Stark Localization in GaAs-GaAlAs Superlattices under an Electric Field

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We have observed that a strong electric field $\mathcal{E}$ shifts to higher energies the photoluminescence and photocurrent peaks of a GaAs-$Ga_{0.65}Al_{0.35}As$ superlattice of period D (=65 Å), which we explain by a field-induced localization of carriers to isolated quantum wells. Good agreement is found between observed and calculated shifts when the large field-induced increase of the exciton binding energy is taken into account. At moderate fields [$\simeq(2-3)\times10^4$ V/cm], the coupling between adjacent wells is manifested by four additional peaks that shift at the rates $\pm e\mathcal{E}D$ and $\pm 2e\mathcal{E}D$ and correspond to transitions that involve different levels of the Stark ladder.

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Although the effects of an electric field on a solid have been studied since the early days of quantum mechanics, some related phenomena predicted long ago,[1] such as Bloch oscillations, Stark ladder, and field-induced localization, have not been observed, and even the physical concepts remain controversial.[2] It has been suggested that, were they to exist, their observation would be easier in superlattices[3] because of the reduced widths of their minibands. In this Letter we consider the localization phenomena and present optical experiments that show that under a very strong field the superlattice states are confined to isolated quantum wells, whose states become analogous to the Stark ladder.

The eigenstates of a periodic potential of period D are distributed in bands. If an electron is in a state of a band of width Δ, an electric field $\mathcal{E}$ would induce, in the absence of scattering and interband transitions, an oscillatory motion of frequency $\nu=e\mathcal{E}D/h$, restricted to a spatial region $\Delta/e\mathcal{E}$ (Bloch oscillation). The energy spectrum would be given by $E_0+ne\mathcal{E}D$ ($n=0,1,2,\ldots$), where E_0 is the eigenenergy of an isolated quantum well (Stark ladder). In a three-dimensional solid, in which D and Δ are of the order of angstroms and electronvolts, respectively, the electron would be confined, for any realistic field ($\mathcal{E}<10^6$ V/cm), to a region that would include many atomic sites. On the other hand, in a superlattice, with a period of $\simeq100$ Å and Δ in the range 0.01–0.1 eV, the electronic motion could be restricted to distances of the order of D for moderately high electric fields. Thus, the field would induce a transition from one regime in which the electron is itinerant through many periods to another in which it is confined to a single period.[4]

We have studied the field-induced localization in superlattices defined by the potential profiles of the conduction- and valence-band edges of very thin GaAs-GaAlAs multilayers. The experiments involve optical transitions between the conduction- and valence-band minibands, as illustrated in Fig. 1(a), where, for simplicity, we have assumed that only one miniband is allowed in each superlattice potential. (The valence-band profile accomodates one heavy-hole and one light-hole miniband.)

The interband transitions at $\mathcal{E}=0$, T_1 and T_2, will have energies corresponding to the differences between the bottom of the conduction miniband and the top of the valence minibands, and will be delocalized since the wave functions extend throughout the superlattice. At very high fields, when $\mathcal{E}>\Delta/eD$, these wave functions will be localized in the GaAs quantum well, and the interband transitions will be restricted to these regions [see

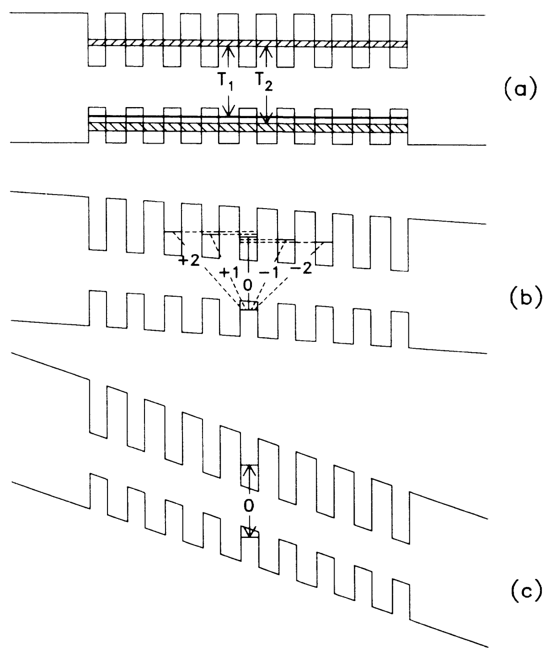

FIG. 1. Sketches of the conduction- and valence-band potential profiles for GaAs-$Ga_{1-x}Al_xAs$ superlattice under a (a) small, (b) moderate, and (c) high electric field, clad by thick $Ga_{1-x}Al_xAs$ regions. The diagrams are approximately scaled for a 30–35-Å superlattice with $x=0.35$, and fields of 2×10^3, 2×10^4, and 1×10^5 V/cm, respectively.

Fig. 1(c)]. In addition, the transition energies will be larger by an amount of the order of $(\Delta_e+\Delta_h)/2$, where Δ_e and Δ_h are the widths of the electron and hole minibands, respectively, since the level for an isolated quantum well lies, to first approximation, at the center of the superlattice miniband.[5]

At intermediate fields a certain degree of delocalization may exist, with the electronic wave functions extending over a few periods [Fig. 1(b)]. Then, in addition to "vertical" transitions, it would be possible to observe transitions involving electron and hole wave functions peaked in different quantum wells or, similarly, involving different levels of a Stark ladder. They would be characterized by shifts to lower energies relative to the "vertical" transition, of the order of $\pm e\mathcal{E}D$, $\pm 2e\mathcal{E}D$, etc.

This simple picture is complicated by the different bandwidths of the electron and hole minibands, and by the Coulomb interaction between electrons and holes (exciton). The former leads to different critical fields for the localization of the two kinds of carriers, and a hybrid situation, in which the heavy holes are localized but the electrons are not, has to be considered. The Coulomb interaction reduces the energy of an interband transition by the exciton binding energy, E_{exc}, which itself depends very strongly on the electric field. At $\mathcal{E}=0$, when the electron and hole are delocalized, the exciton binding energy should be similar to that of bulk GaAs, $\simeq 4$ meV, as recent experiments have confirmed.[6] On the other hand, at very high fields the localization of the exciton would increase its binding up to $\simeq 15$ meV, for an isolated 30-Å quantum well.[7]

The experiments reported here were done on an undoped superlattice made by alternation of 60 30-Å GaAs layers with 59 35-Å $Ga_{0.65}Al_{0.35}As$ films, clad on each end by 600 Å of undoped $Ga_{0.65}Al_{0.35}As$. This heterostructure was in the space-charge region of a p^+-n^+ GaAs junction, doped to 2×10^{18} cm^{-3} carriers on each side. The electric field applied to the superlattice was almost proportional to the voltage drop V between the n^+ and p^+ regions, that is, $\mathcal{E}\simeq e(V-V_{b.i.})/W$, where $V_{b.i.}$ is the built-in voltage (estimated to be 1.62 V in our structure) and W is the total width of the undoped region (5065 Å).

Photoluminescence (PL) and photocurrent (PC) measurements were done at $\simeq 5$ K, with an LD700 dye pumped by a Kr^+ laser to vary the excitation energy in the range 1.60-1.76 eV, with a power density of $\simeq 0.2$ W/cm^2. Photoluminescence excitation (PLE) spectra were also recorded, with the detection wavelength fixed and the excitation energy varied.

The PL signal at $\mathcal{E}=0$ consists of a single structure peaked at 1.697 eV (Fig. 2, 1.6 V), which corresponds to radiative recombination of an electron at the bottom of the conduction miniband with a heavy hole at the top of the valence miniband.

The effects of the electric field are complex, and therefore to focus the discussion we will consider, at the risk of oversimplification, three different regimes: low (1.6 to 1.1 V, $0<\mathcal{E}\leq 1\times 10^4$ V/cm), intermediate (1 to -1 V, $1.2\times 10^4\leq\mathcal{E}\leq 5.2\times 10^4$), and high ($-1.2$ V and below, $5.6\times 10^4\leq\mathcal{E}$).

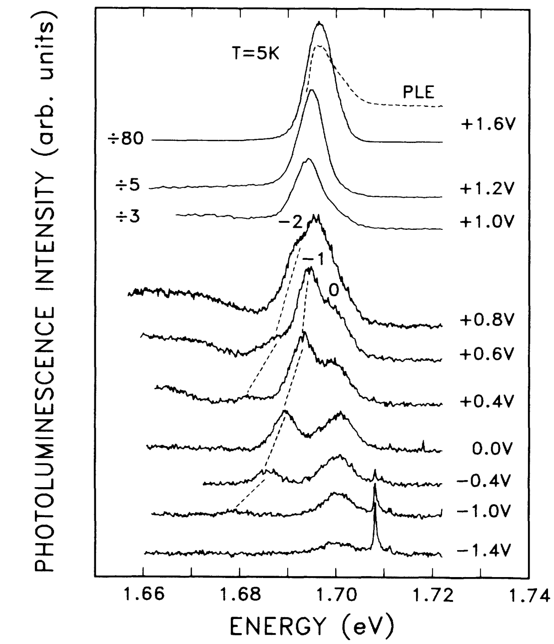

FIG. 2. Photoluminescence (PL) spectra of a 30-35-Å GaAs-$Ga_{0.65}Al_{0.35}As$ superlattice for various voltages applied between the electrodes of a p^+-n^+ junction, on whose space-charge region is the superlattice. The electric field, $\mathcal{E}$ (in kilovolts per centimeter), is related to the external voltage, V, by $\mathcal{E}\simeq 20|V-1.6\text{ V}|$. The peaks labeled 0, -1, and -2 correspond to interband transitions between adjacent wells, as indicated in Fig. 1(b). The photoluminescence excitation spectrum at 1.6 V is shown with a dashed line. The sharp structures observed at $V\leq -0.4$ V are due to field-enhanced resonant Raman scattering.

At low fields, the PL intensity decreases very fast with increasing $\mathcal{E}$, its peak position drops 2 meV, and the height of the PLE exciton peak diminishes relative to the plateau at higher energies. The excitonic nature of the transition is demonstrated by the quenching of the PLE peak by a field such that $e\mathcal{E}a_0^*\simeq E_{exc}$ ($\mathcal{E}\simeq 4\times 10^3$ V/cm), where a_0^* is the exciton effective Bohr radius. The initial 2-meV PL shift, which occurs for both directions of the electric field but is not present in the PLE spectra, could be due to a change of the character of the exciton, from free to bound, but its origin is not well established.

At intermediate fields, the integrated PL intensity decreases much more slowly with $\mathcal{E}$ than at low fields, and the spectra become more complex, as seen in Fig. 2. At ~ 0.8 V, shoulders appear on each side of the 1.695-eV peak, that at lower voltages become distinct structures as they move away from each other. These peaks, labeled

(−2), (−1), and (0), behave very differently with field. Peak (−2), always very weak, shifts downwards linearly with increasing field, at a rate of −1.35 meV/(kV/cm), while peak (−1) shifts at −0.55 meV/kV/cm). The latter dominates between 0.8 and 0.2 V, but below 0 V it is overcome by peak (0), that shifts to higher energies with increasing $\mathscr{E}$, and then it saturates at 1.700 eV at very high fields.

In the intermediate regime the heavy-hole states are localized but the electrons are only partially confined, as sketched in Fig. 1(b). Thus, at $V \simeq 0.4$ V the radiative recombination involves states that, although peaked in individual wells, extend over a few periods, and therefore transition probabilities for electron and hole states that peak in different wells are nonzero. This regime resembles the case of a few coupled quantum wells, like the double-well configurations studied before.[8,9] Peak (0) corresponds then to transitions between electrons and holes in the same well, and peaks (−1) and (−2) to transitions involving electron wave functions whose maxima are one and two periods away from the hole states, respectively. The observed downward shifts of (−1) and (−2) agree with the expected shifts for weakly coupled wells, $-e\mathscr{E}D$ and $-2e\mathscr{E}D$, respectively [that is, −0.65 and −1.30 meV/(kV/cm), respectively, for a 65-Å period]. As the electric field increases, the electronic confinement becomes more important, and the coupling is reduced even further, which decreases the amplitudes of peaks (−2) and (−1), and at very high fields they disappear, once the electron wave functions are fully localized in isolated wells.

Transitions denoted by (+1) and (+2) on Fig. 1(b) are in principle also possible, although they were not observed in PL experiments at low temperature, probably because of thermalization processes. However, they are clearly resolved in the PC spectra of Fig. 3, where up to five transitions (0, ±1, ±2) involving localized heavy-hole states can be simultaneously observed for certain fields. The structures with a positive index shift to higher energies with increasing $\mathscr{E}$ while those with a negative index move toward lower energy, as summarized in Fig. 4. The slopes of the ±1 branches are ±(0.63 ±0.07) meV/(kV/cm), and those of the ±2 series ±(1.30±0.25) meV/(kV/cm), values which again agree with the predicted shifts. The departure from linear behavior of the (−1) transition at low fields, as well as the initial downward shift of the (0) peak, is a consequence of the strong coupling between states at low fields.

The structures in Fig. 3 labeled (0*l*) and (−1*l*) correspond to interband transitions associated with light holes. As seen in Fig. 4 (open circles) their energies as functions of $\mathscr{E}$ run parallel to heavy-hole transitions with the same indexes. Since the states associated with the light-hole Stark ladder have a degree of delocalization comparable to, or even larger than, that of electrons [contrast, e.g, the bandwidths of the corresponding minibands in Fig. 1(a)], the structure labeled for simplicity (−1*l*) in Fig. 3 actually corresponds to two different interband transitions with the same energy: between the (0) light-

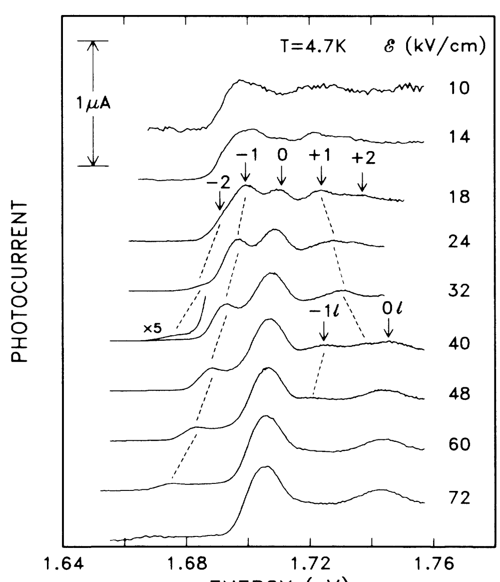

FIG. 3. Photocurrent (PC) spectra for the same superlattice of Fig. 2, at representative electric fields. The peaks labeled 0, ±1, and ±2 are for transitions involving heavy-hole states and electrons weakly delocalized, as illustrated in Fig. 1(b). Analogous transitions for light holes are denotes by 0*l* and −1*l*.

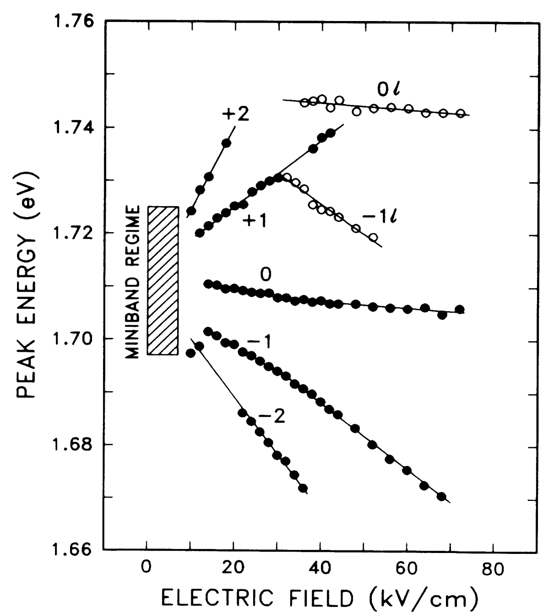

FIG. 4. Transition energies for the PC structures of Fig. 1(a) vs electric field. The filled circles correspond to heavy-hole transitions, whereas the open circles refer to light holes.

hole and the (-1) electron states, and between the $(+1)$ light-hole and (0) electron states. Structures of higher index were not resolved.

The interband energy shift induced by the localization process at high electric field can be deduced, e.g., from the energies of the PL peaks at $\mathscr{E}=0$ and $\mathscr{E}=6\times10^4$ V/cm, after correction for the exciton energy. For this we have used 4 meV (Ref. 6) and 15 meV (Ref. 7), for the two electric-field limits, respectively, which are similar to preliminary estimates derived from magneto-optical experiments performed in the present superlattice. (Although the 15-meV value of Ref. 7 was determined in the absence of any electric field, the field-induced decrease of the exciton energy in an isolated well is smaller[10] than the current uncertainty of the value itself, $\simeq 2$ meV.) The 14-meV interband energy shift thus obtained compares well with a calculated shift of 15 meV of the transition energies in the superlattice and quantum-well limits.[11] The estimated sum of the electron and heavy-hole band widths ($\Delta_e+\Delta_h \simeq 28$ meV) is consistent with a localization field of $\simeq 6\times10^4$ V/cm.

Finally, let us mention the presence of sharp peaks at high fields in the PL and PLE spectra. These structures (see Fig. 2) correspond to electric-field-enhanced resonant Raman scattering of longitudinal and transverse GaAs phonons. The effect observed here is much larger than any field enhancement reported before.[12] A detailed account of this phenomenon is left for a future publication.

In summary, we have shown that a strong electric field induces a carrier localization in a superlattice, which in turn produces a "blue" shift of its interband optical transitions and a significant increase of the exciton binding energy. At moderate fields, wave-function delocalization throughout several periods gives rise to additional transitions that reflect the presence of a Stark ladder. The simultaneous observation of transitions that involve heavy-hole states of a quantum well and electron states up to the second-nearest well (± 2) shows that the coherence length of the superlattice wave functions is at least five periods, and it may be even larger at very low fields (miniband regime).

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Note added.— A recent paper by Bleuse, Bastard, and Voisin,[13] that appeared after this Letter was submitted for publication, has reported tight-binding calculations of electric-field-induced localization phenomena in superlattices. The calculated absorption spectra show field-induced features in good agreement with the PC spectra of Fig. 3.

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