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Excitonic properties of weakly coupled GaAs single quantum wells investigated with high-resolution photoluminescence excitation spectroscopy

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Photoluminescence spectra of three weakly coupled GaAs quantum wells are measured as a function of the excitation energy with a high resolution of both the excitation and the detection energy. The spectra exhibit fine structures corresponding to island terraces as well as superfine structures associated with the coexistence of free and bound excitons. We present direct evidence for the transfer of carriers, excitons or exciton energy between the growth islands of the same well and adjacent wells. The relative occupation of free and bound exciton states depends strongly on temperature, carrier density, and excitation energy. A qualitative model, which explains these experimental results as a consequence of interwell interaction of excitons, is suggested. [S0163-1829(97)04544-X]

Photoluminescence (PL) properties of semiconductor structures can exhibit a considerable complexity since they are determined by photoexcitation (absorption), transfer of carriers, excitons, or energy, and radiative recombination (luminescence). While in PL spectroscopy at a fixed excitation energy the luminescence properties are investigated, photoluminescence excitation (PLE) spectroscopy at a fixed detection energy provides mainly information about the absorption properties. Each of these methods alone, however, can lead to a loss of essential information and consequently to misinterpretations. For example, even a small energetic shift of a PL line at a certain excitation energy appears in a PLE spectrum as an abrupt change of the respective signal. High-resolution PLE spectroscopy, i.e., the measurement of complete PL spectra as a function of the excitation energy with high spectral resolution of the excitation as well as the detection energy using an appropriate representation of the data, will provide more complete information, which is necessary for a reliable interpretation. At the same time, it can reveal novel effects.

High-quality semiconductor single quantum well (SQW) structures with atomically flat terraces can exhibit inter- and intra-growth-island exciton dynamics, which has been studied in detail with continuous-wave and time-resolved spectroscopy in GaAs/$Al_xGa_{1-x}As$ SQW's (Refs. 1–5) as well as in InAs/InP SQW's.[6,7] The dynamics can be described by rate equations, which take into account exciton transfer between islands as well as trapping of excitons at intrinsic defects. Furthermore, the observation of a significant transfer of carriers, excitons, or exciton energy between weakly coupled SQW's has been reported recently.[8,9] This anomalously large interwell transfer through relatively thick barriers is explained by dipole-dipole interaction of excitons[8] or by low potential channels within the barriers.[9]

Direct evidence of both the inter-growth-island and the interwell transfer of excitons can be provided by comprehensive spectroscopic (PL as well as PLE) investigations since resonant excitation of excitons in a certain growth island of a SQW can result in luminescence in other growth islands and wells. Therefore, we have investigated the PL properties of three weakly coupled GaAs SQW's in the excitation energy region between 1.55 eV, which corresponds to the energy gap of the widest well, and 1.76 eV, i.e., the energy gap of the barrier. As a further consequence of the coupling between excitons in adjacent wells, we found a strong correlation between the domination of bound excitons in two of the SQW's and an enhancement of the carrier density in the widest well.

The sample was grown by molecular-beam epitaxy using 2-min growth interruption at the GaAs well interfaces. The wells are 7.8, 5.5, and 3.5 nm wide and separated by 36-nm $Al_{0.17}Ga_{0.83}As$ barriers. Figure 1 displays a schematic band-edge diagram of the sample. For the PL experiments, the sample was mounted in a flow cryostat, which allows for a temperature stability of 0.1 K or better. The optical excitation was carried out with a tunable Ti:sapphire laser pumped by an Ar^+ laser. The PL signal was dispersed in a 1-m monochromator and detected with a cooled charge-coupled-device detector. The laser intensity was adjusted to 0.2 μW

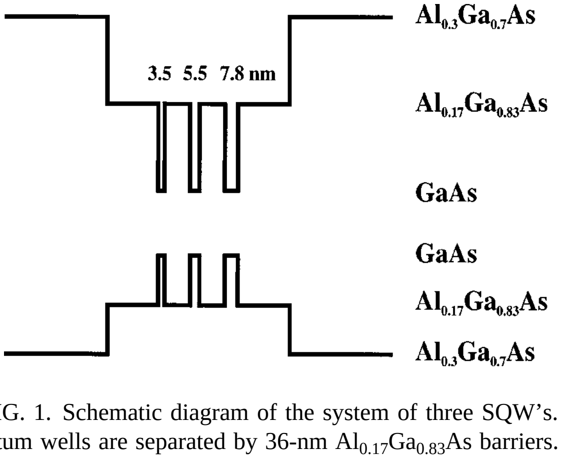

FIG. 1. Schematic diagram of the system of three SQW's. The quantum wells are separated by 36-nm $Al_{0.17}Ga_{0.83}As$ barriers. The whole system is embedded in two 72-nm $Al_{0.17}Ga_{0.83}As$ barriers and two $Al_{0.30}Ga_{0.70}As$ layers with a thickness of 190 (surface side) and 430 nm (substrate side) on an undoped GaAs(100) substrate.

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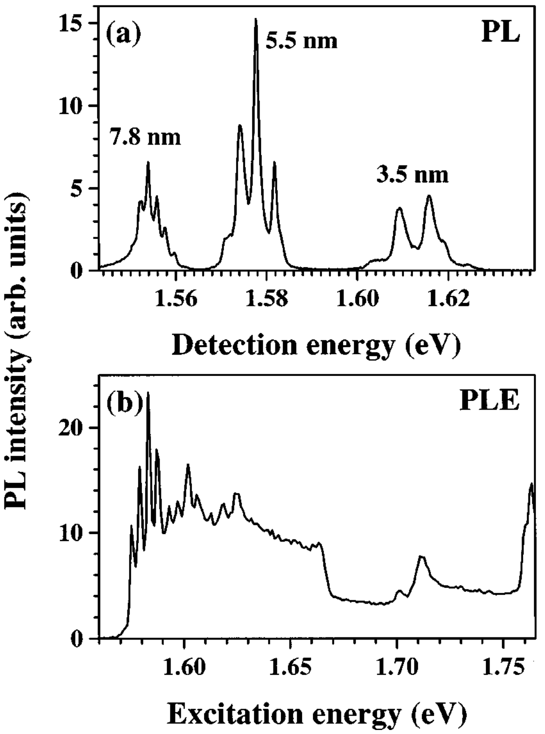

FIG. 2. (a) PL spectrum of the three weakly coupled SQW's at 5 K and an excitation energy of 1.74 eV. The well widths of the respective PL lines are indicated. (b) PLE spectrum of the same system as in (a) at 5 K and a detection energy of 1.576 eV within the 5.5-nm PL line. At excitation energies above 1.76 eV the excitation of electrons and holes in the $Al_{0.17}Ga_{0.83}As$ barriers leads to a considerable increase of the PL intensity.

focused to a diameter of about 50 μm except for the intensity-dependent measurements.

A typical PL spectrum measured at 5 K for an excitation energy of 1.74 eV is shown in Fig. 2(a). The PL lines of the three SQW's at 1.555, 1.578, and 1.614 eV are split into 4–6 peaks with an energy separation of 2, 4, and 6 meV, respectively, due to monolayer (ML) fluctuations in the SQW's. A detailed description of the PL spectrum can be found in Ref. 10. A PLE spectrum measured at 5 K for a detection energy of 1.576 eV, which corresponds to the PL energy of the 5.5-nm SQW, is presented in Fig. 2(b). A large number of transitions are observed. Near 1.58 and 1.62 eV, the absorption from the ground states in different monolayer islands of the 5.5 and 3.5-nm wells, respectively, are present. Near 1.60 eV, excitation from the highest light-hole subband into the lowest conduction subband of the 5.5-nm well is detected. Since the intensities of PLE spectra are usually assigned to the absorption strength, a very surprising observation is the strong reduction of the PLE signal at 1.664 and 1.712 eV. Furthermore, the smaller peaks at 1.7 and 1.71 eV cannot be assigned to a transition in the SQW's. The strong increase at 1.76 eV is due to the onset of absorption in the $Al_{0.17}Ga_{0.83}As$ barriers. Due to the two-dimensional density of states in SQW's, the absorption should remain constant or sharply increase if absorption into the next subband takes place. Therefore, the observation of a strongly reduced intensity far away from any excitonic transition clearly indicates that under certain conditions a single PLE spectrum is not a very reliable method to determine the absorption properties of quantum wells. Furthermore, time-dependent processes such as energy relaxation and carrier transport can influence the PLE spectra.

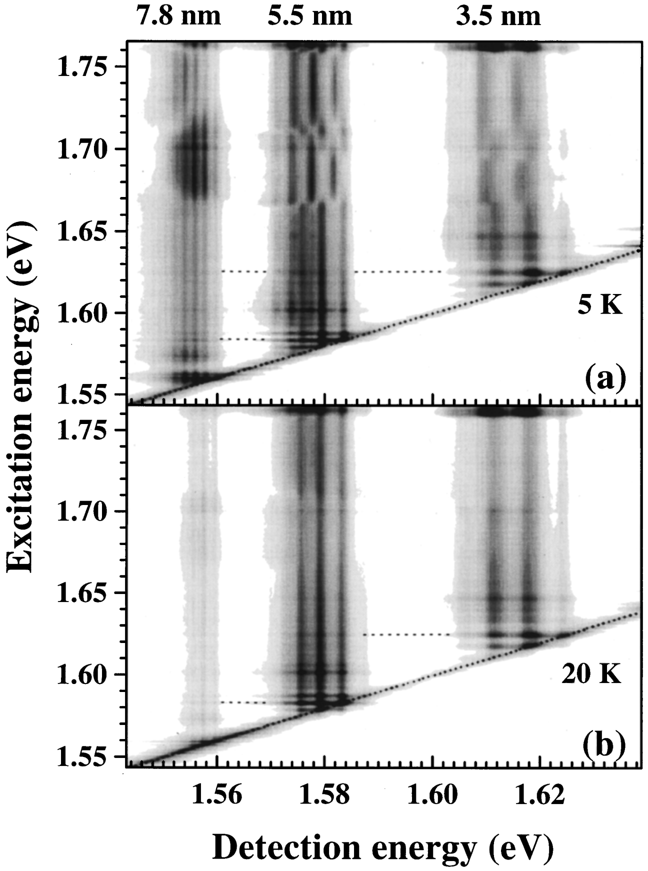

FIG. 3. High-resolution PLE spectra, i.e., PL intensity as a function of the detection as well as the excitation energy, of the three SQW's at (a) 5 K and (b) 20 K (light, small intensity; dark, large intensity). The well widths of the respective PL lines are indicated. The dashed lines mark excitation energies at which interwell coupling is directly visible in the spectra.

In order to resolve the unusual behavior of the PLE spectra, we have performed a detailed investigation of the PL spectra as a function of excitation energy, which we will refer to as high-resolution PLE. In Fig. 3(a) the PL intensity measured at 5 K is shown as a function of both the excitation and the detection energy using a gray scale representation. A horizontal cut in this graph corresponds to a single PL spectrum, a vertical cut to a single PLE spectrum. The PL spectra were recorded as a function of excitation energy using 1-meV steps. These high-resolution PLE spectra reveal that the strong decrease of the PLE signal in Fig. 2(b) is in fact a consequence of a small shift of the PL energy at certain excitation energies. In this representation, the splitting of the PL lines due to ML islands appears as 4–6 narrow, parallel lines in the vertical direction. At excitation energies corresponding to the PL energies of the SQW's, the electrons from the first heavy-hole subband are directly excited into the first conduction subband resulting in pronounced maxima of the PL intensity, which appear as horizontal lines. Weaker maxima in the PL intensity occur at excitation energies of 1.574, 1.603, and 1.646 eV, for which electrons from the first light-hole subband are excited into the first conduction subband. These weaker maxima also exhibit the signature of ML

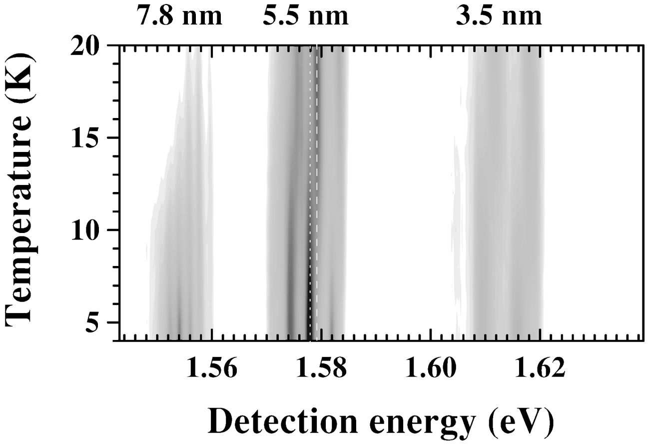

FIG. 4. PL spectra of the three SQW's for temperatures between 4 and 20 K using a temperature step of 1 K at an excitation energy of 1.74 eV and a laser intensity of 0.2 μW. The vertical lines indicate the energetic positions for a particular pair of submaxima in the 5.5-nm SQW with the dotted line corresponding to the bound exciton and the dashed line to the free exciton.

fluctuations since each maximum is split into a series of 4–6 peaks in the vertical direction.

The increased PL signal at certain excitation energies for a particular ML island within a SQW shows a correlation with other ML islands as well as other SQW's, which manifests itself as a narrow horizontal line in this representation. An example for a correlation within one SQW is the increased PL intensity at 1.6 eV in the 5.5-nm well. Coupling between SQW's is clearly visible in the PL line of the widest SQW at the excitation energy of about 1.583 eV, which corresponds to the excitation of a ML island in the 5.5-nm well. Similarly, a maximum appears in the 5.5-nm well at 1.624 eV as a result of excitation in the 3.5-nm well. At this excitation energy, a small enhancement can also be seen in the PL intensity of the 7.8-nm well. The former correlation is caused by the transfer of excitons between different ML islands within the same SQW, while the latter is due to transfer of carriers, excitons or exciton energy between the different SQW's since the horizontal lines cover sometimes the whole range of detection energies. Therefore, efficient transfer of carriers, excitons, or exciton energy between growth islands as well as between different SQW's takes place even at low temperatures in this system.

The most striking feature, however, is the abrupt decrease of the PLE signal at detection energies of 1.67 and 1.71 eV in Fig. 2(b), which is actually caused by an energetic shift of the PL lines by about 2 meV in the 5.5-nm well and 2.5 meV in the 3.5-nm well, as seen in Fig. 3(a). For the 7.8-nm well, the shift cannot be identified because it probably overlaps with the splitting caused by the ML fluctuations. This assumption is confirmed by the investigation of a reference sample, which has the same layer parameters, but was fabricated without growth interruption. The corresponding high-resolution PLE spectra do not show any ML splitting, but still exhibit the abrupt energetic shifts. Even in the excitation energy region below 1.67 eV, a coexistence of PL lines at the different energetic positions can occur. This is clearly visible in Fig. 3(a), e.g., for the two most intense PL maxima of the 5.5-nm well, which are split into peaks at 1.580 and 1.578 as well as at 1.576 and 1.574 eV. Hence the energetic shifts are in fact abrupt changes of the intensities of two submaxima, in which each PL maximum corresponding to a single ML island is split. This additional splitting for each SQW has been previously identified being due to the coexistence of free and bound excitons.[1–5,10–13]

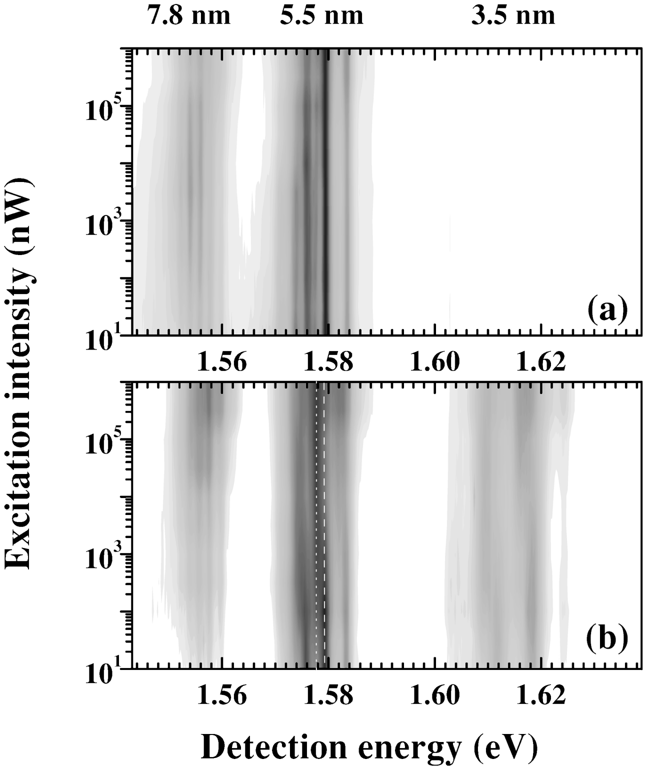

FIG. 5. PL spectra of the three SQW's as a function of the excitation intensity at (a) 5 K and (b) 15 K for an excitation energy of 1.61 and 1.74 eV, respectively. The intensity step was chosen to be equidistant on a logarithmic scale, i.e., 1, $\sqrt{10}$, 10, ... The minimum intensity of 10 nW corresponds to the intensity used in Figs. 2–4 divided by 20. The meaning of the dashed and dotted lines are explained in the caption of Fig. 4.

The energetic shift at 1.67 eV in the two narrower SQW's is correlated with a strong increase in the PL intensity of the widest SQW. However, the integrated intensity of the narrower SQW's remains unchanged. In the energy range between 1.67 and 1.71 eV (1.72 and 1.76 eV), resonant excitation from the second heavy-hole (light-hole) subband into the second electronic subband takes place for the different monolayer islands of the widest SQW, which explains the increased intensity of the PL of the 7.8-nm well in this energy range. It appears that the increased absorption in the widest well leads to a redistribution of excitons in the narrower wells, i.e., a transfer from free to localized excitons.

Since the energetic difference of the subsplitting has a value of about 2 meV, which corresponds to a temperature of 23 K, a significant temperature dependence should be expected over a range of about 20 K. We therefore recorded high-resolution PLE spectra for several temperatures between 5 and 20 K. At 10 K, i.e., after a temperature increase by only 5 K, the spectra exhibit remarkable changes in terms of the intensity distribution between the submaxima. Finally, at 20 K only the energetically higher states remain, as shown in Fig. 3(b), and the energetic shift has completely disappeared. A more detailed comparison of the 5- and 20-K spectra shows that in the excitation energy region, where at 5 K the low-energy peaks dominate, the intensity of the higher-energy submaxima in the 20-K spectra is somewhat reduced. However, the modulation is much weaker than at lower temperatures. In Fig. 4 the temperature dependence of the PL spectra for an excitation energy of 1.74 eV is shown between 4 and 20 K with a temperature step of 1 K. With increasing temperature, the intensity of the low-energy submaximum of each PL maximum decreases, while the high-energy submaximum increases in intensity. Surprisingly, the temperature, at which the two submaxima are of equal intensity is lower in the smaller growth islands (12 K for both the 5.5- and 3.5-nm wells) than in the wider ones (14 K), while the temperature dependence of the PL of the 5.5-nm well as a whole is very similar to that of the 3.5-nm well. For the 7.8-nm well, the low-energy peaks disappear in this temperature range.

In order to obtain further information about the nature of the subsplitting, we also increased the excitation intensity over five orders of magnitude by a factor of $\sqrt{10}$ at each step. Figure 5(a) shows the PL spectra at 5 K for an excitation energy of 1.61 eV as a function of the excitation intensity. At this excitation energy the high-energy submaxima dominate in the low-intensity regime [cf. Fig. 3(a)]. The relative PL intensities of the submaxima are almost independent of the excitation intensity, i.e., of carrier density. No shift towards the low-energy submaxima can be observed. The situation changes drastically when the PL spectra are recorded at 15 K as a function of excitation intensity, as shown in Fig. 5(b) for an excitation energy of 1.74 eV. At this excitation energy the intensities of the low-energy submaxima, which dominate at 5 K, decrease with increasing temperature (cf. Fig. 4), so that at around 20 K the high-energy submaxima dominate. This thermal activation of the high-energy submaxima is partially compensated for at higher excitation intensities beginning at about 10 times the minimum intensity for the wider growth islands of both, the 3.5-nm and 5.5-nm wells, and about a 100 times the minimum intensity for the smaller islands. An increase of the intensity over four orders of magnitude results again in a complete domination of the low-energy submaxima as observed for this excitation energy at 5 K and low intensity. Note that a lower activation temperature (for the smaller growth island) requires a higher intensity necessary for the compensation and vice versa, i.e., a higher activation temperature for the wider islands corresponds to a lower intensity.

The observation of submaxima pairs with a complementary behavior of each submaximum with respect to temperature and carrier density allows us to rule out the existence of regions in the SQW's with different parameters, e.g., different barrier heights due to changes in the barrier composition. Although the 2-meV energy separation of the submaxima approximately agrees with the binding energy reported for biexcitons in GaAs SQW's,[14–17] also the formation of biexcitons can be excluded since they should exhibit a superlinear intensity dependence on excitation power, which is not observed at 5 K. The temperature dependence rather suggests the simultaneous presence of free (at higher energy) and bound (at lower energy) excitons at certain excitation energies. In addition, the compensation of the thermal activation by the increasing carrier density, as discussed above, suggests that also exciton-exciton interaction plays an important role in the interplay of free and bound excitons.

In the framework of this qualitative model, the correlation of the enhanced PL intensity in the widest well with the domination of bound excitons in the narrower wells suggests that, in addition to the observed transfer of carriers, excitons, or exciton energy, another coupling effect through the barriers is present. Due to the interwell interaction of excitons, e.g., the dipole-dipole interaction, the larger density of carriers in the widest well increases the capture probability of free excitons or reduces the probability for thermal activation in the narrower wells. Increasing the temperature results in an increased thermal activation of bound excitons so that the spectra of the narrower wells are dominated by free excitons. At these higher temperatures an increase in excitation power enhances again the density of bound excitons.

For a quantitative interpretation of the experimental results, the interplay of different carrier lifetimes including radiative lifetimes, trapping time (the characteristic time for the formation of bound excitons), and release time (the time for thermal activation) has to be taken into account. An alternative explanation of the correlation could be the existence of low potential channels in the barriers, which possibly mediate the coupling. In this case, states within the barrier are expected to be occupied by electrons or holes so that additional potential fluctuations are created.

In conclusion, we have demonstrated that high-resolution PLE spectroscopy can provide direct information about intra- and interwell coupling of excitons in GaAs/$Al_{0.17}Ga_{0.83}As$ SQW's. We observed clear evidence for a transfer of carriers, excitons, or exciton energy between growth islands within the same wells and also between different wells, which are weakly coupled. Furthermore, a coexistence of free and bound excitons was detected with a relative occupation, which depends strongly on excitation energy, temperature, and excitation intensity. A qualitative model based on coupling mechanisms, which takes into account transfer processes of carriers, excitons, or exciton energy as well as exciton-exciton interaction, has been proposed to explain the experimental observations.

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