x$ for GaAs/Al$_x$Ga$_{1-x}$As/GaAs ADQW (75 Å/300 Å/100 Å). The efficiency was averaged over several different spots.

$x = 0.3$. From this $x$-dependence, photon-reabsorption by the WW can be safely ruled out since the barrier region is always transparent at the photon energies used. It is interesting to note that this “critical $x$” is close to the direct-to-indirect cross over ($x \approx 0.35$). With the exciting photon energies well below the threshold for the threshold for the intervalley transfer, the real space charge transfer [13] is not directly relevant. Furthermore, the issues of type II superlattices or the barrier-confined states [14] do not apply here because of relatively large well thickness. On the other hand, a deeper understanding of the direct-to-indirect crossover in Al$_x$Ga$_{1-x}$As alloy in terms of order parameters or cluster sizes [4] might prove very useful. Finally, we note that even for large $x$, there exists a significant $T \approx 5\%$, which is roughly independent of $x$. This transfer might be due to photon reabsorption or coherent polariton transfer [6–10], processes that are thought to be largely independent of $x$ and $d$.

Since significant transfer persists up to $d = 300$ Å, it is clear that the $d$-dependence is much weaker than what the one-dimensional tunneling model would predict. To see this more closely, we studied the $d$-dependence of the transfer as a function of $d$ for a fixed $x = 0.3$. In Fig. 3, the $d$-dependence of $T$ in GaAs/AlGaAs ADQW is shown for $x = 0.3$ (open circle), along with that for $x = 1$ and $d = 300$ Å (closed circle). The $d$-dependence at $x = 0.3$ is weak, and even at $d = 1500$ Å, $T$ is still greater than that of $x = 1$ and $d = 300$ Å. We now discuss model calculations that emphasize the enormous enhancement of the transmission with the increasing size of clustering.

In the mean field approach, the barrier height in a GaAs/Al$_x$Ga$_{1-x}$As superlattice is assumed to be a constant determined by $x$. On the other hand, recent scanning tunneling microscopy (STM) studies of GaAs/Al$_x$Ga$_{1-x}$As quantum wells, AlAs/GaAs superlattices, and Al$_x$Ga$_{1-x}$As alloys [15] show that there exists elongated “clustering” of Ga rich and Al rich regions in the barrier along the growth direction, thus possibly connecting adjacent GaAs wells. We first considered the effect of atomic scale fluctuations on $t$. We divided the barrier into small cubes of atomic scale representing GaAs or AlAs molecules, and randomly assigned either the potential $V$ for AlAs or 0 for GaAs. $V$ is 1.12 eV (0.26 eV) to simulate the band offsets for electrons (holes) (Fig. 4(a)). We solved the resulting three-dimensional effect mass equation with appropriate boundary conditions to obtain $t$. The resulting $t$ are larger than those obtained from the one-dimensional mean field approach, but still far to small to explain $t \approx 10^{-3} \sim 10^{-4}$ deduced from experiments. Essentially, the wavelength of the incoming waves (around 100 Å, comparable to the well size) is too large to “see” the low but narrow potential pathways.

We then replaced the cubes in the barrier region with rectangular cylinders (or “wires”) long enough to connect the two wells as shown in Fig. 4(b), simulating the possible aligning of GaAs or AlAs roughly along the
Fig. 4. (a) Schematics for the construction of barriers used in our model calculations assuming completely random, atomic alloy fluctuations. Dark squares represent AlAs "molecules". (b) Schematics for our model calculations taking into account the clustering and the formation of channels, and (c) The same as (b) except for the existence of "kinks". (d) $t$ using barriers described in (b), plotted against $x$ for several grid sizes. The incident wave simulates holes in the narrow well, with the effective mass of $-0.5m_e$, and the wavelength of 150 Å.

growth direction [15]. We performed the quantum mechanical calculation for various sizes of rectangles to study the cluster size effect on $t$. Furthermore, the possible effect of "kinks" was considered (Fig. 4(c)) in connection with [15] where the GaAs "quantum wires" were shown to "zigzag" their ways through the barrier. In Fig. 4(d), $t$ of holes as a function of $x$ are plotted for several cluster sizes using the model of Fig. 4(b). Holes are considered because we assume that holes determine the transfer of excitons due to their slower transfer compared with that of electrons. For the grid size of 4 Å, the results are only slightly larger than the prediction of the mean field theory, despite the fact that there exists many low potential quantum wires in the barrier. The physics of this is the same as described earlier: the pathways are much narrower than the wavelength. Increasing the cluster size rapidly enhances $t$, so that for the cluster size of 30 Å, nearly all holes can pass through the barrier for relatively low $x$ during the exciton life time, while for $x > 0.5$, the transmission coefficient is too small to allow any significant transfer. Finally, the results using the model schematically described in Fig. 4(c) show that the effect of the "link" is to decrease $t$ only slightly without changing the overall trends.

Our model calculations suggest that most features of our experiments can be explained, at least qualitatively, if there were large enough clustering of GaAs in a quantum-wire-like fashion, to allow significant coupling between the two GaAs quantum wells. The results of anomalously large anti-Stokes charge transfer from the WW to the NW [6, 16] also support this picture. Although a more realistic approach to the detailed mechanism of clustering and the resulting structure and pattern formation would be much desired, our results imply that the clustering of GaAs in the alloy barrier is a likely source of the enormously enhanced interwell coupling.

In conclusion, we experimentally demonstrated that a mysteriously large transfer of excitons through thick barriers occurs when the barrier is composed of $\text{Al}_x\text{Ga}_{1-x}\text{As}$ alloy. Unlike "solid barriers" such as GaAs/AlAs digital alloys, $\text{Al}_x\text{Ga}_{1-x}\text{As}$ is essentially leaky, or percolating, especially for $x < 0.4$: there may exist regions of low potential in the barrier connecting two adjacent wells, which allow observed huge charge transfer. Our results show that beyond the widely used mean field approach, a three dimensional approach considering the detailed nature of the barrier such as clustering is needed to understand some of the important dynamics of semiconductor superlattices.

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