Triple excitonic mixing associated with recoupling of a Stark-localized state in coupled quantum wells confined by superlattices

Yasunori Tokuda, Kyozo Kanamoto, Yuji Abe, and Noriaki Tsukada
Central Research Laboratory, Mitsubishi Electric Corporation, Amagasaki, Hyogo 661, Japan
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The triple-coupling phenomena of excitonic states were found in the electroabsorption characteristics of GaAs asymmetric coupled quantum wells sandwiched by AlAs-GaAs superlattices. The peculiar optical characteristics can be explained by taking account of the effective recoupling of a Stark-localized state formed in the superlattice confinement region with the coupled states in the quantum wells. The interpretation, furthermore, was supported by a numerical calculation of the eigenenergies of the system in an electric field.

The coupling and decoupling phenomena of the eigenstates in a solid are very interesting subjects. Semiconductor superlattice (SL) and quantum-well (QW) structures are the very systems that are suited to investigate such quantum effects, because their eigenvalues can be artificially designed.[1,2] In particular, investigations of electric-field dependence of the optical properties of such multilayer semiconductor systems have manifested unique features such as Stark ladders formed by a field-induced localization and anticrossing resulting from a strong mixing of the two quantized states.[3–11] Also, the recent works showed that both the peak energies and the oscillator strengths of the optical transitions can be significantly modulated by controlling the interwell coupling strengths and that the characteristics may be useful for creating novel optoelectronic devices.[12–15]

A system in which the state once made localized is coupled again appears to deserve deeper investigations. In this paper, we present an unambiguous evidence of the recoupling of the Stark-localized state which leads to the triple excitonic mixing phenomena observed in electroabsorption characteristics of a multilayer system of coupled GaAs quantum wells confined by AlAs-GaAs superlattices. The interpretation, furthermore, is backed up by a calculation of the eigenenergies of the system under an electric field.

We investigate a GaAs/AlAs/GaAs coupled quantum well confined by two AlAs-GaAs superlattice layers. Figure 1 shows the schematically illustrated potential profiles of the Γ conduction band of the composite system. Under flat-band conditions [Fig. 1(a)], the quantized states in the central double quantum well are hardly mixed because of its asymmetrical configuration, while the minibands are formed in the superlattice confinement regions as the result of the strong mixing effects. By application of an appropriate electric field F [Fig. 1(b)], however, the electron eigenstates in the superlattices are localized to make the ladder formation.[5,6] On the other hand, the strong level mixing may be caused in the central double-quantum-well region under resonance conditions because of the very thin tunnel barrier layer.[7–11] It should be noted that, as the electric field increases, the localized states in the left-hand-side superlattice decrease relative to the quantized levels in the central quantum wells. Therefore, the quantized states in the quantum-well region may well be appreciably affected by some of the Stark-localized states in the superlattice confinement region around fields corresponding to the resonances. In other words, we expect to observe recoupling phenomena of the Stark-localized state.

We calculated the electric-field dependence of the quantized electronic levels in a system of the 100- and 80-Å-thick GaAs quantum wells which are separated by an 8-Å-thick AlAs barrier layer and sandwiched by nine alternating layers of 8-Å-thick AlAs and 25-Å-thick GaAs.[16] Figure 2 shows the calculated results on the con-

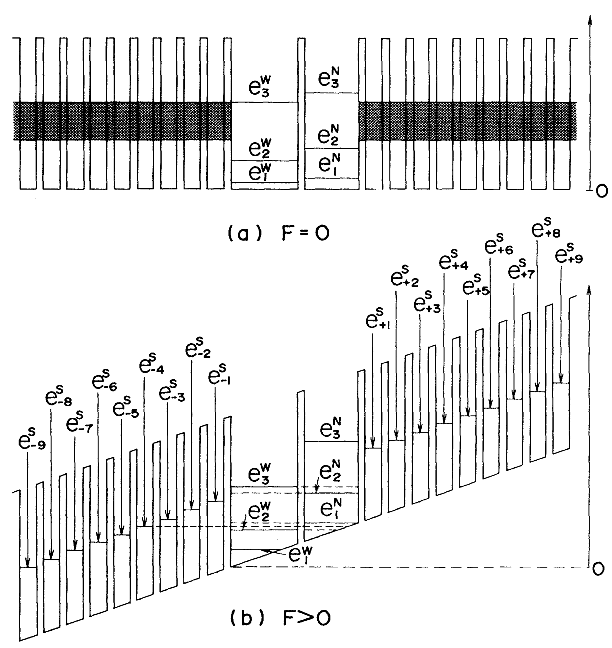

FIG. 1. Schematically illustrated potential profiles of the conduction bands of a GaAs asymmetric coupled quantum well confined by the AlAs-GaAs superlattices in different electric fields F: (a) $F=0$; (b) $F>0$.

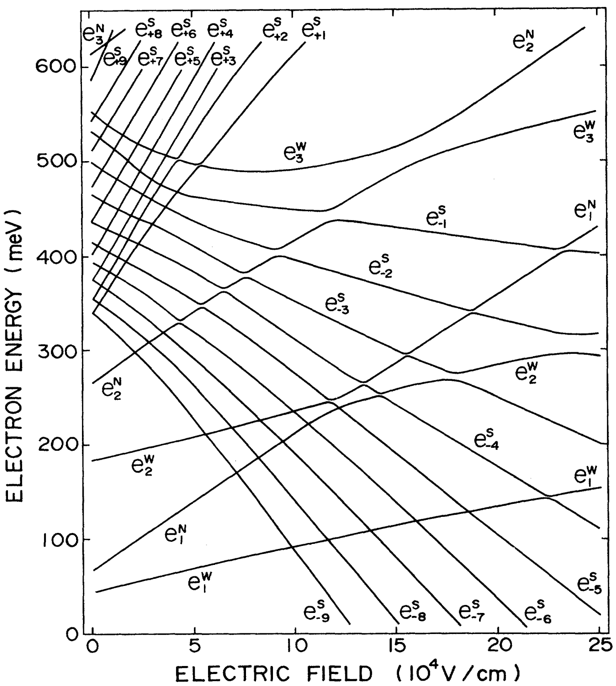

FIG. 2. Calculated electron eigenenergies of the 100- and 80-Å-thick GaAs quantum wells separated by an 8-Å AlAs barrier layer and sandwiched by nine alternating layers of 8-Å-thick AlAs and 25-Å-thick GaAs as a function of the electric field.

duction band, where the electron energies are measured from the minimum point of the bottom in the 100-Å-thick quantum well as indicated in Fig. 1. The symbol $e_n^{W(N)}$ stands for the nth electron state in the wider (narrower) quantum well under decoupling conditions, while $e^S_{-m(+m)}$ refers to the mth Stark-localized state in the left- (right-) hand-side superlattice region.

In Fig. 2, we find clear anticrossing of the two quantized levels in the coupled quantum well, i.e., the e_1^N and e_2^W states around 13×10^4 V/cm as well as the e_2^N and e_3^W states around 16×10^4 V/cm. As predicted, moreover, the quantized levels are intersected with the superlattice localized states, and thus more anticrossing features are seen. The calculated result reveals that the states in the quantum-well region can be modulated by the localized states in the confinement region. It is natural that the repulsive energies at resonance between the $e_n^{W,N}$ and e^S_{-m} levels are larger for a larger number of n (the upper-lying level), for a smaller number of m (the localized level closer to the quantum-well region), and for W rather than N (the quantum well nearer to the left-hand-side superlattice). However, we can recognize, for example, an appreciable repulsion between the e_2^N and e^S_{-6} states in spite of the large spatial separation between the related wells. In addition, the e_3^W level is originally mixed with the states in the superlattice under flat-band conditions and thus the coupled level is pushed up, but the level is decoupled so as to show the original nature with an increase of the electric field. Similar recoupling between the hole states in the central quantum-well region and in the right-hand-side superlattice region can be considered, but the effects are small because of strong localization of the hole states.

The recoupling phenomena of the localized states in the superlattice with the quantized states in the quantum wells may be investigated by the observation of the modulation of the optical transitions in the quantum wells. In the coupled quantum well system, furthermore, we may expect situations in which the Stark-localized state interacts with the mixed states in the quantum wells, and observe more complex coupling behavior than the usual anticrossing of the two quantized states. In Fig. 2, such a situation is seen for a triple mixing between the e_1^N, e_2^W, and e^S_{-4} states around 13.5×10^4 V/cm. Here, $E^{W(N)}_{ijh(l)}$ stands for an intrawell transition between $e_i^{W(N)}$ and $h(l)_j^{W(N)}$ [the jth heavy- (light-) hole level in the wider (narrower) quantum well], while an interwell transition between $e_i^{W(N)}$ and $h(l)_j^{N(W)}$ is referred to as $E^{WN(NW)}_{ijh(l)}$. Furthermore, we indicate the transition between $e^S_{\pm k}$ and $h(l)_j^{W(N)}$ by $E^{SW(N)}_{\pm kj}$.

We carried out a detailed examination of the experimental results of the electroabsorption characteristics of the AlAs/GaAs multilayer structure[17] nearly equivalent to the configuration of Fig. 1. Figure 3(a) shows the absorption peak energies as a function of the reverse bias voltage V_{ex}.[18] This was obtained by measurements of the photocurrent spectra at 77 K, and the fundamental interpretation of the experimental results has been performed in our previous work.[8] The four optical transitions, which are associated with the e_1^W state and dominated almost solely by the quantum-confined Stark effect,[19] are denoted by the small dots along with the identified origins (E^W_{11h},E^W_{11l},E^W_{12h}, and E^W_{13h}). The variations of the remaining absorption peaks, however, are complex and have not been understood completely, though it is sure that the behavior is basically characterized by the anticrossing between the e_1^N and e_2^W states. We classified the remaining observed peaks into three groups indicated by open circles, solid circles, and open triangles. The higher-energy group denoted by the open triangles appears to be composed of four branches. On the other hand, the features of both the two lower-energy groups denoted by the open and solid circles, which consist of three branches, seem to be very different from that of the triangle group. It is evident that the two lower-energy groups denoted by the open and solid circles are involving the same electron states, because their features are just about the same except for the transition energy difference which corresponds to the energy difference between the h_1^N and l_1^N hole levels.

Here, we show that the peculiar electroabsorption characteristics related to the anticrossing of the e_1^N and e_2^W states can interpreted clearly by taking account of further coupling with a Stark-localized state e^S_{-4} in the superlattice. That is, we consider the mixing modes of the three transitions associated with the $h(l)_1^N$ state ($E^N_{11h(l)}$, $E^{WN}_{21h(l)}$, and $E^{SN}_{-41h(l)}$) for the open- (solid-) circle group as well as the three transitions associated with the h_1^W state (E^W_{21h}, E^{NW}_{11h}, and E^{SW}_{-41h}) for the open-triangle group. That the related localized state is e^S_{-4} in the present case

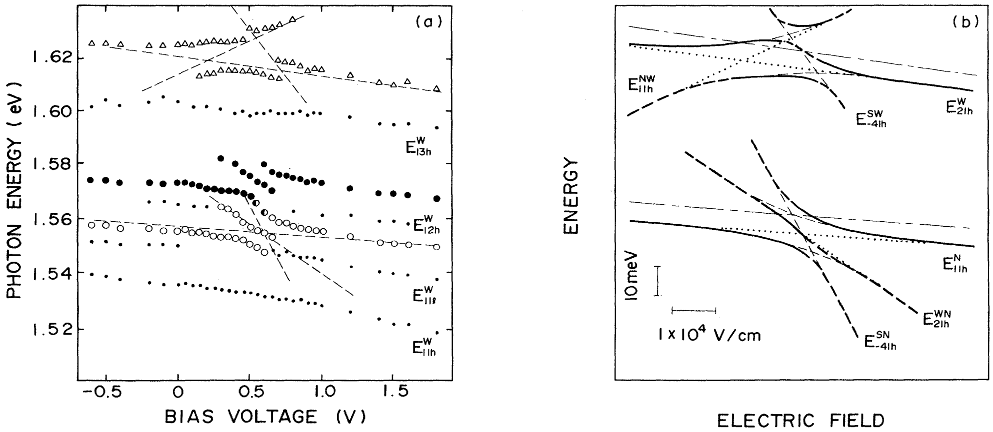

FIG. 3. (a) Absorption peak energies as a function of the external bias voltage V_{ex}. The data were taken from the photocurrent spectra measured at 77 K. (b) A graphical explanation of the experimental results according to the mixing between the three optical transitions. Two dotted lines and a thin dashed straight line in (b) correspond to the three dashed lines in (a). For simplicity, the illustration for the solid circle group is not given.

is based on the calculation of Fig. 2.

In Fig. 3(b), we give a graphical explanation for the present electroabsorption characteristics. We first discuss the result of the higher-energy group denoted by the open triangles. Two dotted and a thin dashed straight lines indicate the electric-field dependence of the E^{W}_{21h}, E^{NW}_{11h}, and E^{SW}_{-41h} transition energies for the uncoupled quantum-well system, respectively. These three straight lines correspond to the dashed lines in Fig. 3(a). Here, the slopes of the lines are based on the calculation of the field dependence of the band-to-band transition energies for the nominal thickness of the intrinsic region of the present sample ($\sim$2850 Å), but the absolute transition energies are determined by comparison with the experimental data. Two thin dashed curves, which are drawn for the two dotted lines and whose large parts cannot be seen because of overlaps with the bold lines, indicate the usual anticrossing characteristics of the E^{W}_{21h} and E^{NW}_{11h} transitions. Here, for the triple coupling of the E^{W}_{21h}, E^{NW}_{11h}, and E^{SW}_{-41h} transitions, we may consider the interaction between these three thin dashed lines, and thus the new three branches denoted by the bold (solid and dashed) lines are obtained. The parts experimentally observed in Fig. 3(a) correspond to the solid bold regions, which nearly agree with the parts where the optical transitions are expected to hold strong oscillator strength.

The characteristics of the two lower-energy groups denoted by the open (solid) circles can also be explained by taking the triple mixing between the $E^{N}_{11h(l)}$, $E^{WN}_{21h(l)}$, and $E^{SN}_{-41h(l)}$ transitions into account. In Fig. 3(b), we only give a brief explanation for the open-circle group, which is also available for the solid-circle group by substituting the light-hole state for the heavy-hole state. In a similar way to the case of the triangle group, we first consider the coupling of the E^{N}_{11h} and E^{WN}_{21h} transitions (two dotted straight lines). Next, by considering the further mixing between the coupled E^{N}_{11h} and E^{WN}_{21h} modes (two thin dashed curves) and the E^{SN}_{-41h} transition (a thin dashed straight line), we obtained three bold lines, solid parts of which also are observed experimentally.

The present results show that the electron states in the quantum-well region are modulated by the electron state which once has been localized at a distance of more than 200 Å and that the modulation becomes remarkable through the triple mixing. Moreover, we find that the observed characteristics of the triple mixing of the absorption transitions, which are more complex than the normal anticrossing of the two transitions, greatly vary with the quantum wells where the holes are localized, because we can observe the only parts where the oscillator strength is large.

In addition, it should be worth noticing that, in Fig. 3(a), the three intersection voltages between the three absorption transitions for the uncoupled system are different between the higher-energy (triangle) and lower-energy (circle) groups. (For the circle groups, the three intersection voltages coincide with one another.) We believe these facts can be attributed to the excitonic effects, as is clarified for the anticrossing behavior of two optical transitions.[10] In Fig. 3(b), we drew thin dash-dotted lines parallel to the E^{W}_{21h} and E^{N}_{11h} dotted lines so as to make the three crossing voltages coincide for the open-triangle and open-circle groups, respectively. These lines indicate the optical transition energies between the bare quantized levels (the band-to-band transitions). The difference energies between the dotted and dash-dotted lines, therefore, may correspond to the exciton binding energies.

In conclusion, we investigated the quantized-state mixing in a system of the asymmetric coupled GaAs quantum wells confined by the AlAs-GaAs superlattices. Theoretical consideration clarified that the field-induced localized states in the superlattice confinement regions can be effectively recoupled with the quantized states in the central well region in spite of the considerably large spatial separation between the wells concerned. We demonstrated that the peculiar electron-field dependence of the absorption properties, which was observed for such an AlAs-GaAs multilayered system, can be explained in terms of the triple excitonic mixing caused by the recoupling with two mixed electron states in the quantum well region.

We have learned of the recent work of Agulló-Rueda *et al.*,[20] whose content is concerned with the *recoupling* of the localized states.

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[16]The eigenvalues of the Schrödinger equation were numerically sought by a finite diffrence method. The following band parameters were used for the calculation: ratio of the conduction- and valence-band discontinuities, 6:4; effective masses of electrons, heavy holes, and light holes in $Al_xGa_{1-x}As$, $(0.067+0.083x)m_0$, $(0.35+0.4x)m_0$, and $(0.08+0.07x)m_0$, respectively. The energy dependence of the effective masses (or the band nonparbolicity) was not taken into account.

[17]The sample used is a *p-i-n* photodiode. The intrinsic region composed of the AlAs-GaAs superlattice, whose nominal width is ~2850 Å, contains four pairs of asymmetric coupled double quantum wells, which consist of 100- and 80-Å-thick GaAs wells and 8-Å-thick AlAs barriers.

[18]$F>0$ even at $V_{ex}=0$ because of the built-in field induced by the *p-n* junction.

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