Anomalous Real Space Charge Transfer through Thick Barriers in GaAs/$Al_xGa_{1-x}As$ Asymmetric Double Quantum Wells: $Al_xGa_{1-x}As$ as a Percolating Barrier

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Abstract

Anomalously large real space charge transfer through thick barriers in GaAs asymmetric double quantum wells is studied by photoluminescence excitation. This inter-well excitonic transfer is very large when the barrier is the $Al_{0.3}Ga_{0.7}As$ alloy, but disappears when the barrier is GaAs/AlAs digital alloy with an equivalent Al concentration of 0.28. These results combined with the observed x and barrier thickness dependence suggest that the spatial fluctuation of the atomic arrangment of Ga and Al in the alloy may be responsible for this transfer. This picture is supported by the quantum mechanical calculation in three dimensions which takes into account the said fluctuation effects.

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Ever since the proposal by Esaki and Tsu [1] and subsequent realization of semiconductor superlattices and quantum wells, the overwhelming majority of the experimental results on alloy superlattices was analyzed using the mean field approach. This approach, which for the most part ignores the disorder and inhomogeneity of the alloy, has been enormously successful in dealing with such dynamic and static problems as the tunneling through thin barriers, energy levels, or the density of states. With this phenomenal success, the nonuniform atomic distribution of these important systems is often put aside. In statistical mechanics, general properties of disordered systems such as the scaling laws near the percolation threshold have been widely investigated [2], and have been applied to the transmission of light through random metal-insulator composites [3]. In semiconductor physics, approaches that fully account for the random fluctuations of the system have only recently begun to be applied, say, to the direct-to-indirect band gap transition in $Al_xGa_{1-x}As$ alloys [4]. Another suggestion on the possible failure of the mean field approach in alloy superlattices was made in the problem of the coherence length of optical phonons in alloys and alloy superlattices: the coherence length of LO phonons in $GaAs/Al_xGa_{1-x}As$ superlattices, deduced from picosecond Raman experiments, is a strong function of x, sharply dropping beyond $x \approx 0.3$ [5].

Recently, the existence of an anomalously large interwell exciton transfer over thick barriers in $GaAs/Al_xGa_{1-x}As$ asymmetric double quantum wells (ADQW) was called into attention [6], although the existence of this unexplained transfer has been observed by many groups for quite some time [7]. The experimentally deduced transfer coefficient was of the order of 10^{-3}, orders of magnitudes larger than the theoretical predictions based on the one-dimensional tunneling model for the given barrier width (d) and the average potential height. The experiments were performed at 2 K so that thermal excitations above the barrier

were ruled out. Reabsorption of the narrow well (NW) luminescence by the wide well (WW), dipole-dipole interaction of excitons, and polariton effects were proposed as possible explanations for this transfer [6-10].

In this Letter, an extensive study of this "mysterious" transfer in GaAs/$Al_xGa_{1-x}As$ ADQW grown by molecular beam epitaxy (MBE) is presented. Photoluminescence excitation (PLE) was performed in a series of samples with variable x and d. Most important observations we would like to report are as follows: (1) the strong x dependence of the transfer (Fig. 1a and 2), (2) drastic reduction in the transfer when the barrier is GaAs or a digital alloy (Fig. 1b), and (3) the very weak d-dependence of the transfer shown in Fig. 3. Our results demonstrate the importance of the nonuniformity of the alloy potential barrier in charge transfer as described in detail later.

In Fig. 1a, PLE spectra at 14 K obtained with a cw Ti:Sapphire laser, with the PL window on the low energy side of the WW exciton, are shown for two ADQW 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, and the PLE spectra are independent of laser intensity. Therefore, intensity dependent nonlinear effects can be immediately ruled out. For x=0.5 in Fig. 1a (bottom), there is virtually no sign of the NW peaks whose positions are indicated by arrows, which is of course what one would expect in the mean field approach.

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 WWHH peak [11], and come up with roughly the same number, ~30%. From the known life time of resonantly excited excitons [12] and the vertical velocities of the first quantized electrons and holes, we can estimate that the transfer coefficient is of the order of 10^{-2} to 10^{-3}, at least 10 orders of magnitude greater than the tunneling coefficient over the one-dimensional mean field barrier.

In Fig. 1b, 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 $In_xGa_{1-x}As/GaAs$ 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 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 $In_xGa_{1-x}As/GaAs$ 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. $Al_xGa_{1-x}As$, 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/$Al_xGa_{1-x}As$ ADQW samples with varying x. In Fig. 2, the normalized transfer efficiency obtained by normalizing the NWHH peak to the WWHH or to the background, is plotted against x. A sharp decrease is observed around 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~0.35). With the exciting photon energies well below 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 $Al_xGa_{1-x}As$ 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 transfer efficiency of ~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 the normalized transfer efficiency 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, it is still greater than that of x=1 and d=300 Å. The weak dependence on d suggests that the apparent penetration depth is of the order of 1000 Å! Since the tunneling coefficient would exponentially decrease with d, with its penetration depth of the order of 10 Å, this dependence is again suggestive of a transfer efficiency that is orders of magnitudes larger than the prediction of tunneling. We now discuss model calculations that take into account the effect of the detailed structure of the alloy barrier and the possible clustering of GaAs and AlAs "molecules".

In the mean field approach, the barrier height in GaAs/$Al_xGa_{1-x}As$ superlattice is assumed to be a constant determined by x. On the other hand, recent scanning tunneling microscopic (STM) studies of GaAs/$Al_xGa_{1-x}As$ quantum wells, AlAs/GaAs superlattices, and $Al_xGa_{1-x}As$ 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 the transmission coefficient. We divided the barrier into small cubes of atomic scale representing GaAs or AlAs molecules, and randomly assigned either the potential V_0 for AlAs or 0 for GaAs. V_0 is 1.12 eV (0.26 eV) to simulate the band offsets for electrons (holes) (Fig. 4a). We solved the resulting three-dimensional effective mass equation with appropriate boundary conditions to obtain the transmission coefficients. The resulting transmission coefficients are larger than those obtained from the one-dimensional mean field approach, but still far too small to explain the large transmission coefficient of the order of 10^{-3} 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. 4b, simulating the possible aligning of GaAs or AlAs roughly along the growth direction in light of recent STM studies [15]. We performed the quantum mechanical calculation for various sizes of rectangles to study the cluster size effect on the transmission coefficients. Furthermore, the possible effect of "kinks" was considered (Fig. 4c) in connection with ref. [15] where the GaAs "quantum wires" were shown to "zigzag" their ways through the barrier. In Fig. 4d, the transmission coefficients of holes as a function of x are plotted for several cluster sizes using the model of Fig. 4b. Holes rather than electrons were simulated because it is generally believed that the transport of excitons is determined mostly by the first quantized holes, whose transport is generally slower than 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 the transmission coefficient, so that for the cluster size of 30 Å, nearly all holes can pass through the barrier for relatively low x. The strong x-dependence and weak d-dependence can then be explained in this model, albeit somewhat trivially. Finally, the results using the model schematically described in Fig. 4c show that the effect of the "kink" is to decrease the transmission coefficient only slightly without changing the overall trends.

Our model calculations suggest that the anomalously large transmission coefficient and most features of our experiments can be explained, at least qualitatively, if there were large enough clustering of GaAs or $Al_xGa_{1-x}As$ with very low x, connecting two GaAs quantum wells in a quantum-wire-like fashion. 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. Finally, even without clustering, quantum wells with completely random substitutional alloy as barriers might have extended states below a certain x [16]. This type of approach has not yet shown its full potential in semiconductor physics.

In conclusion, we experimentally demonstrated that a mysteriously large transfer of excitons through thick barriers occurs when the barrier is composed of $Al_xGa_{1-x}As$ alloy. Unlike "solid barriers" such as GaAs/AlAs digital alloys, $Al_xGa_{1-x}As$ 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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Figure Caption

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/AlAs; 5ml/2ml, digital alloy)/GaAs ADQW (75 Å/300 Å/100 Å) (top), and $In_{0.1}Ga_{0.9}As/GaAs/In_{0.13}Ga_{0.87}As$ (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 ($In_{0.1}Ga_{0.9}As$).

Fig. 2: Transfer efficiency 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 $GaAs/Al_xGa_{1-x}As/GaAs$ ADQW (75 Å/300 Å/100 Å). The efficiency was averaged over several different spots.

Fig. 3: Transfer efficiency at 14 K for $GaAs/Al_{0.3x}Ga_{0.7}As/GaAs$ ADQW (75 Å/100, 300 Å, 700, or 1500 Å/100 Å) plotted against d.

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) Transmission coefficients 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.

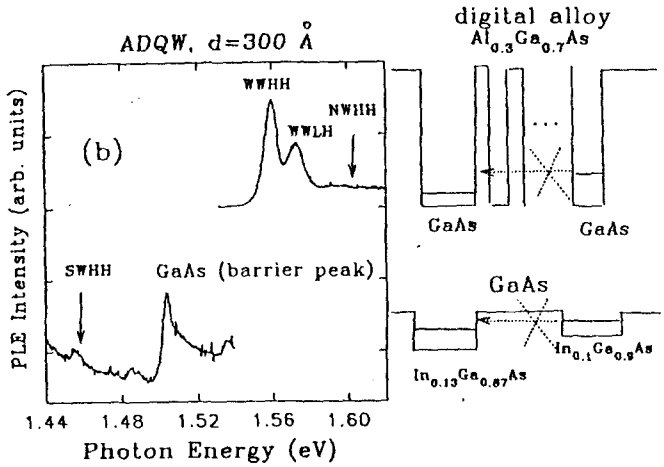

Fig. 1b

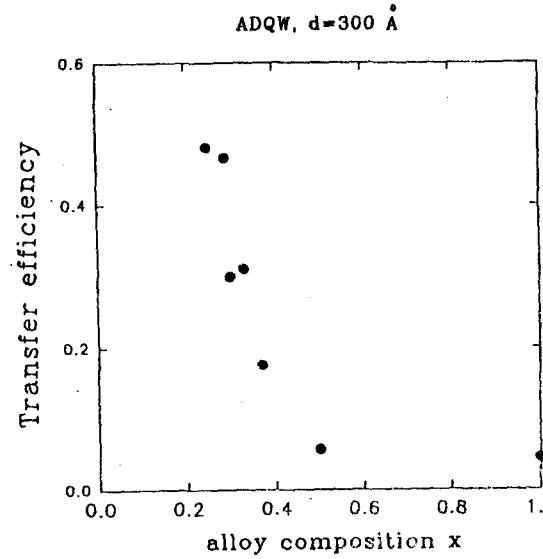

Fig. 2

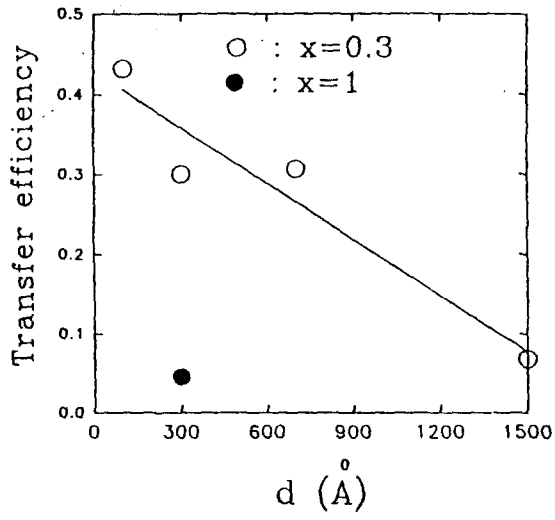

Fig. 3

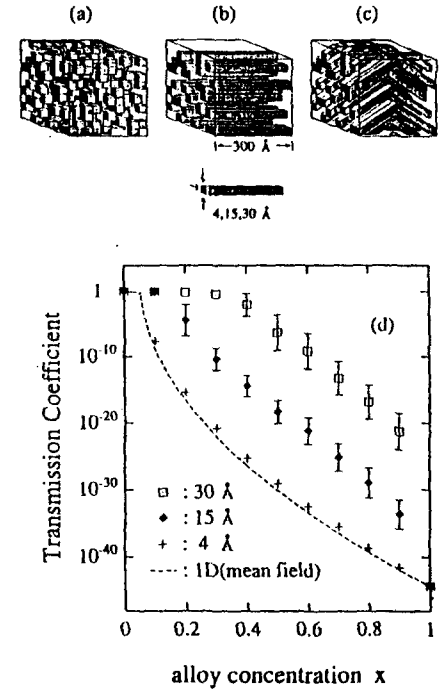

Fig. 4