Thick $Al_xGa_{1-x}As$: An intrinsically percolating barrier owing to its microscopic structural inhomogeneity

D. S. Kim,[a] H. S. Ko, Y. M. Kim, S. J. Rhee, S. C. Hong, Y. H. Yee, D. S. Yee, and J. C. Woo
Department of Physics, Seoul National University, Seoul 151-742, Korea

H. J. Choi and J. Ihm
Department of Physics and Center for Theoretical Physics, Seoul National University, Seoul 151, Korea

D. H. Woo and K. N. Kang
Korea Institute of Science and Technology, Cheongryang, Seoul 136-791, Korea

(Received 13 May 1996; accepted for publication 26 August 1996)

A significant charge transfer, which differs from tunneling, over thick $Al_xGa_{1-x}As$ barrier in GaAs/$Al_xGa_{1-x}As$ asymmetric double quantum wells is studied by cw photoluminescence excitation (PLE) and time-resolved photoluminescence. It is found that 300-Å-thick $Al_{0.3}Ga_{0.7}As$ barrier is universally ''leaky'' with transport time of ~300 ps, while AlAs and AlAs/GaAs digital alloy barriers with same thickness are not. Aided by a model calculation, we suggest that the intrinsic inhomogeneities in the alloy, which recent x-ray and scanning tunneling microscope studies revealed, may be responsible. © *1996 American Institute of Physics.* [S0003-6951(96)02843-4]

GaAs/$Al_xGa_{1-x}As$ quantum wells are perhaps the most thoroughly investigated quantum well system, and yet some puzzles still remain. In this letter, we are interested in one such puzzle: the significant GaAs inter-well charge transfer over very thick $Al_xGa_{1-x}As$ alloy barrier. The existence of this transfer was first recognized by Wilson *et al.* and also by Tomita *et al.*[1] Since the experiments of Ref. 1 were performed at low temperature, thermal excitation is as unlikely an explanation as the normally considered tunneling. In addition, the transfer efficiency was fairly constant over a wide temperature range (2–100 K), a fact that is further confirmed by our photoluminescence excitation (PLE) experiments.

We first demonstrate that this phenomenon is quite general, by showing that the transfer exists in all samples grown by three different molecular beam epitaxy (MBE) machines. Every sample shows significant transfer from the narrow well (NW) to the wide well (WW) through 300-Å-thick $Al_{0.3}Ga_{0.7}As$ barriers [Fig. 1(a)]. Since the ''leak'' over thick AlGaAs is a general phenomenon, to unravel the cause of this leak we devised other barrier structures to stop the leak. We replaced the $Al_{0.3}Ga_{0.7}As$ barrier by an equivalent AlAs/GaAs digital alloy and found that the leak disappears for the digital alloy barrier. We also found that $Al_xGa_{1-x}As$ with $x \geqslant \sim 0.35$ can stop the leak. From these observations, we suggest that the inhomogeneities of the AlGaAs barrier height, which may be unavoidable in the present day growth conditions, is responsible. Our three dimensional quantum mechanical calculation supports this interpretation, assuming the existence of clustering of the size of ~20 Å found in recent scanning tunneling microscope (STM) studies.[2–4]

In Fig. 1(a) PLE spectra of WW PL at 10 K are shown for three GaAs/$Al_{0.3}Ga_{0.7}As$ asymmetric double quantum well (ADQW) samples, each of which was grown by three different MBE machines (labeled I, II, and III). All the samples have the same barrier thickness =300 Å and WW thickness=100 Å, and the nominal NW widths are, from top to bottom, 50, 70, and 75 Å. While the first two peaks correspond to WW heavy hole exciton (WWHH) and WW light hole exciton (WWLH) (not labeled), the strong NW features in the PLE spectra are clearly visible in all three samples. The existence of strong NW peaks in the PLE spectra of WW PL has no other explanation than the existence of transfer from the NW to the WW, an interpretation that is universally accepted and widely used.

We note that the ''quality'' of these different samples varies quite a bit. Nevertheless, all samples show fairly strong NWHH peaks, to a varying degree, demonstrating the intrinsic nature of this effect. Therefore, the fact that $Al_{0.3}Ga_{0.7}As$ is leaky seems to be a fairly general phenomenon, and is not an effect confined to any special MBE machine or the specific choice of sample parameters. From the peak to plateau ratio, or from the ratio of the areas under the WWHH and NWHH peaks in PLE, we can easily see that a significant fraction (from ~30% to nearly all) of excitons excited in the NW eventually end up in the WW. This corresponds to the transfer coefficient per trial t of the order of 10^{-4} to 10^{-3}. While the exact amount of the leak is clearly sample dependent, the fact that there exists a leak that cannot be accounted for by the normally considered tunneling is unmistakable. We also found that the leak persists up to 1500 Å of barrier thickness, and the decrease of the transport efficiency with the barrier thickness is very weak.

In Fig. 1(b), we design various barriers to stop the leak. PLE spectra of ADQWs with digital alloy barrier (top), AlAs barrier (middle), and $Al_{0.5}Ga_{0.5}As$ barrier (bottom) are shown. The barrier thickness was fixed at 300 Å. These samples were grown by MBE II, but samples from other machines show nearly the same results. The digital alloy sample was chosen so that the effective alloy concentration is 0.28. The fact that the leak is largely stopped by all these three barriers is evident from the complete or near absence of the NW features in PLE. We also performed careful x-dependent studies of the leak, and found a dramatic de-

---

[a] Electronic mail: denny@phya.snu.ac.kr

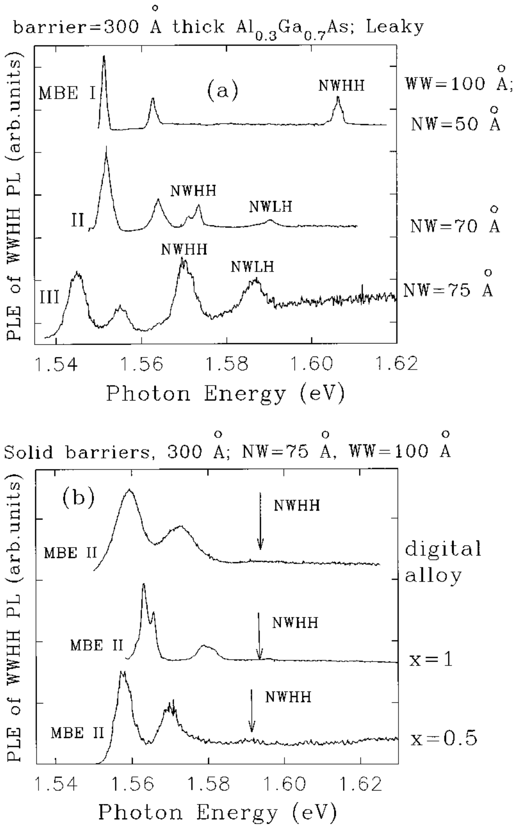

FIG. 1. (a) PLE spectra obtained at 10 K for three GaAs/$Al_{0.3}Ga_{0.7}As$/GaAs ADQW from four different MBE machines. The WW thickness and the barrier thickness are nominally fixed at 100 and 300 Å, respectively, and the nominal NW thicknesses are 50, 75, and 70 Å from top to bottom. (b) PLE spectra at 10 K when the barrier consists of (GaAs/AlAs digital alloy; 5 monolayer/2 monolayer) (top), AlAs (middle), or $Al_{0.5}Ga_{0.5}As$ (bottom). The WW, NW, and the barrier thicknesses are fixed at 100, 75, and 300 Å, respectively.

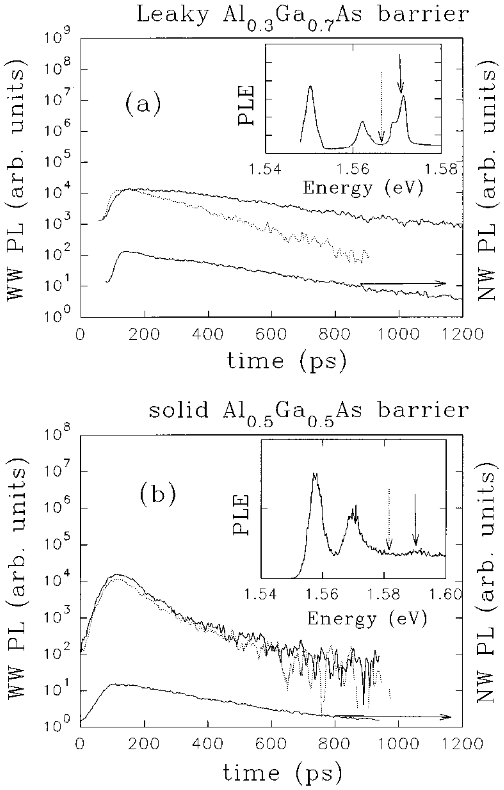

FIG. 2. (a) TR-PL data probed at the WWHH peak for sample II of Fig. 1(a), when exciting slightly below the NWHH (dotted lines) (dotted arrow in the inset), and resonantly at NWHH (solid lines) (solid arrow in the inset). (Inset) PLE spectrum of the same sample on the same spot. (b) The same as (a) for the sample whose PLE is shown at the bottom of Fig. 1(c). (Inset) The same as the inset of Fig. 2(a).

crease with increasing x for $x>0.3$, so that for $x>0.4$, the leak is nearly negligible. This strong x dependence is important, because while our results are surprising, it is consistent with earlier tunneling studies using $x>0.3$.[5]

To further confirm our interpretation based on cw PLE, we performed time-resolved photoluminescence (TR-PL) experiments. TR-PL data obtained using a streak camera at 10 K for one of our samples [sample II of Fig. 1(a)] are shown in Fig. 2(a) along with PLE spectrum for the same sample on the same spot (inset). We first excite WW continuum only (dotted arrow of inset) and probe WW PL (dotted lines). We then excite WW continuum and NWHH simultaneously (solid arrow of inset) (solid lines). At the bottom, near resonant TR-PL for the NW PL is shown. The drastic change in time resolved PL with only a small change of the exciting photon energy again implies the occurrence of a significant transport from the NW to the WW. The fact that WW PL lifetime, which becomes noticeably longer when exciting the WW continuum and NWHH, is nearly the same with that of NW PL leaves no doubt that WW PL at later times is dominated by carriers transported from the NW.

While the details will be given elsewhere, the transport time can be deduced from the combination of the overall efficiency of the transport deduced from cw PLE (about 70%), and the NW PL lifetime: we obtain about 300 ps of transport time from the NW to the WW. Unlike the sample with $Al_{0.3}Ga_{0.5}As$ barrier, the sample with $Al_{0.5}Ga_{0.5}As$ barrier grown by the same MBE shows no change in TR-PL line shapes. Therefore, we conclude that there exists no transport from the NW to the WW for the $Al_{0.5}Ga_{0.5}As$ barrier. We stress that TR-PL experiments on other samples with ‘‘solid barriers’’ shown in Fig. 1(b) also show no transport.

It is clear that an explanation for the universal leak observed in GaAs/$Al_xGa_{1-x}As$ ADQW should be able to explain the following essential features: (1) The large eventual transport efficiency for the $Al_{0.3}Ga_{0.7}As$ barrier. (2) The persistence of the leak over very long distance (up to 1500 Å). (3) The near-disappearance of the leak when the barriers are digital alloy, AlAs, or AlGaAs with $x>0.4$. The original proposed mechanism of dipole–dipole interaction,[1,6] which predicts strong dependence on the barrier thickness [barrier width)$^{-4}$] but no dependence on the alloy composition or the structure of the barrier apparently cannot explain these main features.

We now ask what other mechanisms might be responsible for the observed phenomenon. One hint may lie in recent STM[2–4] and x-ray[7] studies of $Al_xGa_{1-x}As$, which demonstrated clear signature of clustering of up to 20–30 Å of Ga or Al atoms slightly off-axis from the growth direction. These *structural* observations might have far reaching implications on the *dynamics* of the GaAs/AlGaAs quantum wells.

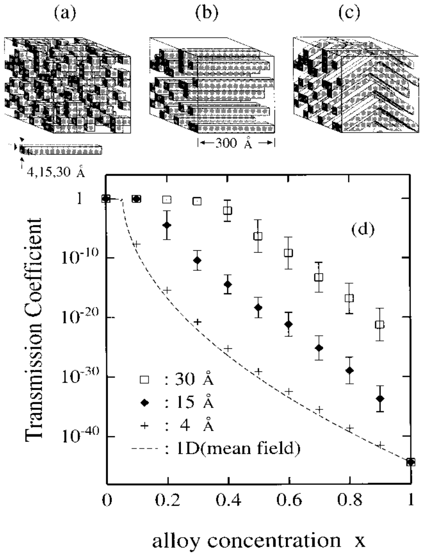

FIG. 3. (a) Schematics for the construction of barriers used in our model calculations assuming completely random, atomic alloy fluctuations. Dark squares represent AlAs ''molecules.'' (b) Schematics for our model calculations taking into account the clustering and the formation of channels, and (c) 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 $\sim 0.5 m_e$, and the wavelength of 150 Å.

To see the effect of clustering on the inter-well dynamics, we first considered the effect of atomic scale fluctuations (4 Å) on the transfer coefficient per single trial (t), as shown in Fig. 3(a). We divided the barrier into small cubes of atomic scale (4 Å) 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). Using a supercomputer, we solved the resulting three-dimensional effective mass equation. The resulting t is close to the prediction of the mean field theory and thus too small to explain the experimental results.

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. 3(b), simulating, with some drastic simplifications, the aligning of GaAs or AlAs clusters. Furthermore, the possible effects of ''kinks'' and the slight deviation of the clustering axis from the growth direction were considered [Fig. 3(c)] in connection with Refs. 2 and 3. In Fig. 3(d) t of holes, which may determine the transfer of excitons due to their larger effective mass, are plotted as a function of x for several cluster sizes using the model of Fig. 3(b). Holes are considered because we assume that holes determine the transfer of excitons due to their slower transfer compared with that of electrons. From the results shown in Fig. 3(d) we can readily see that increasing the cluster size rapidly enhances t, so that for a cluster size between 15 and 30 Å, most of our experimental feature can be explained, both quantitatively ($t \sim 10^{-4} - 10^{-3}$), and qualitatively (the strong x dependence). Finally, the results using the model schematically described in Fig. 3(c) show that the effect of the ''kink'' is to decrease t only slightly, thus suggesting that t is not sensitive to the exact nature of cluster shapes, but to the cluster size. Finally, our calculation is mainly concerned with coherent transport, and we ignore ''incoherent'' effects such as scattering by phonons. However, the temperature independence of the leak partially justifies our considering only the coherent transport.

In conclusion, we experimentally demonstrated that the mysteriously large transfer of excitons through thick barriers is a completely general phenomenon for $x < 0.35$. Unlike ''solid barriers'' such as GaAs/AlAs digital alloys, $Al_xGa_{1-x}As$ is essentially leaky, or percolating, especially for $x < 0.35$. Our results suggest 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.[8]

This work was partially supported by Korean Science and Engineering Foundation (951-0205-023-2), and the Basic Science Research Institute Program of the Ministry of Education (BSRI 96-2421 and 95-E-4009). We wish to thank A. Vinattieri for taking TR-PL data, and T. W. Noh, D. Kim, W. Jhe, R. S. Knox, J. Shah, A. Vinattieri, T. C. Damen, and A. Tomita for helpful discussions.

[1] B. A. Wilson, R. C. Miller, S. K. Sputz, T. D. Harris, R. Sauer, M. G. Lamont, C. W. Tu, and R. F. Kopf, Inst. Phys. Conf. Ser. No. 83, 1986, p. 215; A. Tomita, J. Shah, D. S. Kim, T. C. Damen, J. M. Kuo, S. Schmitt-rink, P. Thomas, and R. S. Knox, Int. Conf. Quantum Electron. Tech. Digest Ser. 9, 1992, p. 116.

[2] A. R. Smith, K. J. Chao, C. K. Shih, Y. C. Shih, and B. G. Streetman, Appl. Phys. Lett. **66**, 478 (1995).

[3] M. B. Johnson, U. Maier, H. P. Meier, and H. W. M. Salemink, Appl. Phys. Lett. **63**, 1273 (1993).

[4] O. Albrektsen, D. J. Arent, H. P. Meier, and H. W. M. Salemink, Appl. Phys. Lett. **57**, 31 (1990).

[5] M. Nido, M. G. W. Alexander, W. W. Rühle, T. Schweizer, and K. Köhler, Appl. Phys. Lett. **56**, 355 (1990).

[6] A. Tomita, J. Shah, and R. S. Knox, Phys. Rev. B **53**, 10793 (1996).

[7] T. S. Kuan, T. F. Keuch, W. I. Wang, and E. L. Wilkie, Phys. Rev. Lett. **54**, 201 (1985).

[8] D. S. Kim, H. S. Ko, Y. S. Lim, Y. M. Kim, J. S. Lee, S. J. Chee, W. S. Kim, S. C. Hong, Y. H. Yee, J. S. Khim, J. M. Jung, S. Huhr, J. H. Lee, J. S. Chang, B. D. Choe, J. C. Woo, P. H. Song, H. J. Choi, S. H. Jhi, J. Ihm, E. J. Shin, D. Kim, D. H. Woo, K. N. Kang, and J. J. Song, J. Opt. Soc. Am. B **13**, 1210 (1996); P. H. Song and D. S. Kim, Phys. Rev. B **54**, 2288 (1996).