Percolation of carriers through low potential channels in thick $Al_xGa_{1-x}As$ ($x<0.35$) barriers

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We study by photoluminescence excitation the heretofore unsolved puzzle of a significant charge transfer over a thick (100 to 1500 Å) $Al_xGa_{1-x}As$ barrier in $GaAs/Al_xGa_{1-x}As$ asymmetric double quantum wells, which the normally considered tunneling cannot account for. This phenomenon is completely general, observed in all the samples grown under standard growth conditions (~600 °C), that originated from many different sources. The existence of such leakage is also confirmed by time-resolved photoluminescence experiments. However, when the alloy barrier is replaced by an equivalent GaAs/AlAs digital alloy, or by AlAs, the leak largely disappears. In addition, a GaAs barrier separating two shallow $In_xGa_{1-x}As$ quantum wells permits only relatively small transfer. The leak has a weak dependence on the barrier thickness at $x=0.3$, but is a very strong function of x around $x=0.3$. We argue that there is no way to explain all of the observed phenomena simultaneously other than by the existence of intrinsic structural inhomogeneities in the alloy. Essentially, there may exist low potential channels in the alloy barrier created by microscopic clustering of like molecules, through which percolationlike transport occurs. This picture is supported by a three-dimensional quantum-mechanical model calculation. Our work pins down the dynamical implications of the partial ordering and clustering in $Al_xGa_{1-x}As$ and related semiconductor ternary alloys. The scope of this paper is exclusively for the transport over thick $Al_xGa_{1-x}As$ barriers ($x<0.35$) and does not include the relatively small but still non-negligible transport over thick homogeneous barriers such as GaAs, AlAs, and AlAs/GaAs digital alloys, and $Al_xGa_{1-x}As$ ($x>0.35$). We contend that transport over these thick, homogeneous barriers may be owing to other mechanisms such as dipole-dipole transfer, photon reabsorption, nonequilibrium distribution of carriers, and polariton transport that are also unrelated to conventional tunneling. [S0163-1829(96)02144-3]

I. INTRODUCTION

Ever since its proposal,[1] and the subsequent realization of the superlattice, $GaAs/Al_xGa_{1-x}As$ has been the most thoroughly investigated quantum-well system, and serves as a model for other quantum wells and superlattices. In particular, the alloy composition $x\sim0.3$ is most widely used. The overwhelming majority of the experimental results on alloy superlattices has been analyzed using the mean-field approach.[2] This approach has been enormously successful in dealing with such dynamic and static problems as tunneling through thin barriers, energy levels, or the density of states. With this phenomenal success, the fact that alloy superlattices are an essentially disordered system due to the alloy fluctuations and structural inhomogeneities is often put aside or temporarily forgotten.

Recently, there has been much interest in ordered III-V ternary alloys.[3–24] While many studies focused on atomic scale ordering and band-gap changes,[3–20] there exists a different kind of partial, longer-range ordering: the microscopic clustering of like ''molecules'' or atoms[21] such as AlAs and GaAs in $Al_xGa_{1-x}As$.[22–25] Therefore randomly substituting different atoms into each site can be a poor approximation of the reality. Needless to say, this microscopic clustering, whose size is of a few lattice constants, should be distinguished from the macroscopic clustering and phase separation in strained alloys.[26–28] In a slightly different approach, such well-known phenomena as the direct-to-indirect band-gap transition in $Al_xGa_{1-x}As$ has been interpreted as a phase transition as a function of an order parameter, without invoking the virtual-crystal approximation and the assumption of well-defined wave vectors.[29] However, thus far, there exist very few studies of the *dynamical* implication of this *structural* information: for instance, what would be the effect of partial ordering and clustering on the effectiveness of $Al_xGa_{1-x}As$ as a barrier?

The question raised above is closely related to an extremely puzzling yet general aspect of a $GaAs/Al_xGa_{1-x}As$ system: the persistence of a significant interwell charge transfer even when the barrier is very thick so that no tunneling is expected. The existence of this puzzle was recognized by many researchers,[30] and the first written evidence was given by Wilson *et al.*[31] These authors correctly ruled out photon recycling as the possible mechanism, as the transfer efficiency was too large to be accounted for by this process. Recently, this puzzle was revisited by Tomita and co-workers,[32,33] and dipole-dipole interaction was proposed as a possible mechanism. In Refs. 30–33, the evidence of

significant interwell transfer was the strong narrow well (NW)-related peaks in photoluminescence excitation (PLE) spectra of the wide well (WW) PL. To appreciate the surprising implication of such transfer, one has only to estimate, from semiclassical mean-field theory,[34,35] what the tunneling time is, say, over the 300-Å-thick $Al_{0.3}Ga_{0.7}As$ barrier used in Refs. 30–33: assuming that a charge starts from a 50-Å well and tunnels to the continuum of a thicker adjacent well, the tunneling time is of the order of 0.1–1 s for electrons, and even larger for holes. Therefore, we expect no tunneling over such a thick barrier. Since the experiments of Refs. 30–33 were performed at low temperature with a prohibitively small Boltzmann factor for overcoming the barrier, thermal excitation is an equally unlikely explanation. In addition, there is no apparent dependence of the amount of leak on temperatures over 2–100 K, further ruling out thermal excitation.

This unexpected phenomenon attracted some theoretical interest, and the proposed mechanisms include dipole-dipole interaction, photon reabsorption, and polariton effects.[31–33,36–38] Naturally, with such an unexpected phenomenon, the most important step is to determine whether or not it is general.

In this paper, we first establish that the leak is quite general, by showing that significant transfer exists in all samples grown by four different molecular-beam-epitaxy (MBE) machines with standard growth conditions (about 600 °C). *Every sample that we investigated shows significant transfer from the NW to the WW through ⩾200-Å $Al_{0.3}Ga_{0.7}As$ barriers [Fig. 1(a)]*. Therefore, the fact that a significant charge transfer occurs over a thick alloy barrier is not due to a specific sample preparation or ''sample imperfection,'' but an intrinsic property of the $Al_{0.3}Ga_{0.7}As$ alloy barrier. Since this ''leak'' over thick $Al_xGa_{1-x}As$ is a universal feature of $GaAs/Al_{0.3}Ga_{0.7}As$ asymmetric double quantum wells (ADQW's) an important question to ask is whether it arises from the structural property of the $Al_xGa_{1-x}As$ alloy that the mean-field theory may overlook, or whether it is related to other proposed mechanisms and perhaps to defects such as the DX centers.

To resolve this question, we have systematically changed the barrier, *in such a way to reduce the leak* significantly: most significantly, we replaced the $Al_{0.3}Ga_{0.7}As$ barrier by an equivalent AlAs/GaAs digital alloy. We found that the leak mostly disappears for the digital alloy barrier. Furthermore, a GaAs barrier separating two shallow $In_xGa_{1-x}As$ wells reduces the leak significantly. These combined results strongly imply that the leak is caused by a fundamental structural property of the $Al_xGa_{1-x}As$ alloy. From these observations, we contend that the inhomogeneities of the $Al_xGa_{1-x}As$ barrier height, which may be unavoidable in the present day standard MBE growth conditions, is responsible. We then perform a three-dimensional quantum-mechanical calculation using a supercomputer, and show that within our simplified model, a drastic increase with the increasing cluster size is indeed expected.

This paper is organized as follows. In Sec. II, we show the universality of the leak by demonstrating that the leak exists in samples from every MBE machine we tried. In Sec. III, we perform barrier-dependent cw-PLE and time-resolved PL studies, and show that the leak is caused by intrinsic structural inhomogeneities of the alloy barrier. In Sec. IV, the barrier thickness and alloy compositional dependencies are shown. In Sec. V, we discuss and rule out many proposed mechanisms, and give a detailed description of how we arrived at the three-dimensional picture of the alloy superlattice, in which the inhomogeneities in the barrier height play an important role. The results of the three-dimensional quantum-mechanical calculation are discussed in Sec. VI. Summary and discussions are contained in Sec. VII.

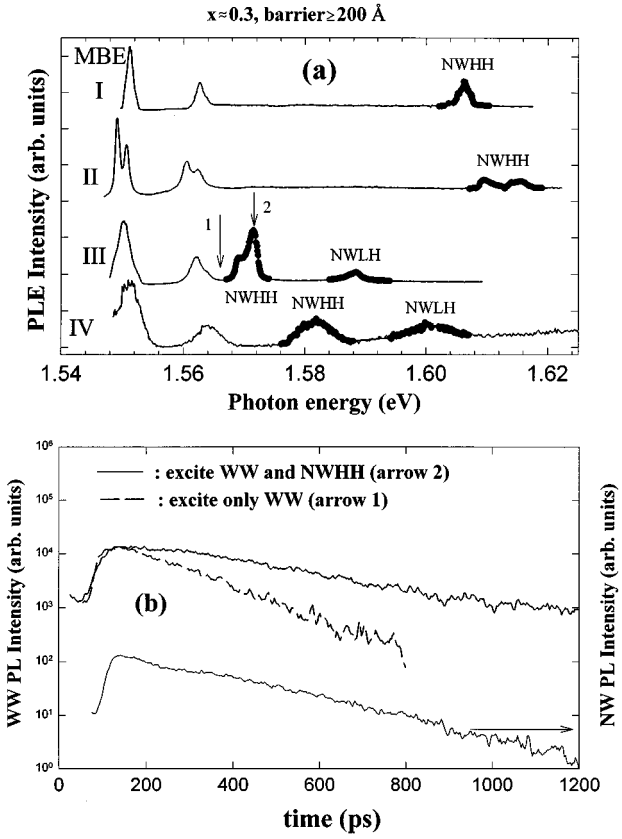

FIG. 1. (a) PLE spectra obtained at 10 K for four $GaAs/Al_{0.3}Ga_{0.7}As/GaAs$ ADQW's from four different MBE machines. The WW thickness is nominally fixed at 100 Å, and the nominal NW and barrier thicknesses are, respectively, (50 Å, 300 Å), (50 Å, 200 Å), (75 Å, 300 Å), and (70 Å, 700 Å) from top to bottom. The approximate Stokes shifts are, from top to bottom, 0.2, 0.1, 0.1, and 0.7 meV. (b) Time-resolved PL data probed at the WW PL peak for sample III, when exciting slightly below the NWHH [arrow 1 of (a)] (dotted lines), and when exciting at NWHH [arrow 2 of (a)] (solid line). At the bottom, time-resolved PL of the near-resonant NW PL is shown.

II. UNIVERSALITY OF THE LEAK AND THE TRANSPORT TIME

In Fig. 1(a), PLE spectra of the WW heavy hole (HH) PL at 10 K are shown for four $GaAs/Al_{0.3}Ga_{0.7}As$ ADQW samples, each of which were grown by four different MBE machines (labeled I, II, III, and IV), some of which are located in different continents. All the samples have thick barriers ⩾200 Å, and the nominal WW width was kept constant at 100 Å. The standard growth conditions optimized for each machine were used. It is important to note that we did not try

to alter the growth conditions to enhance and/or suppress the leak. While it will be interesting to change growth conditions to see their effects on the leak, it is more important to first establish that it is a universal effect for most samples grown under the standard conditions.

The nominal NW width and the barrier width are listed in the figure caption. The first two peaks correspond to WWHH and WWLH (not labeled) (LH stands for light hole), and the NW features in the PLE spectra are represented by thicker lines. 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, as suggested by different PLE linewidth, PL efficiency, the amount of Stokes shift (0.1–0.7 meV), the existence of monolayer fluctuations, etc. Nevertheless, *all samples show fairly strong NWHH peaks, to a varying degree, demonstrating the intrinsic nature of this effect*. Therefore, the fact that the $Al_{0.3}Ga_{0.7}As$ 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. We stress that, although the amount of leak varies quite a bit from sample to sample, it is always greater, by tens of orders, than the prediction of the mean-field theory over such a thick barrier.

Time-resolved PL data at 10 K for one of our samples [sample III of Fig. 1(a)] are shown in Fig. 1(b). The data were taken using a streak camera with time resolution of ~4 ps. We probe WW PL as a function of time, when exciting slightly below the onset of the NW absorption [arrow 1 of Fig. 1(a)], and when exciting the WW continuum and the NWHH simultaneously [arrow 2 of Fig. 1(a)]. At the bottom, NW PL under near-resonant excitation condition is shown. The drastic change in time-resolved PL with only a small change of the exciting photon energy again implies a significant population transport from the NW to the WW. The fact that the PL decay time at later times is more or less the same as that of the NW PL suggests that WW PL at later times is dominated by electrons and holes that originate in the NW.

The transport time can be deduced in the following way. First, we estimate what fraction of the NWHH eventually ends up in the WW from the integrated area below the NWHH peak and the WWHH peak,[39,40] assuming the same oscillator strengths for NWHH and WWHH.[41] We obtain a value of close to 70%. We then note that the near-resonant NW PL decay time is determined by both the PL lifetime inside the NW and the transport time to the WW: NW PL has the *total* decay rate $1/\tau_{t,\ \mathrm{NW}} \approx 1/(220\ \mathrm{ps})$ (bottom curve), which is the sum of the relaxation rate within the NW and the transport rate: $1/\tau_{t,\ \mathrm{NW}} = 1/\tau + 1/\tau_{\mathrm{NW}}$, where τ is the transport time from the NW to the WW, and τ_{NW} is the lifetime within the NW in the absence of transport. Since the eventual transfer efficiency is given by $\tau_{\mathrm{NW}}/(\tau+\tau_{\mathrm{NW}}) \approx 70\%$, we are left with two equations with two unknowns. Therefore, we obtain $\tau \approx 310$ ps and $\tau_{\mathrm{NW}} \approx 720$ ps. This value is longer than tunneling times in samples with thin barriers, but is of course orders magnitudes shorter than the expected tunneling time. For later discussion, we introduce the transfer coefficient per single trial t, which can be readily obtained knowing the round trip time around the QW. In the samples shown in Fig. 1, we obtain $t \sim 10^{-3}$–10^{-4}. This transfer coefficient represents the smaller of the electron or hole transport coefficient, because exciton transfer will be governed by the slower charge transport. It is customary to assume that the hole governs the transport of excitons, because its larger effective mass makes the transport slower. Therefore, we can assume that the transport coefficient deduced from our experiments corresponds to that of the hole. We will use this quantity in Sec. VI to compare with our model calculation.

We stress that a different set of assumptions can be made concerning t without changing the overall interpretation of the data: for instance, we could assume that the transport is governed mostly be electrons. That will make deduced t smaller by a factor of 10 at most, but since we are mainly interested in a discrepancy of tens of orders of magnitude between the mean-field theory and the reality, there will be virtually no change in the final interpretation.

III. HOW TO STOP THE LEAK

In Fig. 2(a), to unravel the origin of the leak, we design various ways to stop it or make it smaller. PLE spectrum of an ADQW with a 300-Å-thick digital alloy barrier (top), and those of an ADQW with a 300-Å-thick AlAs barrier (middle), and 300-Å-thick $Al_{0.5}Ga_{0.5}As$ barrier (bottom) are shown. These samples were grown by MBE III, but samples from other machines show nearly the same results. The parameters of the digital alloy sample was AlAs/GaAs (2/5 ML) so that the effective alloy concentration is 0.28. The idea behind growing the digital alloy barrier was that it has the similar mean alloy concentration as the $x=0.3$ alloy, but any percolationlike transport process would be removed by the successive layers of AlAs. 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. This is also supported by our time-resolved PL experiments, which show little change whether we excite the NWHH or slightly below the NWHH [Fig. 2(b)], in striking contrast with Fig. 1(b). The results shown in Figs. 1 and 2 are enough to rule out photon reabsorption and polariton transport as the dominant contribution. This is because all barriers are transparent to the photon, so that such strong barrier dependence shown in Figs. 1 and 2 is not expected.

In Fig. 3, we have designed a *shallow* $In_xGa_{1-x}As$ quantum-well sample with GaAs as the barrier. The sample parameter is $In_{0.13}Ga_{0.87}As/GaAs/In_{0.1}Ga_{0.9}As$ (100/300/100 Å). The shallower well (SW; $In_{0.1}Ga_{0.9}As$) is equivalent to the NW in $GaAs/Al_xGa_{1-x}As$ system, and the deeper well (DW; $In_{0.13}Ga_{0.87}As$) to the WW. We have made the $In_xGa_{1-x}As$ wells shallow, so as to maximize any transport, if any, over the GaAs barrier. The SW peak in PLE of the DW is overshadowed by the background due to the continuum excitation of the DW, and also by the barrier peak indicating transport from the GaAs barrier into the well. These results demonstrate that there is relatively small well-to-well transfer over the low barrier provided by GaAs. This is of course in agreement with the prediction of the mean-field theory, since, although the barrier is shallow, it is thick enough to be effective. However, we note that the small transport over these homogeneous barriers (GaAs or AlAs/GaAs digital alloy) are still larger than the prediction of the mean-field theory, which essentially says

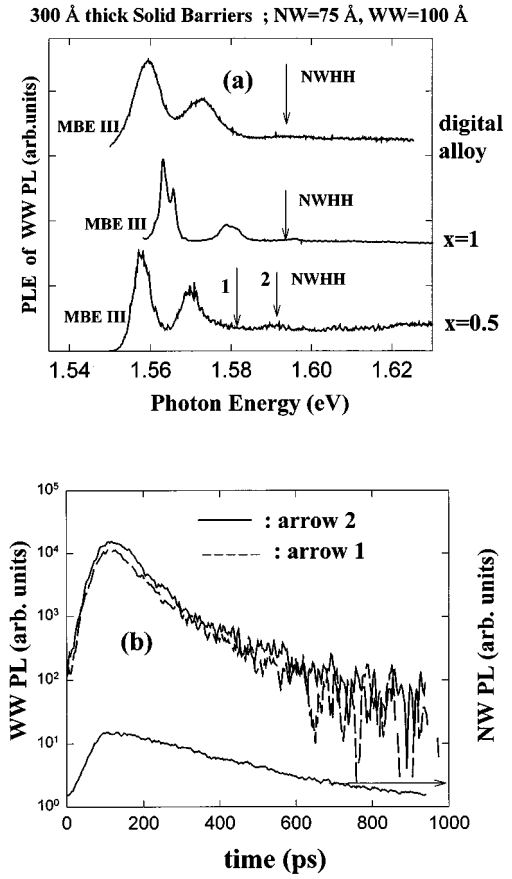

FIG. 2. (a) PLE spectra at 10 K for GaAs ADQW when the barrier consists of (GaAs/AlAs digital alloy; 5 ML/2 ML) (top), AlAs (middle), or $Al_{0.5}Ga_{0.5}As$ (bottom). The well and barrier thicknesses for both samples are WW=100 Å and NW=75 Å, and the barrier is 300 Å. (b) Time-resolved PL data probed at the WW PL peak for the sample with the $Al_{0.5}Ga_{0.5}As$ barrier [bottom of Fig. 3(a)], when exciting slightly below the NWHH [arrow 1 of Fig. 3(a)] (dotted lines), and when exciting at NWHH [arrow 2 of Fig. 3(a)] (solid line). At the bottom, near-resonant time-resolved NW PL is shown.

zero transport. Therefore, in these samples, dipole-dipole interaction or photon-related mechanisms may well be important, although we are mainly interested in alloy barriers throughout this work.

IV. WEAK DEPENDENCE ON THE BARRIER THICKNESS AND STRONG DEPENDENCE ON THE ALLOY COMPOSITION

We now discuss how the leak depends on the barrier thickness and alloy concentration. In Fig. 4, PLE spectra of three samples grown by MBE IV are shown. The samples were grown consecutively in the same run, so as to best isolate the effect of the barrier thickness. While the leak becomes gradually smaller, the persistence of the leak up to the barrier thickness of 1500 Å is suggestive of a very long ''channel'' connecting the two GaAs wells. In Fig. 5, PLE spectra of two samples grown by MBE III in the same run, with identical parameters except for the alloy concentration, are shown. The absence of NW features for $x=0.5$ is in sharp contrast with the strong NW features for $x=0.3$. More careful studies between $x=0.25$ and 1 reveals an almost steplike decrease of the leak with increasing concentration around $x\sim0.3$ (inset, Fig. 5).

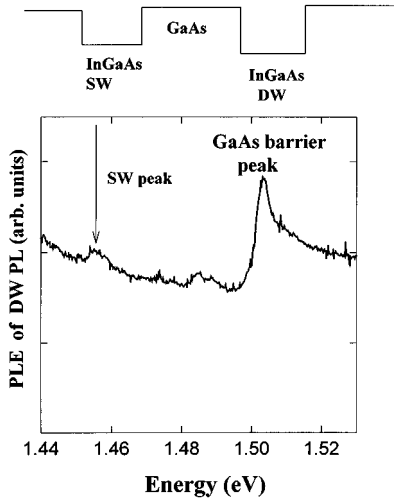

FIG. 3. PLE spectrum of $In_{0.1}Ga_{0.9}As/GaAs/In_{0.13}Ga_{0.87}As$ (100 Å/300 Å/100 Å) ADQW's probed at the deeper well (DW; $In_{0.13}Ga_{0.87}As$). PLE spectra are mostly flat in the shallower well (SW; $In_{0.1}Ga_{0.9}As$) HH regions, indicating little or no transfer. The GaAs peak represents a transfer from the GaAs barrier into the DW.

The strong x dependence shown in Fig. 5 has important implications in establishing a relationship between our work and previous studies. For $x\geqslant\sim0.35$, our PLE shows very slight or no leakage, which is consistent with the prediction of the mean-field theory. Therefore, the agreement found between tunneling experiments performed on samples with barriers with $x\geqslant0.35$ (Refs. 40, 42, and 43) and the mean field theory, is not inconsistent with our observation. Likewise,

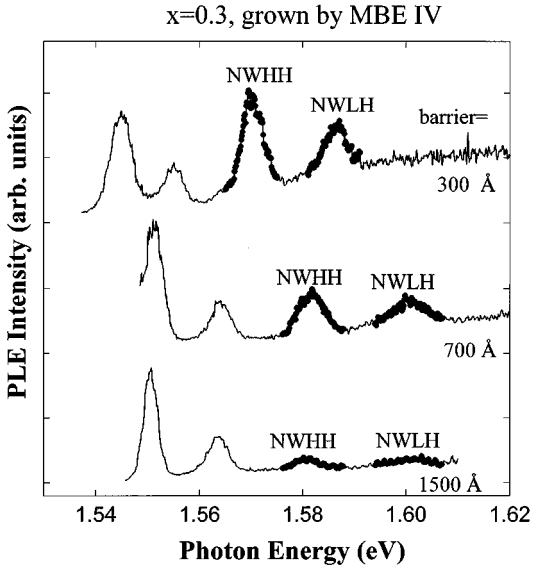

FIG. 4. PLE spectra for three $GaAs/Al_xGa_{1-x}As$ ADQW samples with identical parameters (NW=70 Å, WW=100 Å, and $x=0.3$), except for the barrier thicknesses of 300 Å (top), 700 Å (middle), and 1500 Å (bottom). All three samples are grown by MBE IV, in the same run.

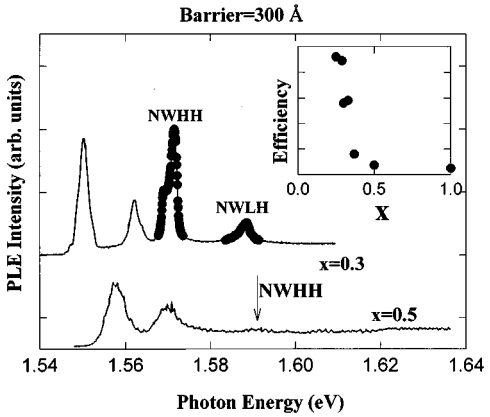

FIG. 5. PLE spectra for two GaAs/$Al_xGa_{1-x}As$ ADQW samples with identical parameters (NW=75 and Å and WW=100 Å, and the barrier is 300 Å), except for $x=0.3$ (top) and 0.5 (bottom). Inset: The x dependence of the transfer efficiency defined as the ratio between the NWHH peak and the WW plateau.

for ''thin'' barriers ($\leqslant$60 Å), we expect that the normal tunneling time would be shorter or comparable to the transport time we observe. In this regard, tunneling studies using thin barrier samples[40,42–45] are not in conflict with our results. However, in samples with $x\leqslant\sim0.3$, the intrinsic leak is significant, and might be the dominant transport mechanism for samples with relatively thick barriers. Thus the importance of the leak is relative, and can probably be ignored for $x\geqslant\sim0.35$, or when the barrier is relatively thin.

V. POSSIBLE MECHANISMS AND FORMING A THREE-DIMENSIONAL PICTURE OF THE ALLOY BARRIER

A mechanism that accounts for all the observed sample dependencies should be able to explain the following essential features of the leak: (1) the large amount of the leak, as specified by the transport time of ~300 ps, and, equivalently, the transport coefficient per single trial of around 10^{-4}–10^{-3}; (2) the near-disappearance of the leak when the barrier is AlAs, AlAs/GaAs digital alloy, or GaAs; and (3) the weak dependence on the barrier thickness and the strong dependence on x. In the earlier experiments, only a single alloy concentration $x=0.3$ was used, with a limited range of barrier thickness. Mainly for this reason, a comprehensive picture was difficult to produce.

One feature in our data that immediately appeals to the senses is the fact that the leak disappears rather strongly with increasing x, around $x=0.3$–0.35 (inset of Fig. 5). This ''critical concentration'' is close to the direct-to-indirect band-gap transition of $Al_xGa_{1-x}As$, and, if we were to use the language of the virtual-crystal approximation, to the Γ-to-X crossover of the conduction band in $Al_xGa_{1-x}As$. Therefore, without close inspection, one may well suspect some direct role of the real-space Γ-to-X charge transport.[46] However, the NWHH exciton energies of our samples lie between 1.57 and 1.62 eV, far below the energy required for the Γ-to-X transition, making it an extremely-unlikely phenomenon in our samples. In addition, since our samples are intrinsic, both holes and electrons should be transported. Therefore, any direct involvement of the Γ-to-X real-space charge transfer can be safely ruled out.

Other mechanisms that can be ruled out as dominant contributions are the photon-related mechanisms such as photon reabsorption and vertical polariton transport,[36–38] which are related to earlier reports of photon recycling.[47,48] It is clear that these could well be the dominant mechanisms of interwell transport over thick homogeneous barriers such as GaAs, AlAs, and a digital alloy. In fact, the small but nonnegligible NWHH peaks (less than 5% in area compared with the WWHH peak) in samples without significant leakage (samples with an AlAs, AlAs/digital alloy, or GaAs with barriers) may well be related to these mechanisms. Nevertheless, since all the barriers used in our investigations are transparent to the photon energies used, we expect virtually no barrier dependencies. Furthermore, the maximum efficiency of these photon-related mechanisms, even assuming 100% luminescence efficiency, is only about 5%, which can be easily estimated from the peak absorption coefficient of the QW exciton peaks. Therefore, while possibly important in case of homogeneous barriers such as GaAs, AlAs, or a digital alloy, the observed strong and well-defined trends with barrier structures and large efficiency help us rule out these photon-related mechanisms as the dominant contributions.

The original proposal in Ref. 32 was the dipole-dipole transfer. Recently, a much more detailed theoretical study of this mechanism was performed.[33] However, this mechanism also did not explain the strong dependence on barrier structures: it could not explain why AlAs, GaAs, or AlAs/GaAs digital alloys can largely stop the leak. Furthermore, dipole-dipole interaction predicts a very rapid falloff of the transport efficiency once the barrier thickness becomes larger than an exciton radius of around 100 Å. Therefore, the weak dependence on the barrier thickness and the persistence of the leak well over a barrier thickness of 300–1500 Å cannot be explained by this mechanism. Therefore, we argue that while this mechanism may be important in the intermediate range of barrier thicknesses where the tunneling is negligible but the barrier thickness is comparable to the exciton radius (i.e., a barrier thickness of around 100–200 Å), it is unlikely to be the dominant mechanism for the universal leak. Nevertheless, as noted in Ref. 33, both the photon-related mechanisms and the dipole-dipole transport are worth further pursuing in samples with *homogeneous barriers* (GaAs, InP, AlAs, or GaAs/AlAs digital alloy) as discussed in this work and in earlier studies,[49] where they may well be the dominant mechanisms of long-range transport.

It is possible that due to nonequilibrium tail of the carrier distribution in the WW, transport over the barrier can occur. Recently, transport over GaAs barriers in $In_xGa_{1-x}As$/GaAs ADQW was explained by this mechanism.[50] This idea is also worth further pursuing, and it is possible that all of the above-discussed mechanisms may contribute to the transport over thick homogeneous barriers.

Finally, the ''critical'' concentration of our experiments happen to coincide roughly with the emergence of the DX center from the resonance state in the $Al_xGa_{1-x}As$ alloy. If the DX centers were in some way related to the observed leak, they would have to *suppress* the leak, for the leak disappears rapidly with increasing x. This is, of course, in contrast with our observation in $In_xGa_{1-x}As$/GaAs ADQW's,

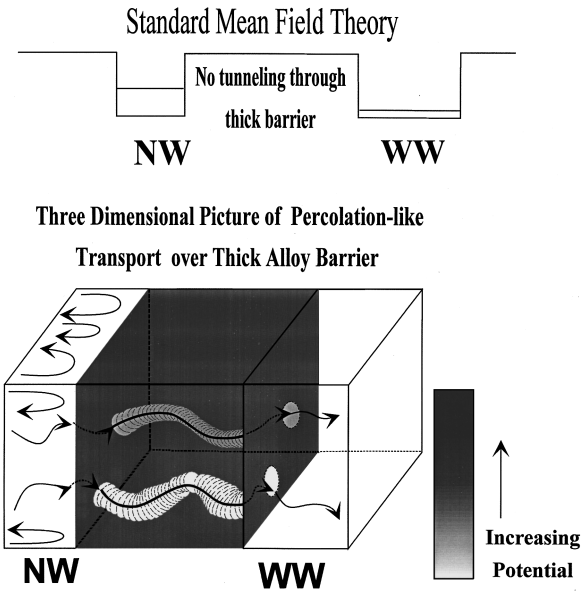

FIG. 6. Schematics for the percolationlike charge transport through low potential channels formed by microscopic clustering. This picture is necessary to explain the universally observed charge transport over thick $Al_xGa_{1-x}As$ alloy barriers that the widely accepted mean-field theory cannot account for.

where the GaAs barrier, without the *DX* center, also stops the leak. Needless to say, *DX* centers are observed in *n*-doped samples, while our work concentrates on intrinsic samples.[51] At present, we are unaware of any ''magic'' defect that can explain all the observed experimental features. As a distantly related question, one may ask whether the increasing number of interfaces in an AlAs/GaAs digital alloy suppresses the leak. However, the suppression of the leak by GaAs barrier in shallow $In_xGa_{1-x}As$/GaAs ADQW's makes this hypothesis inconsistent with our observation.

It is clear that while the mechanisms we discussed thus far have some obvious appeal to common sense, they are not likely to be the dominant mechanism for the leak. On the other hand, the most notable feature in our data is that there is an apparent direct correlation between the degree of homogeneity of the barrier and the amount of the leak. Obviously, AlAs, GaAs, and the digital alloy barrier should have much smaller inhomogeneity than the $Al_xGa_{1-x}As$ alloy, and it is in these barriers that the intrinsic leak observed in the $Al_xGa_{1-x}As$ alloy disappears.

We now ask what other mechanisms might be responsible for the observed phenomenon. One important clue may lie in recent scanning tunneling microscopy (STM) (Refs. 22–25) studies of $Al_xGa_{1-x}As$, which demonstrated a clear signature of clustering of up to 20–30 Å of Ga or Al atoms along well-defined directions. These *structural* observations might have far-reaching implications on the *dynamics* of the GaAs/$Al_xGa_{1-x}As$ quantum wells. Therefore, based upon STM studies of Refs. 22–25 and our observations of the intriguing barrier dependence of the leak we have in mind, we make the following conjecture summarized in Fig. 6: We realize that the tunneling simply cannot account for the observed leak. Therefore, we propose that the existence of low potential channels in the $Al_xGa_{1-x}As$ alloy formed by Ga-rich microscopic clusters, whose existence was revealed by recent STM studies, is responsible for the observed leak. This conjecture is rather attractive because it can explain right away why the leak largely disappears in barriers other than the $Al_xGa_{1-x}As$ alloy. Furthermore, as Sec. VI will show, it can also explain the strong *x* dependence and weak barrier thickness dependence.

VI. MODEL CALCULATION EMPHASIZING THE IMPORTANCE OF CLUSTERING

In Sec. V, we argued that a three-dimensional picture accounting for structural inhomogeneities in the alloy barrier is imperative for understanding the observed leak. In this section, we perform a model calculation to see whether it is possible to explain the leak by clustering and long-range order.

We note that one of the most fundamental assumptions for the mean-field approximation of inhomogeneous media is that the scale of the fluctuation should be much smaller than that of the wavelength of the elementary excitation of one's interest. In $Al_xGa_{1-x}As$, this translates into the assumption of completely random arrangement of Al and Ga atoms, with an atomic scale fluctuation of the barrier height. On the other hand, the wavelength of the electrons and holes we are interested in this work is in the range of ~100 Å. Therefore, if completely random atomic arrangements of Al and Ga were true, the mean-field description of the $Al_xGa_{1-x}As$ alloy would certainly be adequate in all situations, and the leak over the thick barrier may not occur. On the other hand, clustering of Al and Ga atoms, whose size is still smaller than 100 Å,[22–25] may produce, simply by a random arrangement of the clusters, an occasional low potential (thus Ga rich) pathway whose width is in the vicinity of ~100 Å. Then electron and hole waves would be able to ''see'' the pathway. This is essentially the picture proposed in Sec. V. In the following, we present a model calculation using a supercomputer, which tries to isolate the effect of clustering in the transport coefficient per single trial t. We emphasize that because of the dramatic simplification used in this model calculation, our aim is mainly to (and also somewhat quantitatively) understand qualitatively the potential effect of clustering on the interwell transport.

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_0 for AlAs or 0 for GaAs. V_0 is 1.12 eV (0.26 eV) to simulate the standard band offsets for electrons (holes) [Fig. 7(a)]. We solved the resulting three-dimensional effective-mass equation with appropriate boundary conditions to obtain t for an incoming wave with a wavelength of 150 Å, to simulate the transfer from the 75-Å-thick NW. The resulting t's, for both electrons and holes, are larger than those obtained from the one-dimensional mean-field approach, but still far too small to explain $t \sim 10^{-3}$–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. 7(b), simulating the possible

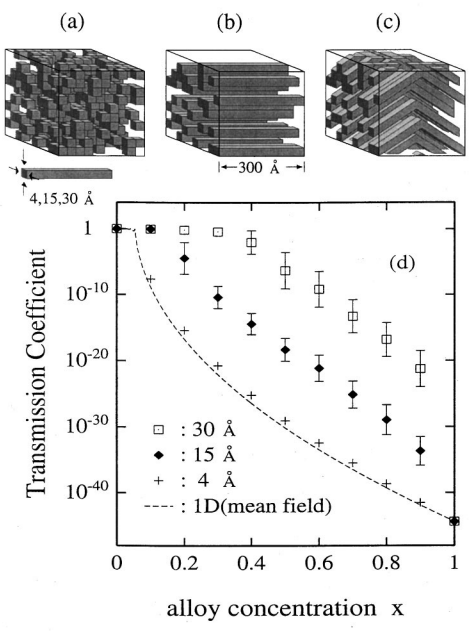

FIG. 7. (a) Schematic for the construction of barriers used in our model-calculations assuming completely random, atomic alloy fluctuations. Dark squares represent AlAs ''molecules.'' (b) Schematic for our model calculations taking into account the clustering and formation of channels. (c) The same as (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 an effective mass of $\sim 0.5 m_e$ and a wavelength of 150 Å.

aligning of GaAs or AlAs roughly along the growth direction.[22–25] We performed a 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. 7(c)] in connection with Refs. 22–25, where the GaAs ''quantum wires'' were shown to ''zigzag'' their ways through the barrier. In Fig. 7(d), values of t of holes as a function of x are plotted for several cluster sizes using the model of Fig. 7(b). Holes are considered because we assume that holes determine the transfer of excitons due to their slower transfer compared with that of electrons. We stress that the essential physics does not change if we were to assume that electrons govern the transfer of excitons instead.

For a grid size of 4 Å, the results are only slightly larger than the prediction of the mean-field theory, despite the fact that there exist 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 a cluster size of 30 Å, nearly all holes can pass through the barrier for relatively low x during the excitonlife time, while, for $x>0.5$, the transmission coefficient is too small to allow any significant transfer. The physical origin of this strong x dependence is simply that, for large enough x, a clustering of 20–30 Å is not enough to create a sufficient number of low potential pathways whose widths are of the order of the wavelength of the excitation, while the opposite is true for smaller x. Finally, results using the model schematically described in Fig. 7(c) show that the effect of the ''kink'' is to decrease t only slightly without changing the overall trends. This helps us to argue that although our model is very simple, the transfer efficiency may be more sensitive to the cluster size, and not to the detailed arrangement and shape of the clusters.

Our model calculations suggest that most features of our experiments can be explained, at least qualitatively, if there were large enough clustering (20–30 Å in size) of GaAs in a quantum-wire-like fashion, to allow significant coupling between the two GaAs quantum wells. This is in fact consistent with STM studies mentioned earlier. Although a more realistic approach to the detailed mechanism of clustering and the resulting structure and pattern formation would be greatly desired, it would be very time consuming; our results strongly imply that the clustering of GaAs in the alloy barrier is a likely source of the enormously enhanced interwell coupling.

Our model can account for the strong x dependence essentially because increasing x rapidly decreases the concentration of wide enough low potential channels. Since the transport through these wide low potential channels is essentially ballistic, the weak barrier thickness dependence can be also explained. Furthermore, necessary clustering in the range of 20–30 Å is in good agreement with the results of the STM studies. Therefore, it is rather likely that the intrinsic structural inhomogeneities in the $Al_xGa_{1-x}As$ alloy barrier is responsible for the universal leak.

VII. SUMMARY AND DISCUSSIONS

In summary, we showed that the ''leak'' over thick $Al_xGa_{1-x}As$ barrier that tunneling cannot explain is a rather universal phenomenon in GaAs/$Al_xGa_{1-x}As$ quantum wells, by performing both cw PLE and time-resolved PL experiments on many samples from many different sources. By designing many different barrier structures, we showed that the degree of inhomogeneity in the barrier is directly correlated to the amount of the leak: while homogeneous barriers such as GaAs, AlAs, and AlAs/GaAs digital alloy are able to stop the leak, $Al_xGa_{1-x}As$ is universally leaky for $x \leqslant 0.3$. This observation strongly suggests that the intrinsic inhomogeneities in the alloy barrier, which today's standard growth conditions may not be able to avoid, are responsible for the leak. Therefore, to understand dynamical transport over thick alloy barriers, it seems imperative to adopt a three-dimensional picture of the alloy barrier. We have performed a three-dimensional quantum-mechanical calculation of transport over a model inhomogeneous barrier, and found that most of the observed phenomena can be explained if there were a microscopic clustering of around 20–30 Å inside the alloy barrier.

Our results can be summarized as follows. For $x<0.35$, and barrier thickness $<$50–100 Å (50 Å for hole transport, and 100 Å for electron transport), tunneling dominates interwell transport. On the other hand, for $x<0.35$, and barrier thickness $>$50–100 Å, the leak would dominate the interwell transport. For the rest of the ''phase space,'' dipole-dipole interaction, polariton transfer, photon reabsorption, and nonthermal distribution of carriers all may contribute to the interwell transport. Therefore, GaAs quantum wells separated by thick alloy barriers with $x<0.35$ should not be viewed as an isolated system, but may have to be viewed as a coupled

system with low potential ''quantum wires'' connecting them in three dimensions. We note that this ''phase diagram'' is very similar to that obtained for the optical-phonon transport studied by time-resolved Raman scattering.[52–54] It is possible that both phonon and carrier transport can be facilitated by the existence of intrinsic clustering and inhomogeneities in the barrier. For the regions of $x>0.35$ and the barrier thickness of $>50-100$ Å, other mechanisms such as the dipole-dipole interaction, photon reabsorption, and polariton transfer may dominate transport.

We further ask the following question that inevitably arises from our investigation: is it a pure coincidence that the direct-to-indirect band gap transition, the critical alloy concentration for the leak, the confined-to-propagating transition of optical phonons all occur around $x=0.3-0.4$? We have already convincingly ruled out the Γ-to-X crossover and the DX center as direct causes for the leak. On the other hand, they may all be a result of a hidden transition that is more fundamental. While more or less a pure speculation, we note that the occurrence of percolation threshold for the diamond lattice, which the zinc-blende structure is based upon, is also close to $x=0.3-0.4$.[55] Deeper insight into the relationship between these seemingly unrelated transitions may be achieved by further theoretical and experimental studies that go beyond the mean-field approach.

Finally, we ask whether there may exist any drastic deviation of the interwell transport from the prediction of the mean-field theory, if the alloy atomic distribution is indeed *completely random*. This may seem a trivial question, and our three-dimensional quantum-mechanical calculation predicts little deviation from the prediction of the mean-field theory if the distribution were completely random [Fig. 7(a)]. However, we note that semiconductor alloys and alloy superlattices have seldom been viewed in light of the Anderson localization.[56] Only recently was a fully three-dimensional Anderson localization theory of alloys developed to study optical-phonon localization in $Al_xGa_{1-x}As$.[57] Without both experimental and theoretical detailed studies, it cannot be ruled out that such a localization approach may be crucial, and might yield surprising results in these *intrinsically disordered systems*, especially in terms of dynamics.

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[1] L. Esaki and R. Tsu, IBM J. Res. Dev. **14**, 61 (1970).
[2] See, for instance, G. Bastard, Phys. Rev. B **24**, 5693 (1981); T. Tada *et al.*, J. Appl. Phys. **63**, 5491 (1988).
[3] K. A. Mäder and A. Zunger, Phys. Rev. B **51**, 10 462 (1995); D. B. Laks, S. H. Wei, and A. Zunger, Phys. Rev. Lett. **69**, 3766 (1992).
[4] C. Wolverton and A. Zunger, Phys. Rev. Lett. **75**, 3162 (1995).
[5] B. L. Gu, Z. F. Huang, J. Ni, J. Z. Yu, K. Ohno, and Y. Kawazoe, Phys. Rev. B **51**, 7104 (1995); J. Ni, X. C. Lai, and B. L. Gu, J. Appl. Phys. **73**, 4260 (1993); A. Chin, B. C. Lin, G. L. Gu, and K. Y. Hsieh, Appl. Phys. Lett. **67**, 3617 (1995).
[6] M. Ishimaru, S. Matsumura, N. Kuwano, and K. Oki, Phys. Rev. B **51**, 9707 (1995).
[7] M. Ishimaru, S. Matsumura, N. Kuwano, and K. Oki, Phys. Rev. B **52**, 5154 (1995).
[8] H. Nakamura, T. Takeguchi, and T. Nishino, Surf. Sci. **363**, 409 (1996).
[9] R. Osorio, J. Bernard, S. Froyen, and A. Zunger, Phys. Rev. B **45**, 11 173 (1992); R. J. Thomas, H. R. Chandrasekhar, M. Chandrasekhar, E. D. Jones, and R. P. Schneider, J. Phys. Chem. Sol. **56**, 357 (1995).
[10] T. S. Kuan, W. I. Wang, and E. L. Wilkie, Appl. Phys. Lett. **51**, 505 (1987); Q. Liu, W. Prost, and F. J. Tagude, *ibid.* **67**, 2807 (1995).
[11] T. S. Kuan, T. F. Keuch, W. I. Wang, and E. L. Wilkie, Phys. Rev. Lett. **54**, 201 (1985).
[12] H. Nakayama, and H. Fujita, Inst. Phys. Conf. Ser. **79**, 289 (1986); F. Glas, *ibid.* **134**, 269 (1993); A. G. Norman, T. Y. Seong, B. A. Phillips, G. R. Booker, S. Mahajan, *ibid.* **134**, 279 (1993).
[13] H. R. Jen, K. Y. Ma, and G. B. Stringfellow, Appl. Phys. Lett. **54**, 1154 (1989); A. M. Mintairov, D. M. Mazurenko, M. A. Sinitsin, and B. S. Yavich, Semiconductors **28**, 866 (1994).
[14] A. Gomyo, K. Makita, I. Hino, and T. Suzuki, Phys. Rev. Lett. **72**, 673 (1994).
[15] M. A. Shahid and S. Mahajan, Phys. Rev. B **38**, 1344 (1988).
[16] O. Ueda, T. Fuji, Y. Nakada, H. Yamada, and J. Umebu, J. Cryst. Growth **95**, 38 (1989).
[17] N. Kuwano and K. Oki, J. Cryst. Growth **163**, 122 (1996).
[18] K. Sinha, A. Mascarenhas, G. Horner, and K. A. Bertness, Phys. Rev. B **50**, 7509 (1994).
[19] M. Konodow, H. Kakibayashi, and S. Minagawa, Inst. Phys. Conf. Ser. **106**, 93 (1990).
[20] K. Sinha, A. Mascarenhas, R. G. Alonso, G. S. Horner, K. A. Bertness, S. R. Kurts, and J. M. Olson, Solid State Commun. **89**, 843 (1994); J. S. Luo, J. M. Olson, and M. C. Woo, J. Vac. Sci. Technol. **13**, 1755 (1995).
[21] B. Kakraborty and Z. Xi, Phys. Rev. B **53**, 5063 (1996); W. Grieshaber, A. Haury, J. Cibert, M. Merle d'Aubigne, A. Wasiela, and J. A. Gaj, Phys. Rev. B **53**, 4891 (1996); Y. Shapira, S. Foner, D. H. Ridgley, K. Dwight, and A. Wold, *ibid.* **30**, 4021 (1984); J. M. Fatah, T. Piorek, P. Harrison, T. Stirner, and W. E. Hagston, *ibid.* **49**, 10 341 (1994).
[22] A. R. Smith, K. J. Chao, C. K. Shih, Y. C. Shih, and B. G. Streetman, Appl. Phys. Lett. **66**, 478 (1995).
[23] A. R. Smith, K. J. Chao, C. K. Shih, Y. C. Shih, K. A. Anselm, and B. G. Streetman, J. Vac. Sci. Technol. B **13**, 1824 (1995).
[24] M. B. Johnson, U. Maier, H. P. Meier, and H. W. M. Salemink, Appl. Phys. Lett. **63**, 1273 (1993).

[25] O. Albrektsen, D. J. Arent, H. P. Meier, and H. W. M. Salemink, Appl. Phys. Lett. **57**, 31 (1990).
[26] S. F. Yoon, Y. M. Miao, K. Radhakrishnan and S. Swamiachan, Superlatt. Microstruct. **17**, 213 (1995).
[27] A. Dimoulas, Z. Hatzopoulos, I. Stoemenos, and A. Christou, Superlatt. Microstruct. **8**, 117 (1990).
[28] F. Peiro, A. Cornet, J. R. Morante, S. A. Clark, and R. H. Williams, J. Appl. Phys. **71**, 2470 (1992).
[29] B. Koiller and R. B. Capaz, Phys. Rev. Lett. **74**, 769 (1995).
[30] For instance, T. C. Damen, J. Shah, and J. J. Song independently found evidence of such transfer or ''leak'' using PLE; T. C. Damen (private communication); J. Shah (private communication); J. J. Song (private communication).
[31] 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. **83**, 215 (1986).
[32] 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**, 116 (1992).
[33] A. Tomita, J. Shah, and R. S. Knox, Phys. Rev. B **53**, 10 793 (1996).
[34] T. Tada, A. Yamaguchi, N. Ninomiya, H. Uchiki, T. Kobayashi, and T. Yao, J. Appl. Phys. **63**, 5491 (1988).
[35] G. Bastard, Phys. Rev. B **24**, 5693 (1981).
[36] M. Batsch, T. Meier, P. Thomas, M. Lindberg, S. W. Koch, and J. Shah, Phys. Rev. B **48**, 11 817 (1993).
[37] N. B. An and G. Mahler, Phys. Rev. B **50**, 17 256 (1994).
[38] D. S. Citrin, Phys. Rev. B **51**, 14 361 (1995).
[39] S. Haacke, N. T. Pelekanos, H. Mariette, M. Zigone, A. P. Heberle, and W. W. Rühle, Phys. Rev. B **47**, 16 643 (1993).
[40] M. Nido, M. G. W. Alexander, W. W. Rühle, T. Schweizer, and K. Köhler, Appl. Phys. Lett. **56**, 355 (1990).
[41] This is a reasonably good assumption, since the increasing oscillator strength of excitons (relative to the continuum) with decreasing well width is partly compensated by the thickness effect between 50 and 100 Å of well thickness; see Y. Masumoto, M. Matsuura, S. Tarucha, and H. Okamoto, Phys. Rev. B **32**, 4275 (1985).
[42] S. Tarucha, K. Ploog, and K. von Klitzing, Phys. Rev. B **36**, 4558 (1987).
[43] A. Nakamura, N. Kawamoto, K. Fujiwara, N. Tsukada, and T. Nakayama, Solid State Commun. **71**, 553 (1989).
[44] D. Y. Oberli, J. Shah, T. C. Damen, C. W. Tu, T. Y. Chang, D. A. B. Miller, J. E. Henry, R. F. Kopf, N. Sauer, and A. E. DiGiovanni, Phys. Rev. B **40**, 3028 (1989).
[45] K. Leo, J. Shah, J. P. Gordon, T. C. Damen, D. A. B. Miller, C. W. Tu, and J. E. Cunningham, Phys. Rev. B **42**, 7065 (1990).
[46] J. Feldmann, C. T. Foxon, E. O. Göbel, P. Dawson, R. Muralidharan, J. Hebling, J. Kuhl, R. Sattmann, and K. Ploog, Phys. Rev. Lett. **62**, 1839 (1989).
[47] J. L. Bradshaw, R. P. Devaty, W. J. Choyke, and R. L. Messham, Appl. Phys. Lett. **55**, 165 (1989).
[48] E. M. Goldys, V. W. L. Chin, T. L. Tansley, and M. R. Vaughan, J. Appl. Phys. **75**, 4194 (1994).
[49] R. Sauer, T. D. Harris, and W. T. Tsang, Appl. Phys. Lett. **50**, 1077 (1987).
[50] A. Vinattieri (unpublished).
[51] D. V. Lang, R. A. Rogan, and M. Jaros, Phys. Rev. B **19**, 1015 (1979).
[52] J. M. Jacob, D. S. Kim, A. Bouchalkha, J. J. Song, J. F. Klem, H. Hou, C. W. Tu, and H. Morkoç, Solid State Commun. **91**, 721 (1994); D. S. Kim, A. Bouchalkha, J. M. Jacob, J. F. Zhou, J. J. Song, and J. F. Klem, Phys. Rev. Lett. **68**, 1002 (1992).
[53] D. S. Kim, A. Bouchalkha, J. M. Jacob, J. J. Song, J. F. Klem, H. Hou, C. W. Tu, and H. Morkç, Phys. Rev. B **51**, 5449 (1995).
[54] D. S. Kim, H. S. Ko, Y. S. Lim, Y. M. Kim, J. S. Lee, S. J. Rhee, 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); D. S. Kim, H. S. Ko, Y. M. Kim, S. J. Rhee, W. S. Kim, J. C. Woo, H. J. Choi, J. Ihm, D. H. Woo, and K. N. Kang, Inst. Phys. Conf. Ser. **145**, 855 (1995).
[55] See, for example, *Introduction to Percolation Theory*, edited by D. Stauffer (Taylor and Francis, London, 1985).
[56] P. W. Anderson, Phys. Rev. **109**, 1492 (1958).
[57] P. H. Song and D. S. Kim, Phys. Rev. B **54**, 2288 (1996); P. H. Song, D. Kim, and D. S. Kim (unpublished).