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Observation of excitonic effects on electroabsorption in coupled quantum wells

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An experimental study was made on the electroabsorption of asymmetric GaAs double quantum wells separated by thin AlAs tunnel barriers to better understand the optical properties in the coupled systems. It was clearly shown that the field-dependent absorption characteristics, i.e., the anticrossing properties, were not fully explained by the predictions for the optical transitions between the quantized electronic levels but better interpreted by taking account of the contribution of the exciton binding energies, i.e., in terms of tunneling between the excitonic states in the adjacent wells.

The coupling effects in semiconductor superlattice (SL) or multiple-quantum-well (MQW) structures in electric fields cause significant phenomena in optical properties, e.g., the Wannier-Stark localization[1-3] and the quantized level mixing.[4-6] The application of the coupling effects to optoelectronic devices[7-11] might bring about fruitful results as with the quantum-confined Stark effect (QCSE).[12]

In conventional quantum-well structures, it is well known that the Coulomb interactions of electron-hole pairs, i.e., the exciton binding energies, are enhanced since the electrons and holes are confined together in very narrow regions.[13] For coupled-quantum-well (CQW) systems, the excitonic effect on the optical transitions would also be of great interest.[1,8,14-17] However, the experimental investigation of the effects has been scarcely reported so far. In this paper, we obtained meaningful information on the excitonic effects from absorption spectra of the CQW structures to better understand the field-dependent characteristics, i.e., anticrossing behavior, of the optical transitions.

We investigated the double-quantum-well (DQW) structures grown by molecular-beam epitaxy (MBE) on Si-doped GaAs substrates. The DQW element consists of 100- and 60-Å-thick GaAs wells separated by 4- or 8-Å-thick AlAs tunnel barriers. Four pairs of the asymmetric DQW's were embedded in undoped AlAs-GaAs SL regions which were sandwiched between Be- and Si-doped AlAs-GaAs SL regions. The epitaxial wafers were processed into *p-i-n* photodiodes with much the same configurations as reported earlier.[4]

For an asymmetric uncoupled DQW system, $E^{WN(NW)}_{nmh(l)}$ stands for an interwell transition between the nth electron level in the wider (narrower) well $e^{W(N)}_n$, and the mth heavy- (light-) hole level in the narrower (wider) well $h(l)^{N(W)}_m$, while a usual intrawell transition between the nth electron and the mth heavy- (light-) hole levels in the wider (narrower) well, $e^{W(N)}_n$ and $h(l)^{W(N)}_m$, is referred to as $E^{W(N)}_{nmh(l)}$.[4,5] The symbols also represent the energy values of the optical transitions. We utilize the notations expansively for the CQW system as well. Figure 1 shows the energy-band diagrams of the DQW intrinsic region. It should be noted that the energy band of the intrinsic region is inherently bent because of the existence of the built-in field. By varying the internal field of the intrinsic regions, we can observe phenomena associated with the quantized level mixing or crossing in the optical spectra.[4-6]

We measured the photocurrent (PC) spectra of the diodes at 77 K in electric fields perpendicular to the layers. Figure 2 shows the spectra of the DQW structure with the 4-Å-thick tunnel barriers at various external bias voltages V_{ex}. To elucidate the peak shift behavior, the peak wavelengths (energies) were summarized as a function of V_{ex} in Fig. 3(a). The absorption peaks were identified by considering the intersection of the lowest- and the first-excited electron levels (e^W_1 and e^N_1) (Refs. 5 and 6) as well as by comparison with the PC spectra of the reference samples. The assigned origins are denoted in Figs. 2 and 3. Thus, the results can be clearly classified into three anticrossing pairs. Also, we can find partial anticrossing characteristics of an intrawell-like forbidden transition of E^W_{12h}. The large energy repulsion (~20

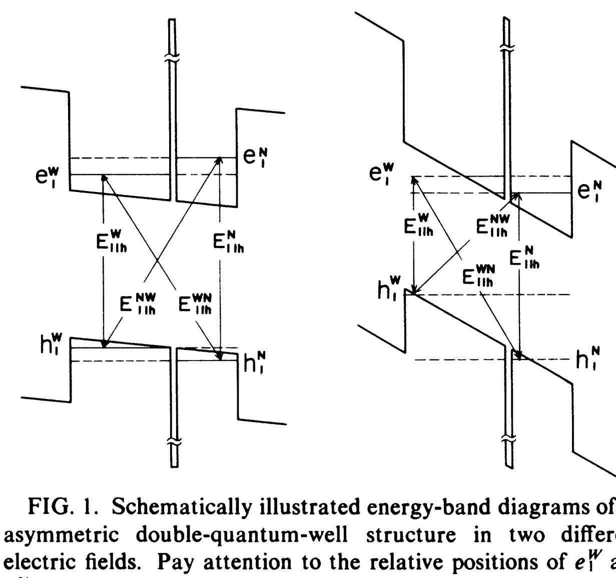

FIG. 1. Schematically illustrated energy-band diagrams of an asymmetric double-quantum-well structure in two different electric fields. Pay attention to the relative positions of e^W_1 and e^N_1 quantized levels. For clarity, the light-hole levels and the higher-lying levels are eliminated. Some optical transitions are inserted.

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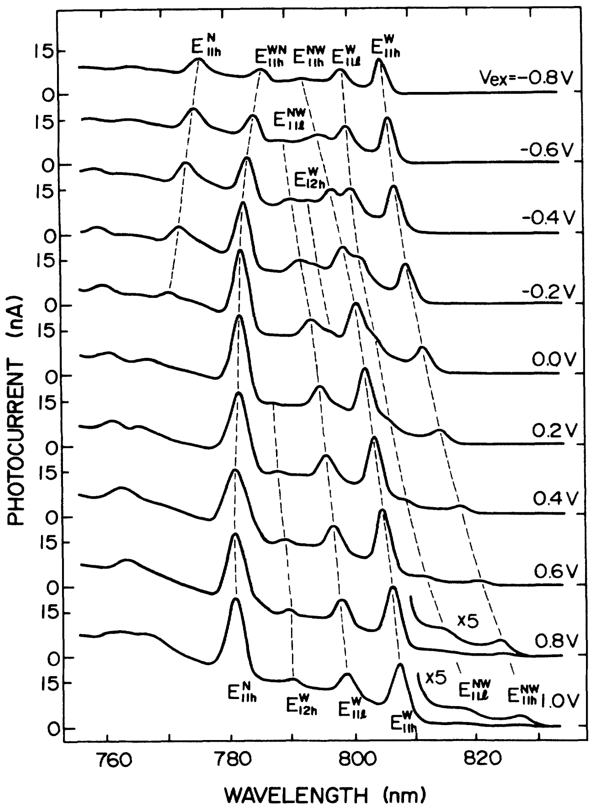

FIG. 2. Photocurrent spectra of an asymmetric coupled-quantum-well structure at different bias voltages. Four pairs of 100- and 60-Å-thick GaAs wells, separated by 4-Å-thick AlAs barriers, are embedded in the intrinsic region sandwiched between *p*- and *n*-type AlAs-GaAs superlattices. Measurements were made at 77 K. Reverse bias is positive.

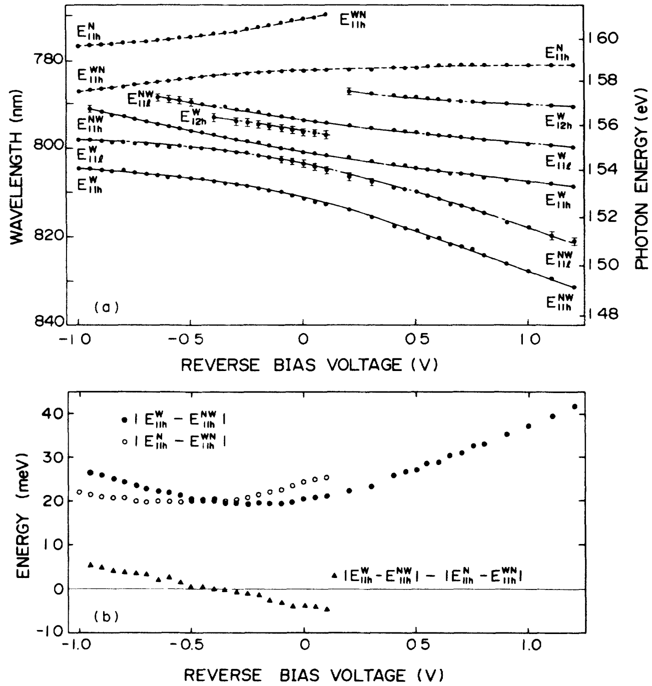

FIG. 3. (a) Absorption peak wavelengths (energies) as a function of bias voltage V_{ex}. The observed optical transitions are divided into four pairs. (b) The V_{ex} dependence of the separation energies between the paired optical transitions associated with the h_1^W heavy-hole level, $|E^{W}_{11h}-E^{NW}_{11h}|$, $|E^{N}_{11h}-E^{WN}_{11h}|$, and their differences, $|E^{W}_{11h}-E^{NW}_{11h}|-|E^{N}_{11h}-E^{WN}_{11h}|$.

meV) observed in the anticrossing characteristics originates from the very thin tunnel barrier thickness, i.e., the strong interwell coupling. In addition, remarkable blue shifts were observed for the transitions associated with the h_1^N quantized level (E^{N}_{11h} and E^{WN}_{11h}, E^{WN}_{11h} and E^{N}_{11h}).

If the observed optical transitions were the transitions between the quantized levels (the band-to-band transitions), the following relation should be satisfied:

$$|E^{W}_{11h(l)}-E^{NW}_{11h(l)}| = |E^{N}_{11h(l)}-E^{WN}_{11h(l)}| = |e_1^W - e_1^N| \ . \tag{1}$$

From the PC spectra, we can estimate both the separation energies associated with the h_1^W and h_1^N heavy-hole levels, i.e., $|E^{W}_{11h}-E^{NW}_{11h}|$ and $|E^{N}_{11h}-E^{WN}_{11h}|$. Figure 3(b) indicates the V_{ex} dependence of the separation energies (solid and open circles). It is obvious that the experimental result is not represented by Eq. (1). In this connection, it should be noted that the bias voltages which give the minimum separation energies are appreciably different. The energy values of $|E^{W}_{11h}-E^{NW}_{11h}|-|E^{N}_{11h}-E^{WN}_{11h}|$ are plotted as a function of V_{ex} in Fig. 3(b) as well (solid triangles).

This marked result should be interpreted in terms of the excitonic effects. In this work, we will give a fundamental explanation for the experimental results by using a simple model. We start with considering the uncoupled system where the e_1^W and e_1^N quantized levels intersect each other at V_C. The solid lines in Fig. 4(a) indicate the energies of the two pairs of transitions between the e_1^W or e_1^N levels and the h_1^W or h_1^N levels as a function of V_{ex} (where, for simplicity, the usual Stark shifts[12] are neglected). In the uncoupled DQW system, the exciton binding energies E_B^{NW} and E_B^{WN} of the interwell E^{NW}_{11h} and E^{WN}_{11h} transitions (which really cannot be observed) should be evaluated as zero. On the other hand, for the intrawell E^{W}_{11h} and E^{N}_{11h} transitions, the sufficient exciton binding energies E_B^W and E_B^N should be considered. The energies of the excitonic E^{W}_{11h} and E^{N}_{11h} transitions are shown by the dashed lines in Fig. 4(a) (where the bias-voltage dependence of the binding energies[12] is also ignored). We name the two crossing voltages of the paired excitonic transitions (E^{W}_{11h} and E^{NW}_{11h}, E^{N}_{11h} and E^{WN}_{11h}), V_W and V_N, respectively. Last, we introduce the excitonic coupling effects,[18] and thus the excitonic transition energies in the coupled system may be represented by the dash-dotted lines. It should be noted that $V_N < V_C < V_W$.

In Fig. 4(b), $|E^{W}_{11h}-E^{NW}_{11h}|$ and $|E^{N}_{11h}-E^{WN}_{11h}|$ are shown for the excitonic transitions in the uncoupled system (dashed lines) and the coupled system (dash-dotted lines). The differences between the separation energies, $|E^{W}_{11h}-E^{NW}_{11h}|-|E^{N}_{11h}-E^{WN}_{11h}|$, are indicated in Fig. 4(c). The bias voltage V_C', where $|E^{W}_{11h}-E^{NW}_{11h}| = |E^{N}_{11h}-E^{WN}_{11h}|$ for the excitonic transitions, may be shifted from V_C due mainly to the difference between E_B^W

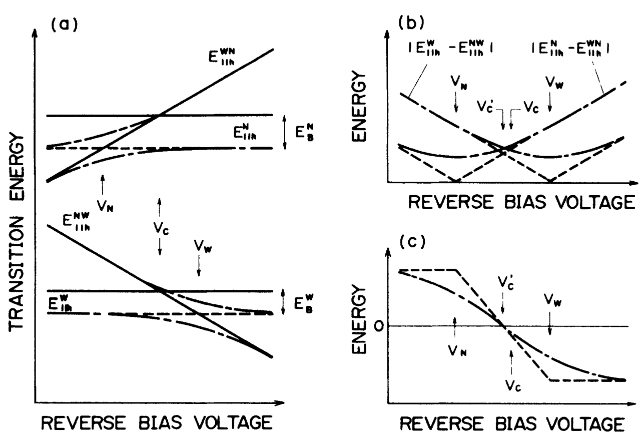

FIG. 4. Graphical explanation of the anticrossing characteristics of the excitonic transitions in double quantum wells. (a) The bias-voltage dependence of E^{W}_{11h} and E^{NW}_{11h}, and E^{N}_{11h} and E^{WN}_{11h}, (b) $|E^{W}_{11h}-E^{NW}_{11h}|$ and $|E^{N}_{11h}-E^{WN}_{11h}|$, and (c) $|E^{W}_{11h}-E^{NW}_{11h}|-|E^{N}_{11h}-E^{WN}_{11h}|$ for the uncoupled system (dashed lines) and the coupled system (dash-dotted lines). In (a), the solid lines denote the band-to-band transition energies for the uncoupled system, whereas the band-to-band transition energies for the coupled system (or the anticrossing characteristics for the bare electronic states) are not shown for simplicity.

and E_B^N. Generally, since $E_B^W < E_B^N$ (Ref. 13), $V_C' < V_C$ or $V_W - V_C < V_C - V_N$.

This simple model accords very well with the experimental results. In the present coupled system, V_W and V_N (Ref. 19) are estimated to be ~ -0.25 and -0.55 V, respectively, while V_C', which may be nearly equal to V_C if $E_B^W \sim E_B^N$, is ~ -0.4 V. As seen in Fig. 4(b), in the voltage ranges far from $V_{W(N)}$, the variations of $|E^{W(N)}_{11h} - E^{NW(WN)}_{11h}|$ are identical for both the coupled and uncoupled systems, and will directly reflect the electric-field change in the intrinsic region if the QCSE and the V_{ex} dependence of $E_B^{W(N)}$ are negligible. Here, the value of $|E^{W}_{11h}-E^{NW}_{11h}|/V_{ex}$ around 1.0 V is $\sim$20 meV/V, which is lower than an expected value of $\sim$30 meV/V estimated from the distance between the centers of the paired GaAs wells (84 Å) and the width of the intrinsic region ($\sim$2800 Å). This may mean that the two transitions are not completely decoupled at $\sim$1.0 V yet.

An equivalent conclusion was also obtained for another DQW structure with 8-Å-thick AlAs barriers, although the minimum repulsion energies of the anticrossings were about half the values for the system with 4-Å-thick AlAs barriers due to the weaker interwell coupling, which gives more definite values of V_W and V_N.

Previously, we have observed the anticrossing characteristics of the radiative transitions for the optical excitation power in the photoluminescence properties of the other asymmetric GaAs/AlAs/GaAs CQW structure.[5] In the results (Fig. 4 in Ref. 5), the excitation powers which give the minimum separation energies appear to be different for the two anticrossings.[20] The property can also be explained by the present model where the exciton binding energies are taken into account. Furthermore, a similar approach may be effective for the investigation of the excitonic effect in the Stark localized SL's, where the serial difference energies between the optical transitions are obtained.

In conculsion, we manifested the excitonic effects on the absorption spectra of asymmetric coupled GaAs quantum wells in an electric field. It was shown that the anticrossing characteristics of the optical transitions in the coupled quantum well systems were appreciably modulated by the excitonic effects. A similar investigation about various coupled systems and sophisticated quantitative calculations will lead to deeper understanding of the tunneling between the excitonic states.

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[18]In a previous work [L. Viña, R. T. Collins, E. E. Mendez, and W. I. Wang, Phys. Rev. Lett. **58**, 832 (1987)], a coupling between different (ground and excited) states of excitons in the same well was demonstrated. In the present case, the excitonic coupling involves the interwell coupling due to tunneling between the excitonic states.
[19]The fact that $V_N < V_W$ can also be confirmed from variation in the oscillator strengths of the paired anticrossing transitions.
[20]This feature becomes more obvious from our recent careful measurement [Y. Tokuda, K. Kanamoto, and N. Tsukada, in *Proceedings of the Thirty-Sixth Spring Meeting of the Japan Society of Applied Physics and the Related Societies, Chiba, Japan, 1989* (The Japan Society of Applied Physics, Tokyo, 1989), No. 3, p. 1034 (in Japanese)].