Observation of Quasibound States in Semiconductor Single Quantum Barriers

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We report the observation of quasibound states in *single* semiconductor *barriers*, in heterostructures with both type-I and type-II band alignments. We discuss the localization and the shape of wave functions of such states, as well as their density of states. In structures with type-II band alignment (CdSe/ZnTe and CdMnSe/ZnTe) we observe transitions involving electrons confined in the conduction band well and quasibound holes localized in the valence band barrier (both in the CdSe layer). In the system with type-I band alignment (ZnMnSe/ZnSe), transitions between states quasibound in the single ZnMnSe barrier also show clear absorption peaks.

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Quasibound states have been studied in a number of semiconductor heterostructures, most extensively in geometries consisting of two separated square potential barriers (so called "double-barrier structures"). The mechanism of the quasilocalization in this case is similar to that of quantum wells, since both systems involve states that are confined between barriers, with the exception that the wave function remains *finite* throughout the structure—a feature representative of all quasibound states. The properties of quasibound states in semiconductor quantum structures have recently been the focus of several other studies, including quantum wells with asymmetric barriers [1], quasibound states in type-I superlattices [2], as well as heterostructures behaving as Bragg reflectors [3]. In such cases quasiconfinement is achieved for states that are outside the well regions, at energies above the potential barriers. In this paper we present the first experimental and theoretical study of the optical properties of quasibound states in semiconductor heterostructures containing *single quantum barriers.*

The single-barrier system is qualitatively different from most of the thoroughly studied semiconductor heterostructures in that its eigenstates are in the form of a continuum, with no truly bound states. However, when the wavelength of an electron λ satisfies $\lambda = NL_b/2$ (where L_b is the barrier width and N is an integer), quasilocalization of the states occurs *in the barrier*, through interference of traveling electron waves. Such conditions are in fact the same as those for resonant tunneling through a barrier (i.e., the wave functions of states responsible for resonant tunneling through single-barrier systems are quasilocalized in the barrier region). These quasibound states will therefore manifest a behavior similar to bound states in quantum wells, such as the dependence of energy on layer width. It is particularly striking—as we will show below—that these similarities to truly bound states are readily observable in the form of absorption peaks in optical experiments on single barriers—despite the fact that in a single barrier there are no discrete levels, or even subbands, but rather a *continuum* of eigenstates.

In this paper we focus on the optical properties associated with quasibound states of semiconductor single-barrier structures, and we show that it is possible to observe optical transitions associated with quasibound states in single barriers with both type-I (ZnSe/ZnMnSe) and type-II (CdSe/ZnTe and $Cd_{1-x}Mn_xSe/ZnTe$) band alignments. We also note that the presence of the magnetic ion, Mn, in these structures is only important for identifying the spatial region within which a given optical transition occurs [2,4,5], as briefly discussed later.

In order to understand why optical transitions involving quasibound states in a single barrier can be observed, it is necessary to compare the density of such states with the rest of the continuum. Since the lack of translational symmetry renders such a calculation intractable for a single-barrier system, we artificially introduce translational symmetry by considering a superlattice consisting of alternating barriers and wells, and calculate the density of states as the density of subbands [6] in the limit of extremely wide "wells" (corresponding to very narrow subbands separated by small gaps, i.e., the quasicontinuous limit). This result is plotted in Fig. 1 (dashed curve) for such a superlattice consisting of barriers that have a width of $L_b = 200$ Å and are separated by wells with a width of 3 μm. The height of the barriers is taken to be 0.64 eV (the same as the valence band offset in CdSe/ZnTe, discussed later), and the effective mass to be the free electron mass.

To demonstrate the effect of quasilocalization, we will use the simple case of an electron incident on a barrier, for which analytic solutions can easily be obtained. This is shown in Fig. 1 (solid curve) by the quantity $(|A|^2 + |B|^2)/|I|^2$, where I is the coefficient of the incoming electron wave outside the barrier region, and A and B are the coefficients of electron waves inside the barrier region traveling in opposite directions. The ratio $(|A|^2$

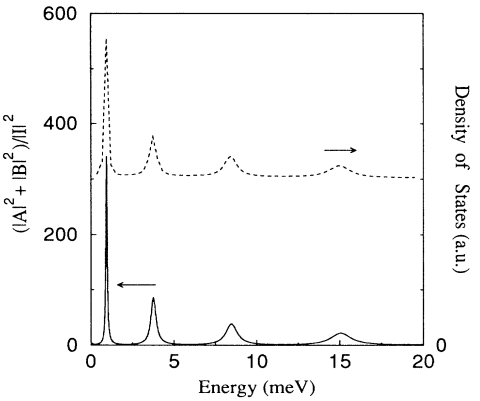

FIG. 1. The ratio of the coefficients $(|A|^2+|B|^2)/|I|^2$ (solid line) provides a measure of the localization of the wave function. The dashed line represents the density of states, which peaks at the same energies as the localization. The energy is measured from the top of the barrier.

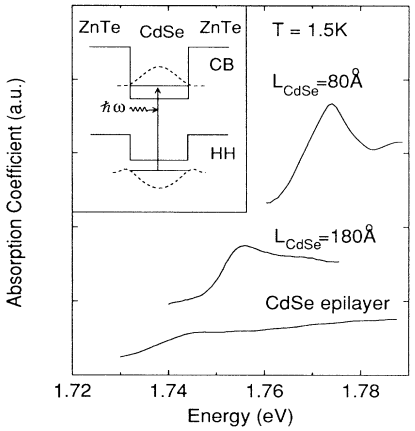

FIG. 2. Absorption spectra of two samples containing single layers of CdSe sandwiched in ZnTe and of a CdSe epilayer are shown on the same scale, indicating the effect of confinement not only in the energies of the transition, but also in the transition probability between the quasibound above-barrier states in the valence band and the bound states in the conduction band (absorption intensity). Inset: The band alignment of the single-barrier heterostructure.

$+|B|^2)/|I|^2$ thus provides a measure of quasilocalization in the single barrier, i.e., a large value of this quantity represents a state with most of its wave function in the barrier region. As is obvious from the figure, the degree of localization is quite large. Furthermore, the state density peaks and the localization in the barrier occur at the same energies—and correspond to wave vectors equal to $k_b = N\pi/L_b$, which is also the condition for resonant tunneling. It is this coincidence of the density-of-states peaks for the quasibound states and the wave function localization that facilitates the observation of such states. It should be emphasized that the energy corresponding to the band edge of the bulk barrier material (i.e., the barrier top, given by $E=0$ in Fig. 1) is now occupied by a delocalized state belonging to the energy continuum, and is not expected to exhibit itself any differently from the rest of the continuum of delocalized states. This is similar to the bottom of a quantum well, which is no longer an eigenstate of the quantum well structure.

We show here that the effects described above can be studied in single-barrier systems with both type-I and type-II band alignments. In the former case, the quasibound states are localized in the barrier region in both the conduction and valence bands. One therefore expects that transitions involving these quasibound states would be characterized by higher intensity than any other transitions between the delocalized states of the continuum. For a type-II structure, the layer that is a barrier in one band (e.g., the valence band) will be a well in the other (in this case the conduction band). In such a system, one again expects to observe the absorption peaks between the quasilocalized states in the valence band barrier and the truly bound states in the conduction band well (as shown in the inset in Fig. 2). Because in our case the heavy-hole-to-conduction-band transitions are much stronger than those originating from the light hole states, we will only focus on transitions involving the heavy hole.

An important feature to note is that quasibound states (i.e., those with their wave vectors in the barrier, k_b, equal to $k_b = N\pi/L_b$) have the *same* wave function parities and shapes as those for states in an infinitely deep quantum well with the corresponding quantum number N. The same is true for the confinement energy if we define it more generally (so that it is suitable for both a quantum well and a barrier) as the energy measured from the bulk band edge of the layer within which the state is localized. Thus the transitions between quasilocalized states in a type-I barrier (or between a quasibound state in a barrier and a "truly" bound state in a well in a type-II structure) follow the same well-known selection rules as those for states in a quantum well, i.e., $\Delta N = 0$.

We first discuss a type-II structure comprised of a single thin layer of CdSe embedded in ZnTe. The band alignment for this system is schematically shown in the inset in Fig. 2. (From the periodic table, one notes that CdSe/ZnTe is a II-VI analog of the well-known III-V system InAs/GaSb, which is strongly type II.) The valence band offset of CdSe/ZnTe is $E^v_{ZnTe} - E^v_{CdSe} = 0.64$ eV [7], which corresponds to a conduction band offset $E^c_{ZnTe} - E^c_{CdSe} = 1.35$ eV. Both optical absorption and photomodulated reflectivity experiments were performed on this system. Several CdSe/ZnTe samples were studied, with CdSe layers ranging from 80 to 180 Å. All samples were grown by molecular beam epitaxy on (100) GaAs [8], with thick (1 to 3 μm) ZnTe buffer layers. The CdSe/ZnTe heterostructure is nearly lattice matched (0.3%), again in close similarity to the case of InAs/

GaSb already mentioned.

Transitions involving the lowest quasibound state (heavy hole) as well as higher quasibound states are observable using modulation techniques. A detailed discussion of all the transitions will be reported elsewhere. Here we will focus on the confinement effect corresponding to the strongest transition, i.e., the transition from the lowest quasibound state in the heavy-hole barrier to the ground state in the conduction band well (both in CdSe layer). Optical transmission results are particularly useful for this discussion. The absorption coefficients of two single CdSe barrier samples (80 and 180 Å) are shown in Fig. 2 on the same scale, with the lines shifted vertically for clarity. Also plotted in Fig. 2 is the absorption coefficient for a CdSe epilayer. The energy gap determined from the CdSe epilayer is 1.745 ± 0.002 eV. Figure 2 clearly shows that a reduction of the CdSe layer thickness leads to a blueshift of the transition. The effective mass calculation, including the Coulomb interaction between the electron and the hole, gives transitions at 1.776 and 1.754 eV for the 80 and the 180 Å barrier, respectively. The effective mass values for the electron and the heavy hole used in the calculation are $0.15m_0$ and $1.0m_0$, respectively [9], where m_0 is the free electron mass. The calculated results are in reasonable agreement with the observed blueshifts. Notice also that there is a significant enhancement of the absorption coefficient as the CdSe layer width decreases, in *exact* analogy to excitonic transitions in type-I quantum wells. Based on this enhancement, we conclude that the observed transition is excitonic, since probabilities of free carrier (or band-to-band) transitions are insensitive to changes in layer width.

Although the observed transitions originate from a continuum, the effects discussed earlier concerning localization and density of states lead to fairly narrow absorption peaks: The full width at half maximum of the absorption peak in the 80 Å CdSe sample is 8 ± 2 meV at $T = 1.5$ K. The confinement-enhanced absorption coefficient α for this sample, shown in Fig. 2, is as large as 8×10^4 cm^{-1}, comparable to transitions between the ground states in type-I quantum wells.

To unambiguously identify the location of the observed excitons, we also studied single layers of CdMnSe in ZnTe, in which the giant spin splitting of optical transitions in the magnetic layers serves as a probe for identifying the actual (spatial) region in which the optical transition takes place [2,5]. Note that because of the large band offsets, the presence of a small concentration of Mn does not change the type-II band alignment. Studies of several single-layer samples of CdMnSe in ZnTe show a very large spin splitting of the observed excitonic transition above the CdMnSe energy gap, reinforcing our argument that observed transitions occur within the thin CdMnSe layers.

It should be emphasized that the observed exciton absorption peak in CdSe layers is primarily the result of the wave function localization of the quasibound hole states, rather than from the electrons bound in the conduction band well "pulling" the holes via Coulomb attraction into the CdSe barrier layer. Excitons involving delocalized holes can of course also form, as has already been demonstrated for type-II systems [10]. However, since in the present case of a single barrier in the valence band there is a *continuum* of delocalized hole states, such Coulomb pulling alone would result in a continuous absorption spectrum rather than in the strong narrow peaks observed in our experiments. In fact, there is a weak background in the observed absorption spectrum, and it is the result of just such transitions. The behavior observed in type-I single-barrier systems discussed immediately below, where there are no bound states to provide the Coulomb attraction of the type just described, further confirms the above interpretation.

To illustrate the generality of the confinement phenomenon in single barriers, we have also carried out experiments on the single-barrier type-I system $Zn_{0.85}Mn_{0.15}Se/ZnSe$ ($Zn_{0.85}Mn_{0.15}Se$ being the barrier in *both* the conduction and the valence band [11]). Several samples were grown, containing single barriers of varying thickness. The spectra shown in Fig. 3 are from two samples. One of them (which we will refer to as sample A) contains a 360 and a 150 Å ZnMnSe barrier, separated by 1 μm of ZnSe. The other sample (sample B, grown under identical conditions as sample A) consists of ten 90 Å ZnMnSe barriers separated by 1000 Å ZnSe layers. The measurements for the two single barriers in sample A were carried out separately, by etching away one of the barriers so as to eliminate ambiguities concerning the origin of a particular transition. Because the single barriers in sample B are relatively thin, resulting in weak transitions, we grew ten barriers to enhance the signal in the absorption spectrum. Details of the intensity and the line-shape behavior as a function of the barrier width will be given elsewhere. Here we will only emphasize that

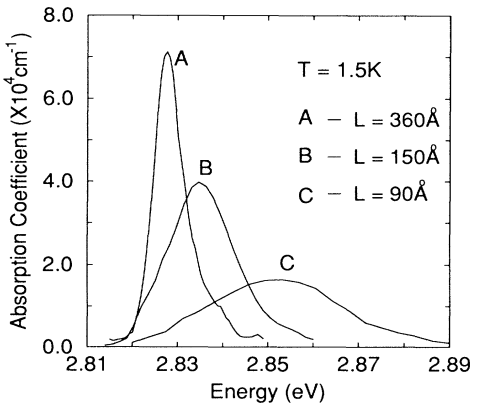

FIG. 3. The absorption coefficients of three single-barrier (ZnMnSe/ZnSe) systems. The clear blueshift of the absorption peak position with decreasing barrier width indicates that the states involved are localized in the barrier.

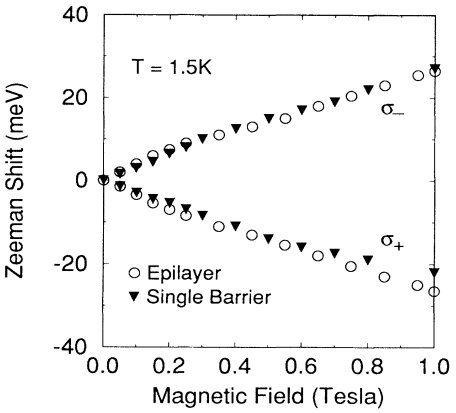

FIG. 4. Experimental results for the Zeeman splittings in the $Zn_{0.85}Mn_{0.15}Se$ epilayer (circles) and for a 150 Å single barrier (triangles) are plotted as a function of magnetic field. The amount of splitting is very close, indicating that the transition in the single-barrier sample occurs in the $Zn_{0.85}Mn_{0.15}Se$ barrier layer.

there occurs a definite blueshift associated with the barrier width (see Fig. 3), and that the width dependence parallels that of a quantum well. Using the effective mass approximation, we calculate the transition energies for the 360, the 150, and the 90 Å barriers to be 2.827, 2.835, and 2.857 eV, respectively. As can be seen in Fig. 3, the results agree with the observed peak positions within experimental error, clearly indicating that the states involved in the transitions are localized in the barriers.

This conclusion is further confirmed by the study of the Zeeman splitting for the transitions observed in magnetotransmission. Magnetotransmission experiments were carried out on all single-barrier samples and on a ZnMnSe epilayer grown under identical conditions as the single barriers (used for determining the concentration of Mn in the single barriers). If the transitions shown in Fig. 3 occur in the barriers (i.e., in ZnMnSe layers), they would show Zeeman splittings comparable to that in the ZnMnSe epilayer. Figure 4 compares the Zeeman splittings for the 150 Å barrier (the same as that used in Fig. 3) and for the ZnMnSe epilayer. As we can see, the splittings for the single barrier and for the epilayer are nearly the same. Furthermore, the remaining barriers corresponding to Fig. 3 also show the same Zeeman splitting, within experimental error. We should add that ZnSe has no observable Zeeman splitting in the field range shown in Fig. 4. Considering the drastic difference between ZnMnSe and ZnSe, the nearly identical Zeeman splittings shown in Fig. 4 provide unambiguous evidence for identifying the spatial region within which states involved in the observed transitions are localized (i.e., the barriers).

In conclusion, we have demonstrated the properties of quasibound states localized in a *single barrier*, and we presented the first optical observation of transitions involving such states. The localization effect is demonstrated by the dependence of the transition energy on the barrier thickness. The spatial localization of the wave function is further experimentally confirmed by the introduction of Mn into the barrier, which results in large Zeeman splittings for transitions involving states quasibound in the barrier. Finally, we would like to point out that such an effect is not limited to barriers in the form of layers (essentially a one-dimensional square potential barrier). It can be easily shown that analogous confinement effects exist in other dimensions, such as barriers in the form of quantum wires and quantum dots, respectively.

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