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Reduced binding energy of the symmetric heavy-hole exciton in GaAs/$Al_xGa_{1-x}As$ symmetric coupled double quantum wells

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Discrete peaks, which are identified as the excited 2s state of the $n=1$ symmetric heavy-hole exciton, are observed in GaAs/$Al_xGa_{1-x}As$ symmetric coupled double-quantum-well (SCDQW) samples by combining unpolarized and polarized photoluminescence-excitation measurements. The 1s-2s splitting of symmetric heavy-hole excitons is accurately determined in the SCDQW's. The 1s-2s splitting is significantly reduced compared to uncoupled quantum wells.

Coupled double quantum wells (CDQW's) are interesting not only for their fundamental physics but also for applications.[1] If the barriers in the double-quantum-well structures are sufficiently narrow, there will be an interaction between the two wells. The electronic states corresponding to single-well one-particle states split into symmetric and antisymmetric states. A schematic diagram of the band structure is shown in Fig. 1 for a symmetric CDQW (SCDQW) structure. Energy levels and allowed optical transitions under flat-band condition are also indicated. The exciton states are formed from either symmetric or antisymmetric hole and electron one-particle wave functions, and are consequently denoted "symmetric" and "antisymmetric" excitons. Both types of excitons have even parity. The splitting of the energy levels due to the coupling between the two wells is very sensitive to the barrier width, but it is also dependent on the well widths. The exciton effects and band mixing in CDQW's have often been neglected, however, recently their importance was realized.[2-4] There has been no report up to now, to our knowledge, on experimental determination of the exciton binding energy in SCDQW systems.

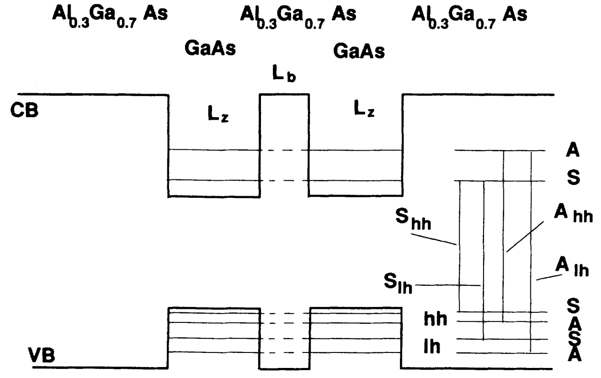

FIG. 1. Schematic diagram of the band structure of a SCDQW and the SCDQW energy levels. Allowed transitions are also shown.

In the previous studies on noninteracting $Al_xGa_{1-x}As$/GaAs quantum wells (QW's),[5-8] the experimental data of the 1s-2s exciton energy splitting combined with theoretical calculations of the 2s exciton binding energy is used to determine the total exciton binding energy. This is shown to be a precise way to obtain the 1s exciton binding energy.

The energy position of the 2s state of excitons is often difficult to determine in optical spectra, due to its weak nature and the spectral overlap with the light-hole exciton transition. In this Rapid Communication we demonstrate that a combination of ordinary photoluminescence excitation (PLE) and polarized PLE (PPLE) measurements allows us to accurately obtain the energy separation of 1s-2s exciton transitions. By using this technique a clearly resolved peak, which we interpret as the 2s state of the symmetric heavy-hole excitons in a GaAs/$Al_xGa_{1-x}As$ SCDQW has been observed. The observation of the 2s state allows precise determination of the 1s-2s splitting in the SCDQW structures, and thereby provides important data for the determination of the 1s exciton binding energy in these systems.

The samples were grown in a Varian Gen II modular molecular-beam epitaxy machine on semi-insulating substrates oriented in the [001] direction. Three samples

TABLE I. Barrier (L_b) and well widths (L_z) of the symmetric coupled double-quantum-well samples which were used in this study. The width is given in Å and in monolayers (ML) of GaAs, respectively. The barrier material is $Al_{0.3}Ga_{0.7}As$.

Structure	Sample *A*		Sample *B*		Sample *C*	
Width	Å	ML	Å	ML	Å	ML
Barrier	14.2	5	14.2	5	19.8	7
Well 1	39.6	14	59.4	21	39.6	14
Well 2	79.2	28	99.1	35	59.4	21
Well 3	124.5	44	150.0	53	79.2	28

have been used in this study; each sample contains three different SCDQW structures with the same thickness of the $Al_xGa_{1-x}As$ barrier layer. Sample *A* contains three double wells with nominal widths $L_z = 124.5$, 79.2, and 39.6 Å, respectively, while sample *B* contains three double wells with nominal widths $L_z = 150.0$, 99.1, and 59.4 Å, respectively. All the double wells in both samples have nominal barrier widths $L_b = 14.2$ Å. Sample *C* contains three double wells with nominal width $L_z = 79.2$, 59.4, and 39.6 Å, with a nominal barrier width $L_b = 19.8$ Å. The parameters of the samples are summarized in Table I. The detailed growth conditions of the samples were reported in Ref. 3.

The PL, PLE, and PPLE measurements were done in an exchange-gas-type He cryostat, where the temperature could be varied from 1.8 K up to room temperature. The excitation source was either an Ar^+ laser (5145 Å) or a tunable sapphire:Ti solid-state laser pumped with an Ar^+ laser, which covers the wavelength range from 700 to 1000 nm. A double-grating monochromator and a GaAs photomultiplier were used to disperse and detect the PL signals. For the polarized PLE experiments we have used a photoelastic modulator, whereby the intensity difference between σ^+ and σ^- polarization can be measured in PPLE experiments.

In the PL spectra only one emission from each SCDQW appears. The emission corresponds to the lowest symmetric heavy-hole excitons. The PLE spectra are measured with detection at the low-energy side of the symmetric heavy-hole exciton of each SCDQW. Two typical PL and PLE spectra are shown in Fig. 2 for the 79.2-Å SCDQW's with barrier width $L_b = 14.2$ and 19.8 Å, respectively. Increasing the barrier thickness of SCDQW means reducing the coupling strength between the two wells, resulting in a decrease of the splitting between symmetric and antisymmetric states. The main exciton transitions related to heavy holes (symmetric and antisymmetric light holes) are denoted as S_{hh}, A_{hh}, S_{lh}, and A_{lh}, respectively (see Fig. 1), corresponding to labels 1, 3, 2, and 4, respectively, in a previous publication.[3] To explain the change of the relative oscillator strengths of the excitons with varying coupling strength, exciton effects and mixing of different exciton states must be taken into account in detail.[3]

Additional small peaks sometimes appear at the position of the exciton energy levels for adjacent quantum wells. They are labeled *T* in Figs. 2(b) and 3(b). These peaks are explained as charge-transfer effects due to the fact that resonant optical excitation causes a relatively high concentration of charge carriers in the adjacent 59.4-Å wells. Some of these charge carriers will be excited thermally or optically over the $Al_xGa_{1-x}As$ barrier layer and are transferred to the probed quantum well where they finally recombine.[3]

In addition to the four main exciton transitions (see Fig. 2), a discrete peak at the high-energy side of S_{hh} appears, which is interpreted as the excited 2*s* state of S_{hh} excitons. To support our interpretation the PPLE spectra are measured, which are shown in Fig. 3, compared with ordinary PLE spectra for a 79.2-Å SCDQW with two different $Al_xGa_{1-x}As$ barrier thicknesses. Since the transitions related to heavy-hole excitons and light-hole excitons have different polarization, they show different behavior in PPLE spectra, i.e., a heavy-hole exciton transition has opposite sign from a light-hole exciton transi-

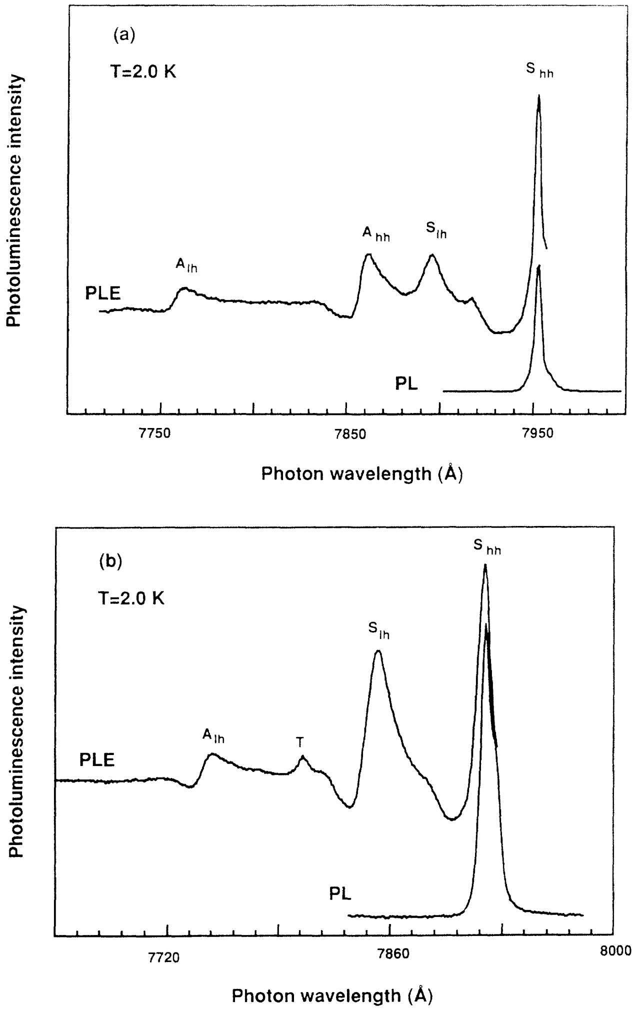

FIG. 2. PL and PLE spectra measured at 2.0 K for a 79.2-Å SCDQW (a) with 14.2-Å barrier and (b) with 19.8-Å barrier. The excitation wavelength is 7200 Å for PL measurements. In the PLE measurements, the detection was at the low-energy side of the PL peak. Peak *T* is due to transfer from the symmetric heavy-hole exciton in the 59.4-Å SCDQW's.

TABLE II. The energy separation between 2*s* and 1*s* exciton states (meV).

SCDQW This work					Uncoupled QW previous work			
L_z (Å) SCDQW	L_b (Å) ($x=0.3$)	1*s*-2*s* S_{hh}	1*s*-2*s* S_{lh}	1*s*-2*s* A_{hh}	L_z (Å) QW	x	1*s*-2*s* hh	1*s*-2*s* lh
99.1	14.2	5.65			100[a]	0.35	7.5	8.5
					100[b]	0.22	8.0	9.1
79.2	14.2	6.50		5.72[c]	80[a]	0.35	8.1	9 or 10
59.4	14.2	7.79			92[d]	0.35	8.5	10.2
79.2	19.8	6.75	7.82		75[d]	0.40	9.5	10.3
59.4	19.8	7.98	8.07		64[b]	0.22	9.4	10.6
					45[b]	0.22	10.2	11.4

[a] Reference 5.
[b] Reference 8.
[c] The assignment of the 2*s* state of the A_{hh} exciton is tentative, and the splitting of 1*s*-2*s* has a large uncertainty due to the less-well-defined peak position.
[d] Reference 6.

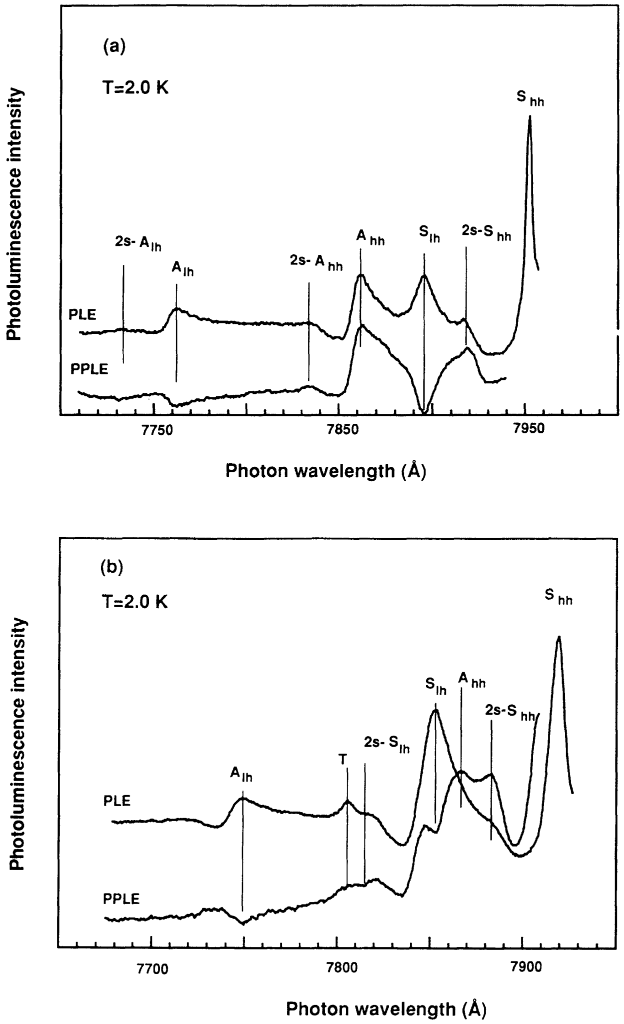

FIG. 3. PLE and PPLE spectra measured at 2.0 K for 79.2-Å SCDQW (a) with 14.2-Å barrier and (b) with 19.8-Å barrier. The detection used for PLE spectra was the same as in the PLE measurements of Fig. 2. In the PPLE measurements, the detection is always at the maximum intensity of the PL peak in order to enhance the intensity and polarization effect.

tion in PPLE spectra. Furthermore, the excited 2*s* states of heavy-hole excitons (light-hole excitons) has the same polarization as heavy-hole excitons (light-hole excitons), so they should have the same sign in PPLE spectra. The results shown in Fig. 3 are consistent with our interpretation. In addition to the 2*s* state of S_{hh} excitons, there are a few not-well-defined peaks (or dips). The one at the high-energy side of A_{hh} (or S_{lh}) is tentatively interpretated as the excited 2*s* state of A_{hh} (or S_{lh}). For some SCDQW's there is also a peak at the high-energy side of A_{lh}, which we tentatively interpret as the excited 2*s* state of A_{lh}. Our experimental data show that when the well width is larger than 100 Å, the 2*s* states of the excitons are very difficult to observe. The values of the splitting between 2*s* and 1*s* states obtained from PLE and PPLE spectra are shown in Table II. The results from isolated quantum wells, which exist in literature, are also shown in the last four columns for comparison. These results are illustrated graphically in Fig. 4.

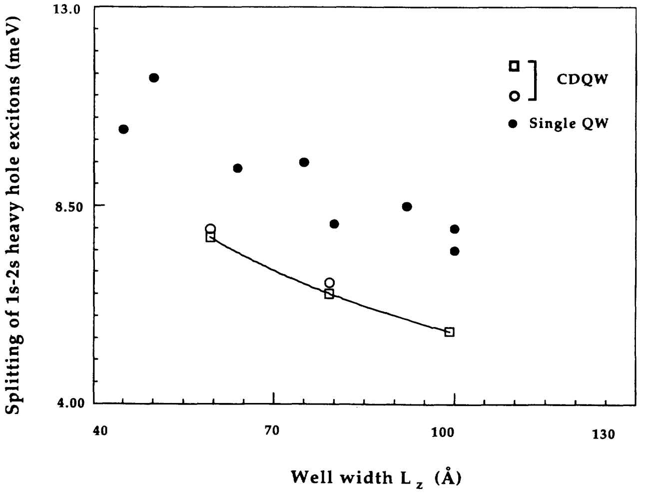

FIG. 4. The splitting of 1*s*-2*s* symmetric heavy-hole excitons. The CDQW data are from this work on SCDQW structures with 14.2-Å (□) and 19.8-Å (○) barrier layer, respectively. The single QW data are from literature (see Table II). The solid line is used to guide the eye.

The results show a large reduction of the 1*s*-2*s* splitting for the CDQW compared to the single quantum well. Theoretical calculations for CDQW's are more complicated than in the single QW case, since more electron and hole subbands are interacting with each other. To our knowledge no calculations of the 2*s* exciton state in SCDQW structures exist so far. Qualitatively a reduction of the 1*s*-2*s* splitting for the SCDQW's is expected from the density distributions of carriers in the wells, since electrons and holes are much more delocalized in SCDQW's than in single QW's. The reduced 1*s*-2*s* splitting for SCDQW's with 14.2-Å barrier as compared to uncoupled QW's is larger than 1.0 meV. Comparing the same well width of the SCDQW's with different barrier thickness, the 1*s*-2*s* splitting increases with increasing barrier thickness. If the binding energy of the 2*s* exciton state, which is not very sensitive to the change of exciton wave functions, is assumed to be the same for a SCDQW and a QW, the difference in binding energy of heavy-hole excitons for QW and SCDQW with 14.2-Å barrier layer varies from about 1.0 up to 3.0 meV, when the well widths change from 100 to 60 Å. However, to accurately determine the binding energy of symmetric heavy-hole excitons in SCDQW's, the experimental value of 1*s*-2*s* splitting should be supplemented by the binding energy of 2*s* excitons, which has to be calculated theoretically. The results also show that the 1*s*-2*s* splittings are not equal for S_{hh} and A_{hh} excitons for the 79.2-Å SCDQW with 14.2-Å barrier (see Table II). This unequal splitting is probably due to a different degree of delocalization of the corresponding S_{hh} and A_{hh} exciton wave functions.

It is worth noticing that the excited 2*s* state of the excitons is conveniently determined by combining PLE and PPLE measurements. The 2*s* state energy position is otherwise often difficult to determine in ordinary PLE measurements due to the strong light-hole excitons.

In summary we have presented optical measurements on the excited 2*s* states in SCDQW structures. The splitting between 2*s* and 1*s* excitons is accurately obtained for SCDQW's with different well widths. In comparison with an uncoupled QW with the corresponding well width, a strong reduction of the splitting of 1*s*-2*s* exciton states in the SCDQW is observed. We have demonstrated that the combination of PPLE and PLE is a useful method to study excited states of excitons. These measurements provide a precise experimental value to determine the exciton binding energies in the SCDQW system. The data presented here also provide a basis to develop a more sophisticated theory to calculate the electronic states in SCDQW structures.

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