Cathodoluminescence imaging of quantum wells: The influence of exciton transfer on the apparent island size

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We demonstrate the influence of lateral exciton transfer on the resolution in cathodoluminescence images and thereby the *apparent* island size in monolayer-flat (ML-flat) areas in quantum wells (QW's). The QW used in this study exhibits two peaks originating in regions of two different QW thicknesses, differing by 1 ML. A variation of the exciton transfer is achieved by altering the temperature. When the temperature is increased from 20 to 120 K, the ratio between the low-energy and the high-energy peak is increased by over a factor of 5, caused by an increased lateral transfer. At 20 K the images of the two peaks show a high degree of complementary behavior and features of about 0.5 μm can be resolved. The contrast and the complementary behavior is reduced with increased temperature, i.e., increased transfer, and at 110 K the smallest features observed are about 1 μm. We also suggest a model, where the ML-flat areas in reality are clusters of microislands. These clusters *appear* to be extended islands of a single thickness, due to a one-way transfer of excitons from thinner to thicker microislands. The sizes of these *apparently* extended islands are discussed in terms of a diffusion-length-dependent critical density of microislands.

Monolayer-flat (ML-flat) islands, or terraces, in quantum wells (QW's) is a concept that has been used over the last decade to describe the quality of the interfaces between the barrier and the QW. Several methods have been used to probe the size of these islands.[1–7] One very common, although blunt method is to use the emission peaks from the QW, where an important factor is the diameter of the exciton ($\approx$30 nm), which determines the area of the interfaces it samples when it recombines. Flat areas on a larger scale (>0.1 μm) can be observed by cathodoluminescence (CL) imaging (CLI).[2,7] Images corresponding to the different ML peaks can be recorded. There is a problem inherent to this technique: *The intensity of the emitted luminescence is detected as a function of the spatial position where it was generated and not where it recombined.* CL images can therefore be seriously distorted by transfer (diffusion) of carriers or excitons. There are ways to reduce the effect of lateral transfer,[5,8] but in general CLI has a submicron resolution; a resolution better than 100 nm has been reported for work at low accelerating voltage, 1–3 keV.[9,10]

Several studies have shown that there is a significant transfer of excitons, or electron-hole pairs, between regions of different ML thicknesses within a single QW. This transfer is already effective at low temperatures, e.g., 2–5 K, and can be observed by several methods,[6,11,12] and the intensity ratio between the emission peaks, originating in areas of different thickness, can differ from the actual area coverage by up to one order of magnitude, in favor of the low-energy peak. This means that the apparent island sizes in the images of the low-energy peak tends to be severely overestimated. In studies of diffusion lengths (L_{diff}) in strained and unstrained QW's, diffusion lengths of 1–10 μm have been reported.[5,13,14] The diffusion lengths were assessed by CL-based methods at 300 and at 5 K. A different approach was taken in Ref. 15, where pulsed lasers were used to simultaneously determine the diffusion coefficient (D) and the radiative lifetime (τ) in QW's. In this study, both D and τ increased with temperature, implying that the diffusion length ($L_{\text{diff}}=[D\tau]^{1/2}$) increased with temperature.

The growth conditions for the $Ga_{1-x}In_xAs/InP$ QW used in this study are described in Ref. 6. The emission exhibits two separated peaks, attributed to recombination of excitons in areas with thickness of 4 and 5 ML, respectively. Figure 1 shows a series of CL spectra from the whole area of Fig. 2, where the temperature has been increased from 20 to 120 K. At 20 K, the separation of the peaks is about 45 meV, and the peak width is about 12–16 meV, which is gradually increased up to about 20–25 meV at 120 K. At 20 K the intensity ratio between the 4- and 5-ML peaks is 2:1. With increasing temperature this is gradually reduced 1:3 at 120 K, caused by increased exciton transfer. The part of the sample chosen for the imaging is where the two emission peaks are about equal in intensity at 20 K. In this case the area distribution of the apparent ML-flat islands should be about equal for the two thicknesses, even though the area of the thicker, low-energy, islands will be over-represented in the CL images due to exciton transfer, as discussed above.

The CLI was performed at 5 keV in order to have as small generation volume as possible, with sufficient penetration depth of the electrons to excite the QW directly. The temperature was varied between 20 and 200 K. The probe current was kept at 100 pA. This is well below the 500 pA needed to induce any visible saturation effect in the spectra and images. This effect appears as a change in the ratio of the two peaks involved, where the high-energy peak grows compared to the low-energy peak. The width of the detection window was 8 meV, and it was positioned at the center of the respective peaks. For clarity the images presented here are expanded in contrast to show only the intensity variations. This

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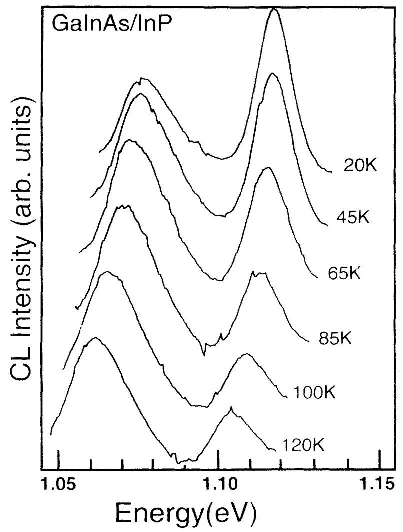

FIG. 1. A series of CL spectra from the same area, obtained at different temperatures, 20–120 K. The two peaks are attributed to a recombination of excitons in areas of 4- and 5-ML thickness of a single QW. At 20 K the ratio between the 4- and 5-ML peaks is 2:1. With increasing temperature this is gradually reduced, and at 120 K it is 1:3.

is done by subtracting a constant intensity level from the images. At 20 K this level is about 10–20 %, whereas at 110 K this is about 50% of the maximum intensity. The line scans, however, show the true variations.

Figure 2 shows the images, and Fig. 3 shows the corresponding line scans obtained at 20 K. As expected from the discussion above, the lateral extension of the *apparent* island of both thicknesses are about equal in size and have about the same area coverage. The borders between the different islands are not well defined, as can be seen even more clearly in the line scans. It can also be noted that the image of the high-energy peak appears to have more well-defined edges of the islands. It is also worth pointing out the high degree of complementary behavior between the two images.

As the temperature is increased, the images of both peaks are less well defined. Figure 4 shows the same area as in Fig. 2, but at 65 K. Already at this temperature a smearing of the features in the images can be observed, especially in the image of the low-energy peak, Fig. 4(b). The features in Fig. 2 can, however, still be recognized. At an even higher temperature, the smearing of the features is increased and, in Fig. 5, only the most prominent features of Fig. 2 can be observed. With increasing temperature it is not only the contrast that is reduced, the finer details of the intensity variations are also lost. In the 20-K images of Fig. 2, features of about 0.5 μm can be resolved. At 110 K, Fig. 5, the smallest features that can be observed are about a factor of 2 larger. In Fig. 5, the complementary behavior is not as clear as in Fig. 2. At even higher temperature, $\approx$200 K, most of the intensity variations are lost and thereby most of the complementary behavior in the images is lost.

This can all be explained in the terms of transfer, or diffusion, of excitons. In the ideal case with no transfer, the resolution in the images is only limited by the size and shape of the excitation volume. In the real case there is, as discussed above, a considerable transfer between regions of different thicknesses within a single QW. The nearer to a boundary between areas of different thicknesses, the more significant the transfer will be. This will lead to a reduction of the contrast in the CL images. Several studies of the temperature dependence of the exciton transfer within QW's have identified three different temperature ranges.[6,16–18] At low temperature, the "trapping" range,[6] the excitons are localized by small scale fluctuations in the potential of the QW. These fluc-

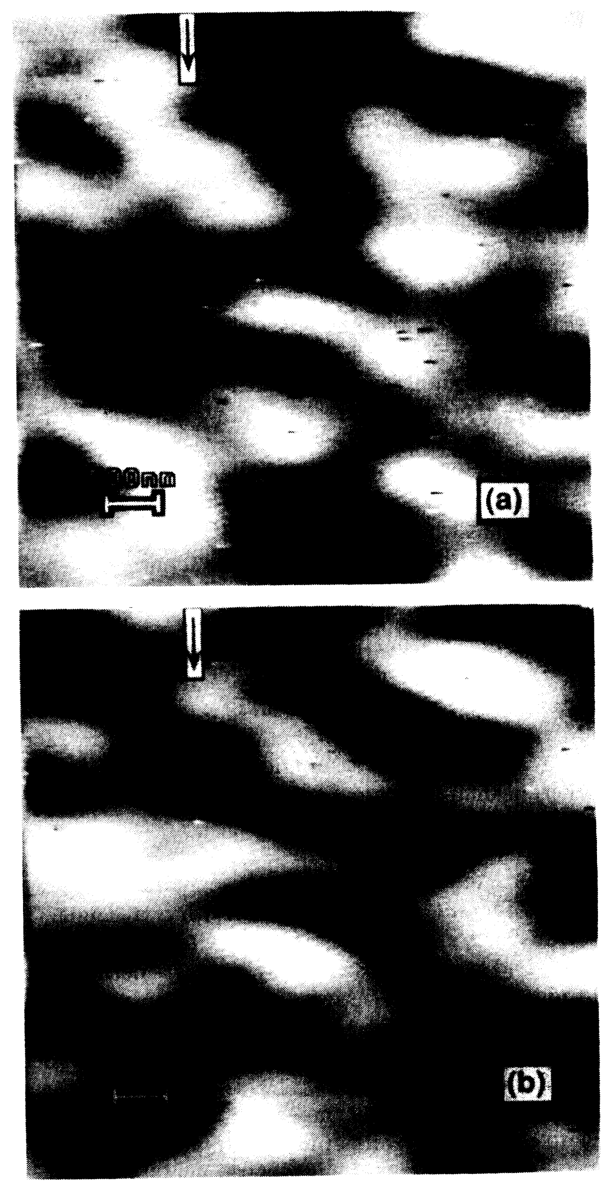

FIG. 2. Images recorded at 20 K show the expanded contrast variations of the 5-ML peak (a) and 4-ML peak (b). Features about 0.5 μm can be observed in both images, and there is a high degree of complementary behavior between the two images. The starting point and the direction of the line scans of Figs. 3 and 6 are indicated by arrows in the images.

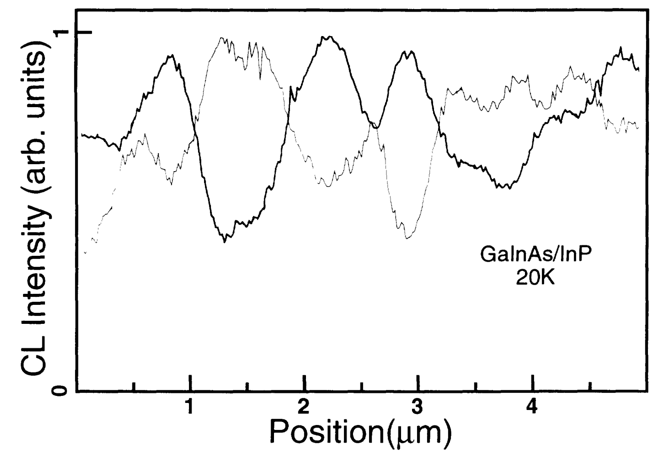

FIG. 3. Line scans of identical lines using the luminescence from the 4-ML (thick line) and 5-ML areas (thin line) of the QW recorded at 20 K, and in the area of Fig. 2. In the line scans, the complementary behavior of the two images is even clearer.

tuations can be caused by fluctuations in the alloy composition or by small scale thickness variations (microroughness).[6,16] Already in this temperature range there is a significant transfer from thinner to thicker areas of the QW as discussed above and in Ref. 6. At intermediate temperatures, the "down-hill" range,[6] the excitons are thermally delocalized. Furthermore, in this temperature range the transfer takes place only from the thinner areas to the thicker areas; additional energy is needed for the transfer to go in the opposite direction, from thicker to thinner areas. This in turn leads to an asymmetry in the contrast of the images. The images of the thinner area will in principle only be affected by outdiffusion from the area. If this is effective from the whole area, but most significant from the boundaries, the result is an overall reduction of intensity. In the image of the thicker area this leads to an overall increase in intensity. This results in a reduction of the contrast in the image of the low-energy peak, whereas the image of the high-energy peak is more or less unaffected. This can be observed in the images of Fig. 2 and even better in the line scans in Fig. 3.

As the temperature is increased this one-way transfer is enhanced. This leads to a general reduction of the contrast in both images, but still with a higher contrast in the image of the high-energy peak. As the temperature is increased even further, the "up-hill" temperature range,[6] the Boltzmann-like transfer from thicker, low-energy regions, to thinner, high-energy regions, starts to play a significant role and both images start to have similar contrasts. Though not shown in the Fig. 1, as the temperature is increased further, the "up-hill" transfer becomes more significant and the intensity ratio starts to increase. As the transfer is increased the finer details of the images are lost. The evolution of the contrast variations with increasing temperature is illustrated in Fig. 6. At 20 K we could resolve structures of about 0.5 μm, whereas at 110 K the finest features we could resolve are about 1 μm. This leads us to conclude that transfer between areas of different thickness plays an important role in determining the size of extended ML-flat islands.

It is of fundamental importance to reach an understanding of the detailed nature of the extended ML-flat islands, which appear to be flat on the ML level in the CL images. This is especially important since, based on transmission electron microscope investigations and PL investigations, it has been claimed that all QW interfaces have microroughness on different length scales.[19,20] We believe that our quantitative investigations of the lateral transfer within a single QW and of the temperature dependence of these processes[6] makes it possible to give a consistent explanation of the different experiments. The two models of the ML-flat islands are shown in Fig. 7.

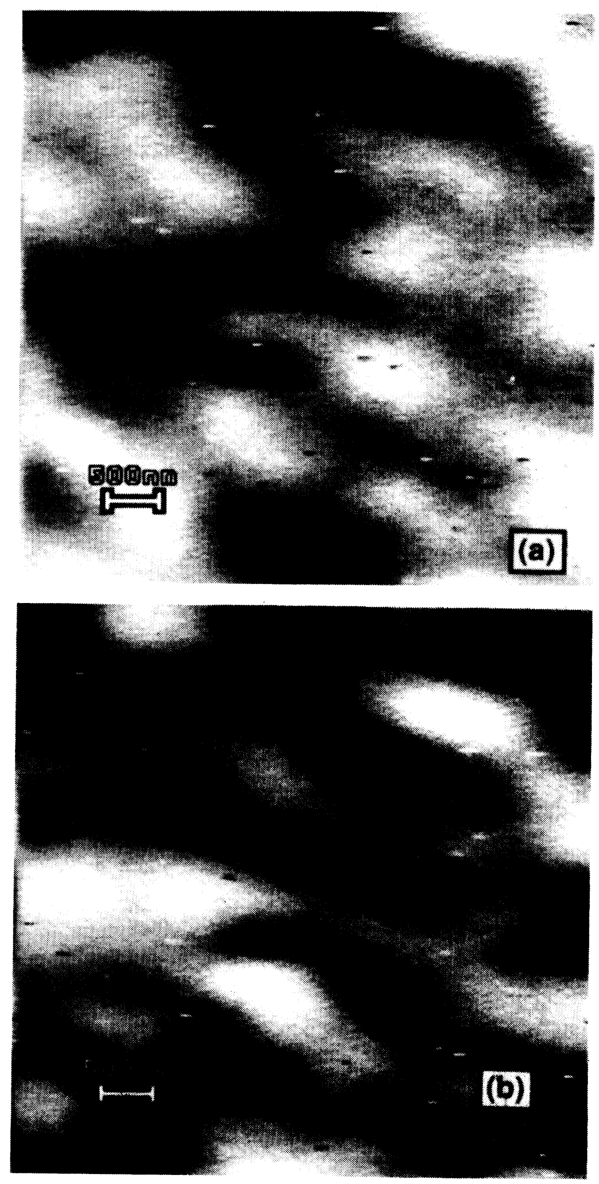

FIG. 4. Images recorded at 65 K of the same area show the expanded contrast variations of the 5-ML peak (a) and 4-ML peak (b). The same features as in the images of Fig. 2 can readily be observed. However, the images appear smeared, as compared to Fig. 2.

The first is the conventional model, with an extended island of, say, 5-ML-thick areas embedded inside a sea of 4-ML-thick QW material. In this model the effect of the temperature-dependent diffusion and transfer corresponds to varying the range of a "halo" extending around each such extended ML-flat island.

To resolve the controversy we would like to propose a second model, based on the model proposed by Warwick, Jan, and Ourmzad in Ref. 20. Our model is shown schematically in Fig. 7. We assume that no extended ML-flat island on the μm scale exists, but instead there are very small islands of 5-ML-flat material with dimensions larger than the exciton diameter, i.e., with sizes in the range of 30 nm to a few hundred nm, distributed in a sea of 4 ML. Clearly, no such microisland can be expected to be resolved either by PL imaging or by CLI, since even CLI is limited to observing objects $> 0.1-1$ μm (depending on sample and experimental conditions). In a

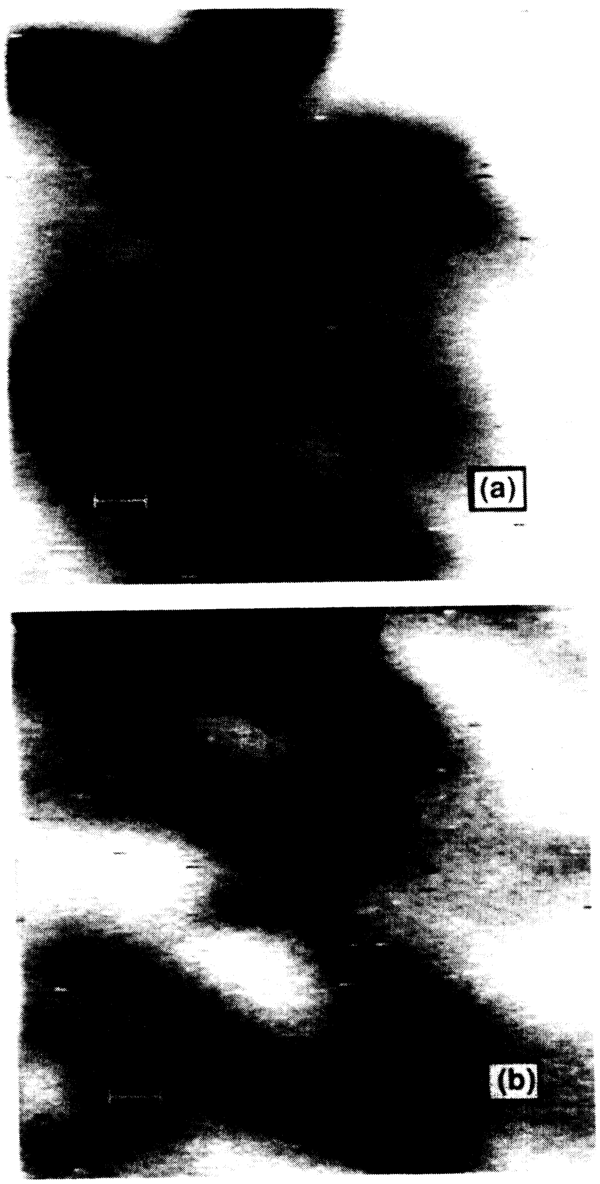

FIG. 5. Images recorded at 110 K of the same area show the expanded contrast variations of the 5-ML peak (a) and 4-ML peak (b). In this image most of the intensity variations are lost. The complementary behavior is not as obvious, and the smallest features that can be observed are about 1 μm.

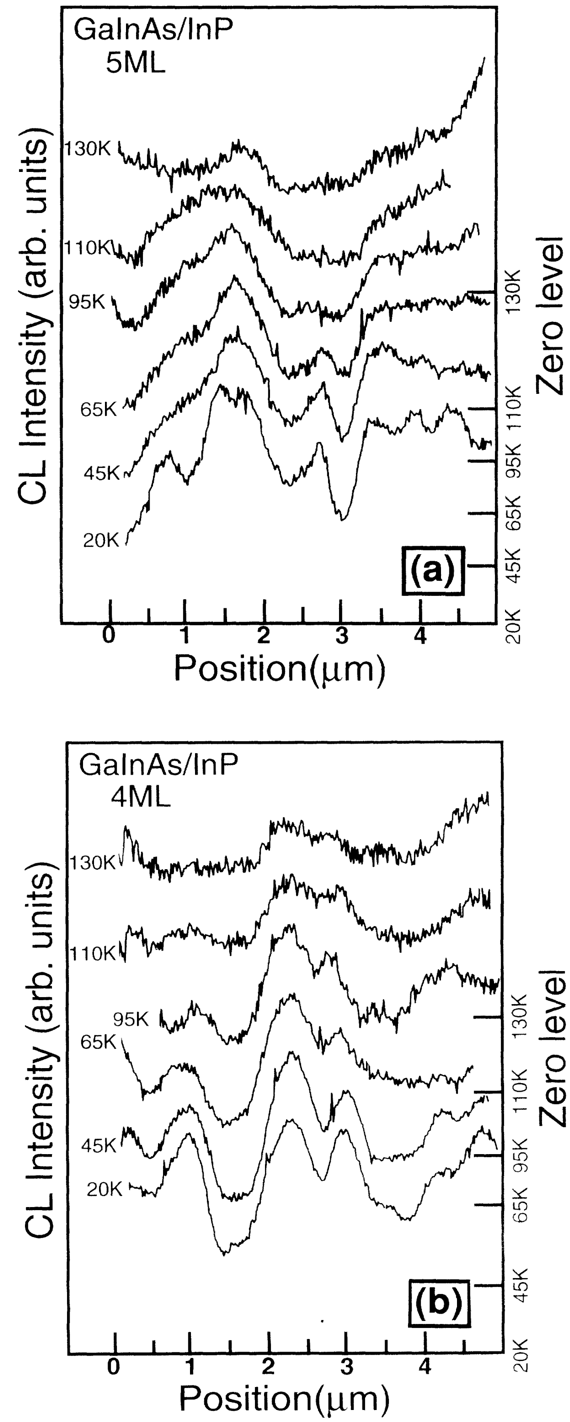

FIG. 6. Line scans of identical lines using the luminescence from (a) the 5-ML areas and (b) 4-ML areas of the QW, obtained at different temperatures. As the temperature is increased, the finer features of the line scans (≈ 0.5 μm) are lost. However, the large features (≈ 1 μm) are still present at 130 K.

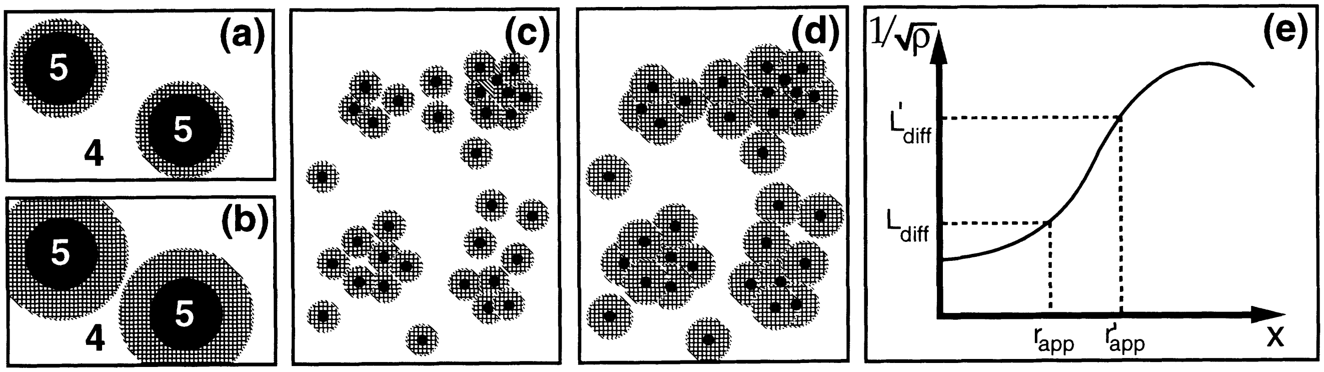

FIG. 7. In the conventional model of the detailed nature of the thickness variations in a single QW, there are extended ML-flat islands, either 4 (white) or 5 (black) ML thick. (a) With a one-way transfer (cross hatched) of excitons from thinner to thicker islands, the thicker islands appear larger in the CL images. (b) With increasing transfer they appear even larger. In the model suggested in this paper there is a varying density of 5-ML-thick microislands on a 4-ML-thick background. (c) The individual microislands are not resolved in the CL images, but when the density of the thicker microislands is above a critical value, depending on the diffusion length, these will appear as solid extended islands in the CL images. (d) With increasing diffusion length, the thicker islands appear even larger. (e) shows a graph, where the apparent size r_{app} of the islands depends mainly on the diffusion length.

simple model we assume that the microislands, with an area density of ρ, are regularly distributed in a regular hexagonal lattice. In this case the shortest distance between the centers of two adjacent microislands is $(\rho)^{-1/2}$, and the maximum distance from any one point to the center of the nearest microisland is $(3\rho)^{-1/2}$. However, it is reasonable to assume that the density of such microislands will vary, resulting in certain areas of the QW sample where ρ is sufficiently high (above a critical density ρ_c) for most of the excitation in the surrounding 4-ML material to transfer into 5-ML microislands by lateral exciton diffusion (of diffusion length L_{diff}). If the extension of the microislands, d_{MI}, is small compared to L_{diff}, we can neglect d_{MI}, and this corresponds to the condition for the critical density, $\rho_c = 1/(3L_{diff}^2)$. For the general case with randomly distributed microislands of various sizes, this can be approximated by

$$\rho_c = 1/(L_{diff}^2) \ . \tag{1}$$

On the other hand, for a low density of microislands the effect of the transfer will be quite negligible and the sea of 4-ML material will also be recorded as QW material of this thickness. For a density just below ρ_c, the QW will be recorded as a mixture of both thicknesses, resulting in diffuse borders of the apparent islands in the images. With a temperature-dependent and varying L_{diff} of the excitons, the critical microisland density ρ_c for Eq. (1) to be fulfilled will vary. We have thus replaced the (probably) oversimplified concept of extended ML-flat islands with the existence of areas of a QW within which the critical condition in Eq. (1) results in CL spectra and images which appear as if the area was almost an ideal 5-ML QW. This picture for the temperature dependence of what will be recorded as a 5-ML island is shown schematically in Fig. 7.

This model also gives an explanation of why only the contrast variations, and not the total intensity, behave complementarily; the intensity is not 0 between the apparent islands. Since the thicker apparent islands are not totally homogeneous and the transfer from the thinner areas relies on diffusion, a fraction of the excitation will stay in the thinner areas. As for the thinner areas in between the thicker apparent islands, even a low density of thicker microislands will, via diffusion, lead to a significant signal.

In summary, we have demonstrated that by increasing the transfer between monolayer-flat islands of different thicknesses, the resolution in the cathodoluminescence images has deteriorated. At 20 K features about 0.5 μm can be resolved, whereas at 110 K, where the transfer is more significant, the smallest features are about 1 μm. Based on these observations we have proposed a model of the nature of the extended monolayer-flat islands generally observed in cathodoluminescence images. In this model the thicker extended islands are made up of microislands, which are distributed with a locally varying density. In an area, where the density of thicker microislands is above a critical density, the dominating emission will be that of the thicker microislands. This is caused by transfer from thinner to thicker regions, and the whole area will be recorded as a thicker extended ML-flat island in the cathodoluminescence images. On the other hand, well below the critical density, the dominating emission is that of the thinner regions in between the thicker microislands, and the whole area will be recorded as a thinner extended ML-flat island.

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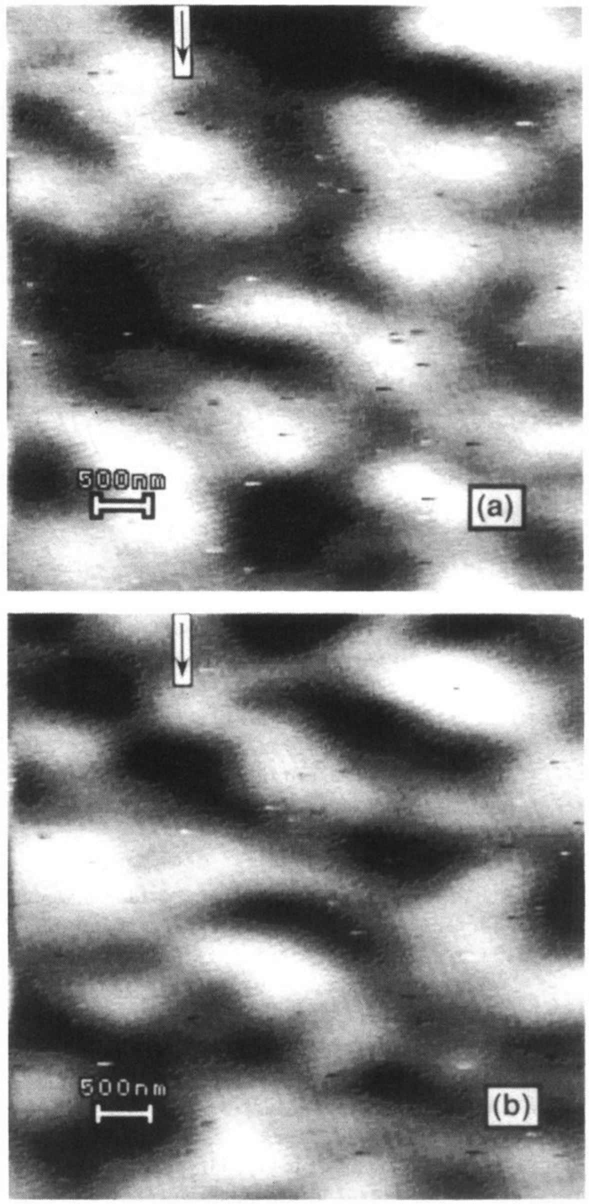

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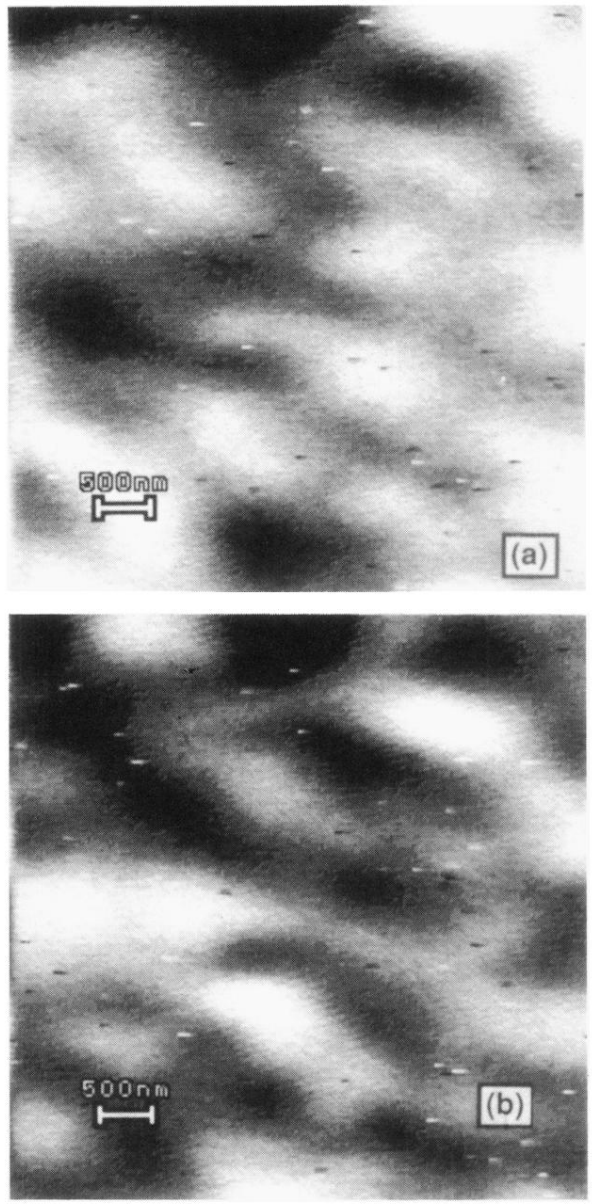

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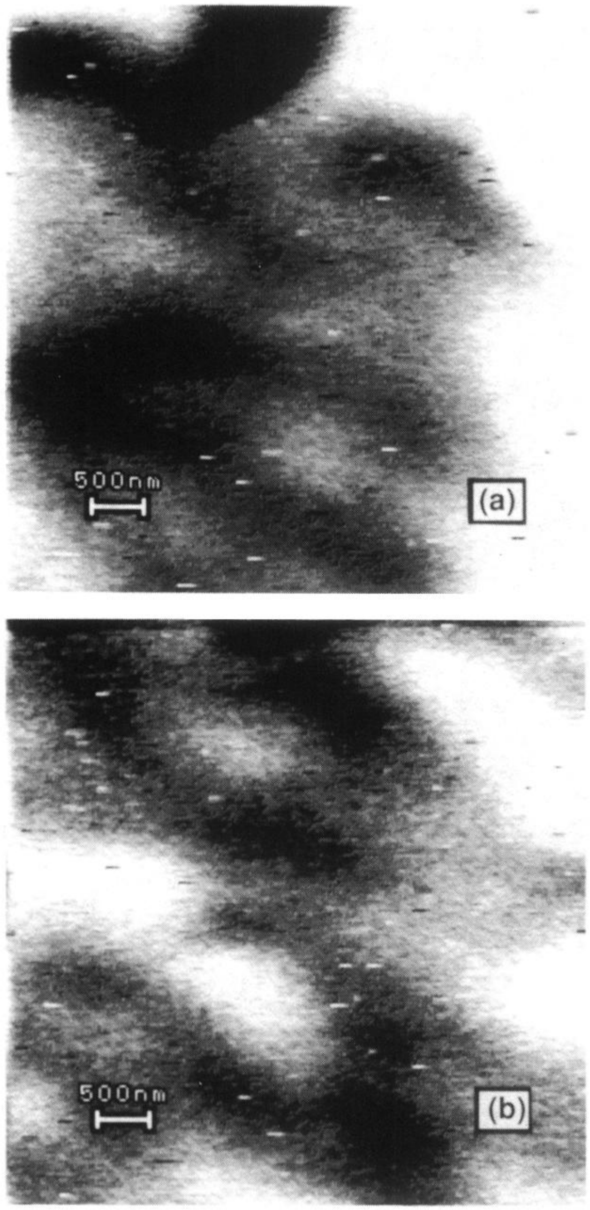

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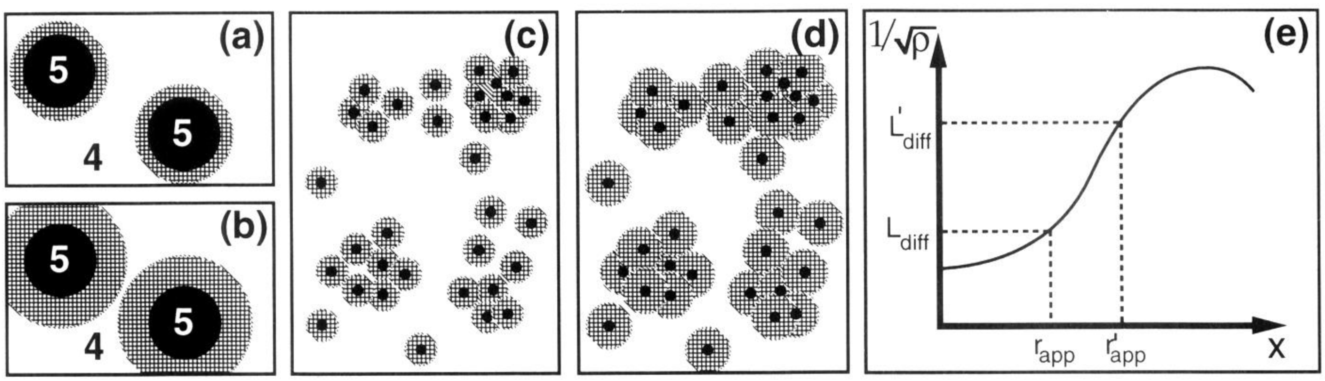

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