Indirect-Direct Anticrossing in GaAs-AlAs Superlattices Induced by an Electric Field: Evidence of Γ-X Mixing

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We demonstrate that a GaAs-AlAs superlattice can be switched from indirect to direct, in both real and reciprocal spaces, by application of a modest axial electric field. The crossover region is found to be an anticrossing, manifesting the presence of Γ-X mixing by a potential measured to be of the order of 1 meV. This value is corroborated by time-decay measurements performed on different superlattices.

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Since the conception of superlattices,[1] their band structure and properties have been reasonably understood as consequences of the periodic modulation of crystal properties along the growth axis. Electrons and holes are partially confined in the layers of their lowest energy, and their energies increased by quantum effects of this confinement. The layering of the charge distributions has interesting consequences, in that the crystal properties differ significantly depending on whether electrons and holes are confined in the same layer (type-I superlattices) or in adjacent layers (type-II superlattices).[2] Other features of interest can develop when individual layer thicknesses are reduced and the confinement energies increase. In particular, the superlattice conduction-band minimum can be driven higher in energy than another minimum in the Brillouin zone, resulting in a material direct in reciprocal space for a certain range of thicknesses and indirect for other thicknesses.[3-5]

We report here dramatic electric field effects in type-II GaAs-AlAs superlattices designed with layer thicknesses such that they are just barely indirect in band structure, i.e., with electrons lying at an X minimum of AlAs. We show here for the first time that these electrons can be transferred into the GaAs Γ point by making use of energy shifts of the quantum states under application of an electric field. The material is therefore electric-field switched from indirect and type II to direct and type I, an extremely fast process which had not previously been realized.

In GaAs-AlAs short-period superlattices, for GaAs layers of thickness $\lesssim 35$ Å (and thicker AlAs layers), the Γ conduction-band minimum, whose wave function is localized predominantly in GaAs, lies at an energy near that of the bulk AlAs X-valley minimum. Thus for thin GaAs layers (i.e., $d_1 < 35$ Å), the lowest conduction-band state is that of the heavier X-AlAs electrons confined in the AlAs layers. Since the valence-band extremum remains at Γ in all cases, the short-period superlattice is then indirect and in both real (type-II) and k space, with electrons and holes spatially separated and at different points of the Brillouin zone (Fig. 1).[3-9] From simple perturbation-theory arguments one can expect, however, that for small energy separations between the X and Γ minima, mixing effects might substantially affect the electronic wave functions.[8,9]

In a type-II superlattice, as a result of the spatial separation of electrons and holes, the indirect band-gap energy is shifted towards higher energy under application of an electric field perpendicular to the layers.[10] The quantum confined Stark effect on the spatially direct Γ band gap remains a red shift.[11-13] It is these opposing blue and red shifts under application of an electric field which we utilize in the present work to switch a superlattice from type II indirect to type I direct. In the past the direct-indirect conversion has been possible only by means of alloying, which is irreversible, or by application of pressure,[14] thus requiring heavy laboratory equip-

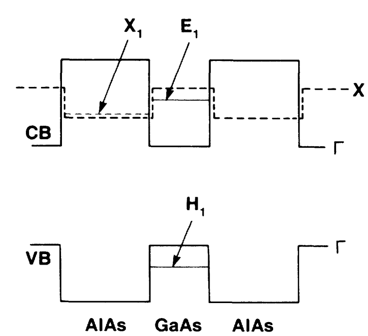

FIG. 1. X (dashed line) and Γ (full line) conduction- and valence-band profiles in a type-II GaAs-AlAs superlattice. X_1, E_1, and H_1 refer to the lowest quantum states of the X electron, Γ electron, and Γ heavy hole, respectively.

ment. We show here that not only this switching, but also the type-I to type-II switching, is attainable with very modest axial electric fields, and should take place in picosecond time scales. We also analyze the blue Stark shift of the indirect transition in the low electric field regime to extract the quantum confined Stark shift of the X electron; this provides a measurement of its mass along ⟨001⟩ (the growth axis, z), which is found to be $\approx 1.2m_0$, consistent with this state being the ⟨001⟩, X_2 orbital. At higher fields, the direct and indirect band gaps are found to anticross, manifesting the existence of a mixing potential $V_{\rm mix}$ which is measured to be about 1 meV. Consistent with this, we obtained other measurements of this potential on a set of samples with different Γ-X configurations by means of photoluminescence time-decay studies. They further show that the mixing scales as the inverse square of the energy separation between Γ and X, in agreement with perturbation theory. This mixing potential could be either a random potential due to alloy fluctuations at the superlattice interfaces or the ⟨001⟩ component of an atomic potential with the superlattice periodicity.

All superlattices investigated were grown by molecular-beam epitaxy. The sample used for Stark-effect measurements is a p-i-n structure, where the superlattice, designed to be indirect by only a small energy, is made of 43 periods of 35-Å GaAs and 80-Å AlAs layers, undoped, and is embedded between 0.5-μm-thick layers of GaAs doped in the range of 10^{18} cm^{-3} with Si (bottom) and Be (top). The substrate is n^+-GaAs. Ring-shaped metal contacts were evaporated on the top p side, and the voltage was applied between these and the back of the sample. The other samples, on which photoluminescence time decay was measured, are undoped, 0.5-μm-thick superlattices, with periods ranging from 14 to 31 monolayers.

Photoluminescence spectra of the p-i-n, 35/80 sample, taken for various applied voltages, are shown in Fig. 2. At very low fields, they consist of two weak lines at 1.735 and 1.720 V, labeled Γ and X, respectively. Both lines have similar excitation spectra, showing two peaks at 1.758 and 1.807 eV, corresponding to E_1H_1 and E_1L_1 direction excitons made of the fundamental Γ electron with heavy and light holes, respectively. The lower energy part of the excitation spectrum (E_1H_1 exciton) is enhanced for the 1.735-eV Γ line, indicating a better feeding for the corresponding electronic state. The Γ-line energy is within the tail of the E_1H_1 free-exciton absorption line, which is about 40 meV wide. This corresponds to ≈ 1 monolayer thickness fluctuation of the GaAs layer. We therefore attribute this Γ line to the recombination of the direct E_1H_1 exciton on the lowest energy sites.

As voltage is applied, from 2.25 to 3.45 V, the X-line intensity increases drastically and moves to higher energies. This intensity change is expected for type-II transitions, since for larger fields the spatially separated electron and hole wave functions are brought closer to an interface where their overlap is larger. The blue shift of the X line results from the existence of a voltage drop between adjacent layers. This, together with the increase in intensity, shows unambiguously that it is a type-II transition between separated electron-hole pairs. This is consistent with previous work in which this line was associated with an indirect exciton consisting of an electron in the AlAs X minimum and a hole in the GaAs Γ minimum.[5]

Both Γ and X minima are significantly populated at fields ranging from 4 to 5 V. Above 4.5 V, Γ becomes the lowest-lying line. At 5 V the X line has been driven to an energy of 1.749 eV, a shift of ≈ 30 meV from its original position, after which the X minimum is no longer populated and the transition cannot be followed. Note, however, that larger Stark shifts of the X transition energy should be observable in a more indirect sample, i.e., one with a Γ minimum higher in energy.

The blue shift of the X line is given by

$$\delta E = eFd/2 - \delta X - \delta H, \tag{1}$$

where the first term is positive and corresponds to a volt-

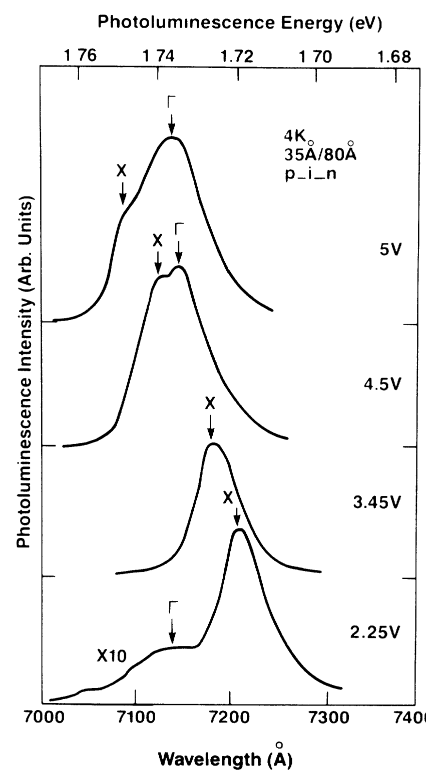

FIG. 2. Photoluminescence spectra of a 35-Å GaAs, 80-Å AlAs superlattice embedded in a p-i-n structure, taken at various applied voltages. Excitation density is about 10^2 W cm^{-2}. X and Γ are the indirect and direct optical transitions, respectively.

age drop across a half period, and the two other are the negative quantum confined Stark shifts of the X-electron and heavy-hole states, respectively. The Stark shift on the binding energy of the exciton is a second-order effect, expected to be of the order of a few millielectronvolts only, and will be neglected. δX and δH are given in the low-field regime by perturbation theory.[11] δX can be calculated as

$$\delta X=\sum_{n\neq 1}\left(\frac{\langle\psi_{x1}|eFz|\psi_{xn}\rangle}{E_{x1}-E_{xn}}\right)^2, \qquad (2)$$

where ψ_{xn} and E_{xn} are the wave function and energy, respectively, of the nth X quantum confined state, and F is the field. In the approximation of an infinite quantum well, which is reasonable here for these thicknesses and masses, Eq. (2) has a simple form and δX scales as $m^*(X)F^2d_2^4$, where d_2 is the thickness of the AlAs layer and $m^*(X)$ the effective mass of the X valley along the growth axis. δH is similarly calculated, and scales as $m^*(H)F^2d_1^4$. Since $d_2/d_1=2.3$ and $m^*(X)>m^*(H)$, we can neglect the hole Stark shift. The quantity $eFd/2-\delta E$ should therefore, in the low-field regime, be linear in F^2, with a slope proportional to the mass of the X electron along the z axis. This has been plotted in Fig. 3; the agreement is excellent up to $F\approx 3\times 10^4$ V cm^{-1}, and provides a measurement of the X-electron mass which is found to be $1.2m_0$. This large mass justifies neglect of the hole contribution ($\delta H\approx 0.01{*}\delta X$) and demonstrates that for this AlAs thickness the X orbital lying along the growth axis, X_2, is the lowest conduction-band minimum. A different result was found formerly on samples with smaller periods[5] where it was concluded that the in-plane orbitals $X_{x,y}$ had the lowest energy. Our findings suggest that the ordering of the X_z and $X_{x,y}$ minima could be thickness dependent, as was also proposed in recent pseudopotential calculations.[8]

At higher fields, the red Stark shift δX of the X state saturates, and the overall shift approximates closely $eFd/2$ towards higher energies. Crossing of the Γ and X transition energies occurs at about 4.5×10^4 V cm^{-1}, as can be seen in Fig. 4. However, the transitions are found to anticross, with a splitting at crossover of about 2.5 meV, manifesting the existence of mixing. The potential responsible for the mixing of the ⟨000⟩ and ⟨001⟩ Bloch wave functions, which is twice smaller, is therefore of the order of 1 meV.

The existence of mixing between Γ and X in indirect type-II superlattices can also be probed by the measurement of the time decay of the indirect luminescence. The radiative recombination rate of this transition in the absence of phonons ought to be proportional to the square of the Γ component of the X-electron wave function. Such experiments were performed on a series of superlattices with periods ranging from 38 to 88 Å, corresponding to Γ-X spacings of 100 to 200 meV. The X-exciton and E_1H_1 energies were measured by photoluminescence and excitation spectroscopy, and the radiative recombination rate of the no-phonon X luminescence was obtained from an exponential fit to the luminescence decay. Decay times ranged from 5 μs for the 200-meV Γ-X spacing (78-Å period) sample to 220 ns for the 100 Γ-X spacing (38-Å period) sample. The resulting potential V responsible for the mixing is then given by perturbation theory as

$$\frac{\omega(X)}{\omega(\Gamma)}=\left(\frac{V_{\text{mix}}}{E(X)-E(\Gamma)}\right)^2, \qquad (3)$$

where $\omega(X)$ and $\omega(\Gamma)$ are the radiative recombination rates of the X and Γ transitions, respectively, and V_{mix} is the potential responsible for the mixing. Assuming that

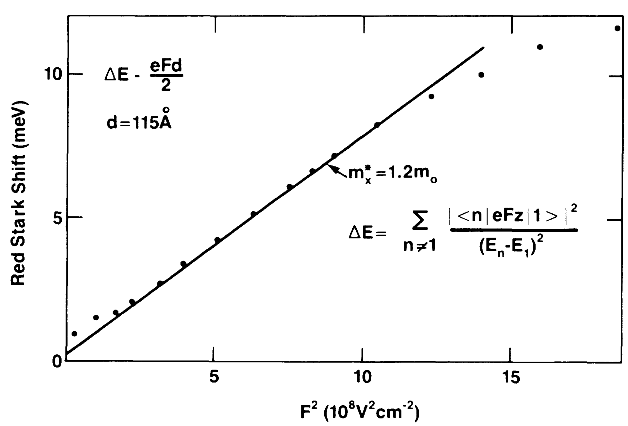

FIG. 3. Red Stark shift of the X minimum at low fields as extracted from the overall blue shift ($\delta E-eFd/2$) plotted as a function of the square of the field. The linear fit is obtained from perturbation theory for an X electron mass of $1.2m_0$ along ⟨001⟩.

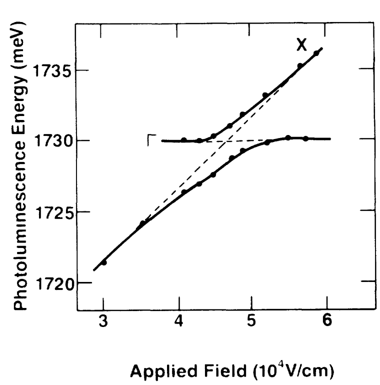

FIG. 4. Energies of the direct and indirect transitions at high fields showing the anticrossing between the X and Γ transitions. The linear blue shift of the X transition far from the anticrossing follows the drop of voltage across a half period. The red quantum confined Stark shift is saturated in this field regime.

the direct recombination rate is constant and equal to 5×10^9 s^{-1} for all samples,[15] we obtain potentials V_{mix} ranging from 1 to 3 meV on all 8 samples investigated. However, the product $V_{mix}n$, where n is the number of monolayers in a period, is found to be constant within 10% over this range of thicknesses and equal to about 40 meV. The corresponding potential $V_0 = V_{mix}n$ deduced from the anticrossing for the *p-i-n* sample is 50 meV. The agreement is therefore very good, and seems to indicate that indeed the potential scales as $V_{mix} = V_0/n$, where V_0 is of the order of 50 meV.

We suggest two mechanisms which could be responsible for this mixing: (i) the scattering between Γ and X induced by a random potential due to fluctuations at the superlattice interfaces,[5] or (ii) the mixing of the wave functions due to the superperiodicity itself. In the latter case, V_0 corresponds to the ⟨001⟩ component of the atomic potentials of Ga and Al modulated with the superlattice periodicity. Experiments on samples with graded aluminum concentration profiles might allow determination of the correct mechanism.

In summary, we have succeeded in switching a GaAs-AlAs superlattice from a type-II, indirect material to a type-I, direct one by means of an applied electric field. The crossover is found to be an anticrossing induced by a potential of ≈1 meV responsible for Γ-X mixing. The switching should therefore take place on a picosecond time scale, and is not limited by transfer or lifetime of the carriers. This opens possibilities for new optics and transport phenomena as well as device applications, as the direct and indirect configurations have very different properties. Finally, the mixing of the wave functions was corroborated by time-resolved measurements evidencing the existence of a Γ-X mixing potential, which is found to scale with the inverse of the superlattice period. While the exact nature of this potential is not entirely known yet, its strength and effects are clearly observable both in the anticrossing and in the reduction of the indirect radiative lifetime in superlattices with decreasing Γ-X energy separations. The electric field tunability in the crossover region will allow further examination of the effect of that mixing on other electronic properties. We have demonstrated here, for the first time, the possibility of monitoring externally and simply the character of the Bloch wave function at the conduction-band minimum. This superlattice effect is of an entirely new type, unattainable either in alloys or in conventional type-I superlattices.

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