Large intersubband infrared transitions in GaAs-$Ga_{1-x}Al_xAs$ superlattices

L. D. L. Brown and M. Jaros
Department of Physics, The University, Newcastle upon Tyne NE1 7RU, United Kingdom

D. C. Herbert
The Royal Signals and Radar Establishment, Malvern, Worcester WR14 3Ps, United Kingdom
(Received 30 November 1988)

We performed pseudopotential calculations of intersubband optical transitions in GaAs-$Ga_{1-x}Al_xAs$ superlattices throughout the full width of the superlattice Brillouin zone. We show that such transitions are strong and tunable over the spectral range 2–30 μm. This is achieved by exploiting the effect of momentum mixing involving bulk contributions from the primary and secondary conduction-band minima.

The purpose of this study is to investigate how the superlattice parameters may be adjusted to vary the underlying superlattice band structure and consequently tune the magnitude of the transition probabilities between the superlattice states. We focus upon transitions in the infrared between the lowest conduction states of GaAs-$Ga_{1-x}Al_xAs$(001) superlattices. We show how these transitions are tuned by mixing of bulk Bloch states derived from the edge of the bulk Brillouin zone into the superlattice wave function.

Earlier investigations[1] concentrated upon the infrared transition between the two lowest confined conduction states in thick isolated GaAs quantum wells. This transition was shown to be strong. In these thick wells the wave functions of the lowest confined conduction states are standing waves obtained from the bulk wave functions associated with the bottom of the conduction band of bulk GaAs. Consequently, the energy levels in such systems may be understood in terms of the particle in a box model in which the bulk wave vector remains a good quantum number.[2] This model also provides an adequate description of the optical transitions between the lowest superlattice conduction states.

However, it is well known that in GaAs and in alloys of GaAs and AlAs the conduction band exhibits secondary minima located at the Brillouin-zone boundary (e.g., the X point). These minima are separated by only a few tenths of an electron volt from the principal Γ valley at the center of the Brillouin zone. When the well and barrier widths are reduced, and the quantum states derived from the bulk states of Γ and X character become near degenerate in energy, the simple picture collapses. The crystal potential of the superlattice couples the bulk states of Γ and X character and the resulting superlattice wave function can be expressed as a linear combination of such contributions. The implications of this breakdown of the particle in a box model concerning the magnitude of the optical transition probability across the fundamental gap have been described in some detail.[3–7] Such transitions have recently been observed in luminescence experiments.[6,8,9] In the present study we focus on intersubband transitions in the conduction band. We show that the magnitude and spectral range of these transitions are a sensitive function of the mixing of bulk Γ and X momentum components. Such transitions are being investigated with a view to identifying applications for detection and generation of infrared radiation.[10,11] In particular, we will show that strong transitions can also be found at points lying further from Γ towards the edge (Z point) of the superlattice Brillouin zone. The strength and frequency of such transitions can be tuned by the effect of momentum mixing.

We use a full scale pseudopotential scheme to model the GaAs-$Ga_{1-x}Al_xAs$ superlattice. The superlattice wave function Ψ for a particular superlattice state is constructed as a linear combination of bulk GaAs wave functions $\varphi_{n,k}$, i.e., $\Psi=\sum_{n,k} A_{n,k}\varphi_{n,k}$: n is the band index and k is the reduced wave vector in units of $2\pi/A$ (A is the bulk GaAs lattice constant). $\varphi_{n,k}$ satisfy the Schrödinger equation $H_0\varphi_{n,k}=E_{n,k}\varphi_{n,k}$ where H_0 is the Hamiltonian of bulk GaAs. The Schrödinger equation for the superlattice problem is $H\Psi=E\Psi$ where $H=H_0+V$ and

$$V=\sum_j [V(Ga_{1-x}Al_xAs)_j - V(GaAs)_j]$$

is the difference between the microscopic potentials of $Ga_{1-x}Al_xAs$ and GaAs at all sites j in the alloy (barrier) layers. This equation is solved numerically by direct matrix diagonalization. A detailed description of our method has been given in Ref. 4. The values of k in the expansion for Ψ are uniquely determined by the periodicity of the superlattice along the growth direction $\langle 001 \rangle$. In particular, the wave functions $\varphi_{n,k}$ associated with the bulk Γ and X points are included in this expansion. It has been shown that the potential V can induce a mixing of bulk momentum states $\varphi_{n,k}$ of Γ and X character. This means the wave-function expansion coefficients A_{nk} are large for $\varphi_{n,k}$ with $k\approx 0$ and $k=2\pi/A$. If Ψ is constructed only from bulk functions centered around Γ then we say that Ψ is Γ-like. Similarly if Ψ is constructed only from bulk functions from around $k=2\pi/A$ we say Ψ is X-like.

Since we are dealing with a superlattice, i.e., a struc-

ture of period a, we compute Ψ and E along the cubic axis $\langle 001 \rangle$ from $k=0$ (Γ point) to the edge of the superlattice Brillouin zone at $k=\pm\pi/a$ (Z point), i.e., $\Psi \equiv \Psi_{\kappa,m}$ and $E \equiv E_{\kappa,m}$ where κ is the reduced wave vector in the small (superlattice) Brillouin zone and $m=1,2,...$ labels subbands. Hence we have two sets of quantum numbers, one labeling the bulk wave vectors and bands (k,n) and the other the superlattice wave vectors and minibands (κ,m). In our geometry k ranges from Γ ($k=0$) to X ($\pm\pi/A$) and κ ranges from $\Gamma(\kappa=0)$ to $Z(\pm\pi/a)$. In the absence of momentum mixing, the new quantum states $|\kappa,m\rangle$ are obtained simply by folding the bulk band-structure states $|k,n\rangle$ into the small Brillouin zone of length $2\pi/a$.[12] The character of the rapidly varying part of the wave function is not changed in this process and it is therefore useful to refer to the superlattice states in terms of their bulk origin, i.e., Γ-like, X-like, etc.

To begin with, the superlattice parameters are chosen so that the wave function of the lowest conduction state $\Gamma 1$ at all points across the superlattice Brillouin zone is derived from the region of k space around the principal Γ minimum of the bulk GaAs band structure, i.e., the wave function of this state has appreciable A_{nk} for $\varphi_{n,k}$ with $k=0$ and n corresponding to the first GaAs (bulk) conduction band. The second conduction state in the superlattice is derived from the secondary (bulk) X minimum. The wave function for this state is therefore dominated by large A_{nk}'s with $k \approx \pi/A$.

In Fig. 1 we show the subband dispersion along the superlattice growth axis from Γ to Z for a GaAs(6 monolayer)–$Ga_{0.8}Al_{0.2}As$(6 monolayer) superlattice. (One monolayer is defined to consist of one Ga atom and one As atom in the GaAs layer.) The state $X1$ is derived from the X minima of the alloy layer which lie lower in energy than the bulk X point in GaAs. The states labeled $\Gamma 2$ and $X2$ are the first excited Γ-like and X-like states, respectively. For clarity all higher subbands have been omitted.

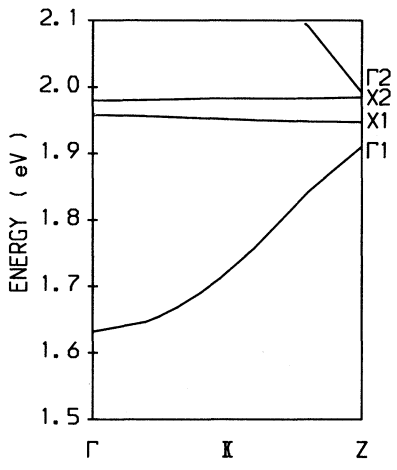

FIG. 1. The subband dispersion along the superlattice growth direction from Γ to Z for a GaAs(6 monolayer)–$Ga_{0.8}Al_{0.2}As$(6 monolayer) superlattice. State $\Gamma 1$ is the lowest conduction state. The superlattice wave function Ψ for this state is constructed from bulk GaAs wave functions derived from the Γ point at the center of the bulk Brillouin zone. $\Gamma 2$ is the first excited superlattice state whose wave function is also derived from points around the bulk Brillouin-zone center. The wave function of state $X1$ contains contributions primarily from the bulk X minima. The energies are measured from the top of the valence band of bulk GaAs. The band gap at Γ is 1.523 eV. κ is the reduced wave vector lying in the small superlattice Brillouin zone.

Let us now consider the effect upon the subbands if the aluminum fraction in the alloy is reduced. In Fig. 2 the subband dispersion for a GaAs(6 monolayer)–$Ga_{0.9}Al_{0.1}As$(6 monolayer) superlattice is shown. Figure 2 shows that the state at Z labeled $\Gamma 2$ crosses the ground X-related state labeled $X1$ around $\frac{4}{5}$ of the way from Γ to Z. As a result of this crossing the relative magnitude of the bulk contributions to the wave functions of states $\Gamma 2$ and $X1$ change. The effect of altering the aluminum composition of the alloy layers upon this crossing may be clearly seen by comparing Fig. 2 to Fig. 1. The increase in the aluminum fraction drives the excited Γ-like state $\Gamma 2$ above $X1$ at Z. The important point to be observed from Figs. 1 and 2 is that in Fig. 2 the subbands ($X1$ and $\Gamma 2$) lying nearest the lowest conduction subband interact. In contrast, increasing the aluminum fraction (Fig.1) prevents $X1$ and $\Gamma 2$ from interacting since in this system $\Gamma 2$ lies above $X1$ over the full width of the superlattice Brillouin zone.

As the interacting states become momentum mixed, their wave functions take on both bulk X-like and Γ-like characteristics. The effect of this mixing on optical transitions between the lowest-energy state $\Gamma 1$ and the states $\Gamma 2$ and $X1$ is best shown by considering the variation of the oscillator strength with κ across the superlattice Brillouin zone. For comparison, this variation is calculated for both the GaAs-$Ga_{0.9}Al_{0.1}As$ and the GaAs-$Ga_{0.8}Al_{0.2}As$ superlattices and is shown in Fig. 3. The

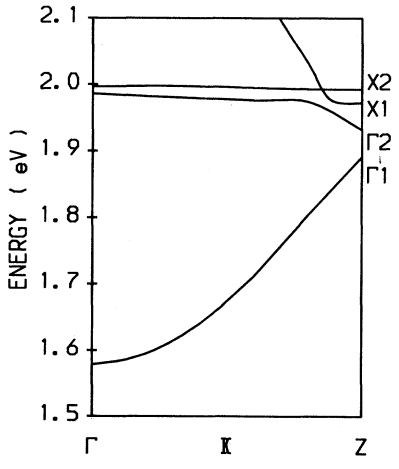

FIG. 2. The subband dispersion along the superlattice growth direction from Γ to Z for a GaAs(6 monolayer)–$Ga_{0.9}Al_{0.1}As$(6 monolayer) superlattice. From this figure it can be seen that state $\Gamma 2$ crosses state $X1$ around $\frac{4}{5}$ of the way from Γ to Z.

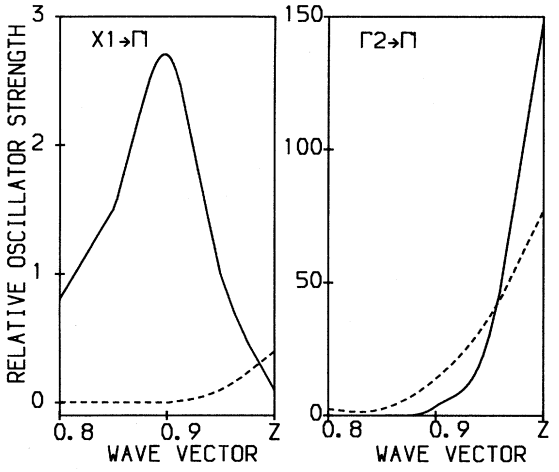

FIG. 3. The variation in the oscillator strength for the transitions between the states $\Gamma 1$ and $X1$ and $\Gamma 2$ across the superlattice zone for κ from $\frac{4}{5}$ (Γ to Z) to Z is shown. The solid curve corresponds to the GaAs(6 monolayer)–$Ga_{0.9}Al_{0.1}As$(6 monolayer) superlattice, the dashed curve to the GaAs(6 monolayer)–$Ga_{0.8}Al_{0.2}As$(6 monolayer) superlattice. Notice that the $\Gamma 1 \rightarrow X1$ transition in the GaAs-$Ga_{0.9}Al_{0.1}As$ superlattice shows a maximum away from Z corresponding to the $\Gamma 2$-$X1$ crossing. No maximum is seen in the $\Gamma 2$-$X1$ curve for GaAs-$Ga_{0.8}Al_{0.2}As$. All oscillator strengths are quoted as a ratio to the oscillator strength between the two lowest Γ-related states at the superlattice minizone center in a GaAs(16)-$Ga_{0.75}Al_{0.25}As$(16) superlattice. [The oscillator strength is defined to be $2|M_{if}|^2/(E_{if}m)$: M_{if} is the optical matrix element between superlattice states i and f in the usual dipole approximation. E_{if} is the energy separation between these states and m is the free electron mass.]

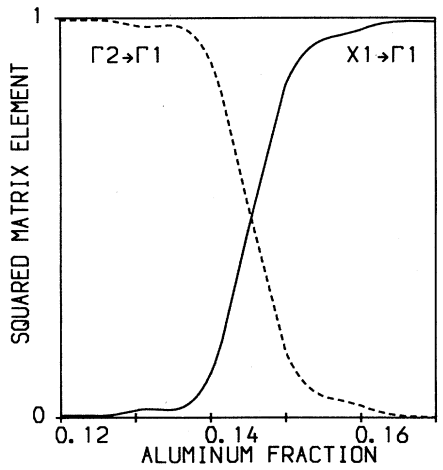

FIG. 4. The variation of the modulus squared of the optical matrix elements at Z with aluminum fraction x in a GaAs(6 monolayer)–$Ga_xAl_{1-x}As$(6 monolayer) superlattice. The matrix elements between $X1$ and $\Gamma 1$ are shown as a solid line, the dashed line corresponds to the $\Gamma 2$ to $\Gamma 1$ transition. The curves cross at $x \sim 0.15$. At this point $\Gamma 2$ crosses $X1$ at Z. These matrix elements are given as a ratio to the matrix element between $\Gamma 2$ and $\Gamma 1$ at Z in the GaAs(6 monolayer)–$Ga_{0.9}Al_{0.1}As$(6 monolayer) superlattice.

latter is depicted by a broken curve. Notice that the magnitude of the oscillator strength for the $\Gamma 1$ to $X1$ transition for the GaAs-$Ga_{0.9}Al_{0.1}As$ superlattice exhibits a maximum. This is in contrast to the GaAs-$Ga_{0.8}Al_{0.2}As$ superlattice where this oscillator strength is roughly constant and much smaller across the full width of the superlattice zone.

We can also see in Fig. 2 that the energy separation of the levels $\Gamma 1$ and $X1$ at the crossing is ~ 150 meV. Since state $\Gamma 2$ may be driven to cross $X1$ at Z by altering the aluminum fraction in the barrier layer, the energy separation between $\Gamma 1$ and $X1$ at the crossing may be tuned over a range of energies within the infrared band.

This crossing is shown in more detail in Fig. 4 where the variation of the modulus squared of the optical matrix elements between states $X1$ and $\Gamma 1$ and $\Gamma 2$ and $\Gamma 1$ with aluminum fraction at Z is presented. From this figure it is seen that $\Gamma 2$ is driven to cross $X1$ at the superlattice minizone edge when the aluminum fraction contained in the alloy layer is $x \sim 0.15$.

Let us inspect the variation of the superlattice energy levels of the three lowest conduction states at Z with aluminum fraction. This variation is shown in Fig. 5 which illustrates the crossing of $X1$ and $\Gamma 2$. By comparing this figure to Fig. 4 it is seen that the large oscillator strength for the transition between $\Gamma 1$ and $X1$ occurs for energy separations between these states of around 50 meV.

This demonstrates quite clearly the degree to which Γ-X mixing can be used to widen the scope for optimization of the infrared transition energies and bandwidths.

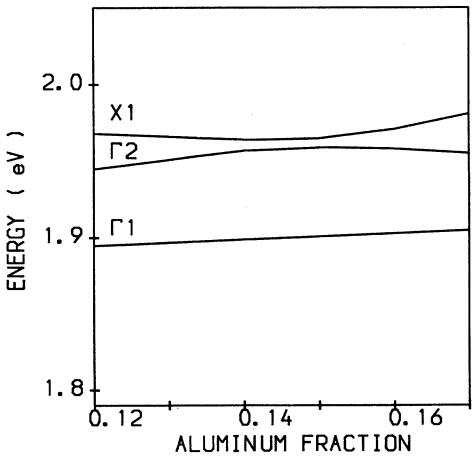

FIG. 5. The variation of the three lowest superlattice energy levels with aluminum fraction. Comparing this figure with Fig. 4 it is verified that the crossing curves in Fig. 4 correspond to the crossing energy levels of $X1$ and $\Gamma 2$. The energy is measured from the top of the valence band of bulk GaAs.

Without this mixing, the energies follow the particle in a box theory and there is a rigid link between the optical gaps and subband widths.[2] We have also shown that the Γ-X mixing alters the transition probabilities between the crossing levels and the remaining subbands.

In conclusion, we have given an example of novel strong optical transitions between conduction subbands which can be tuned over a wide range of frequencies in the infrared spectrum. We predict that this tunability is greatly increased by exploiting band-structure effects such as momentum mixing across the bulk Brillouin zone (i.e., Γ-X mixing). The transition energy, bandwidth, and the strength of the optical transition can be optimized. Such optimization cannot be achieved by simple zone folding. Our predictions provide quantitative guidelines for fresh optical experiments on intrinsic and lightly doped GaAs-$Ga_{1-x}Al_xAs$ superlattices.

We would like to thank the Science and Engineering Research Council (U.K.), the Procurement Executive (Ministry of Defence) Royal Signals and Radar Establishment, Malvern, and the Office of Naval Research (U.S.) and ONR (U.S.A.) Contract No. N00014-88-J-100 for financial support.

[1]See, for example, L. C. West and S. J. Eglash, Appl. Phys. Lett. **46**, 1156 (1985); S. Y. Yuen, *ibid.* **43**, 813 (1983).

[2]See, for example, M. Jaros, *Physics and Applications of Semiconductor Microstructures* (Oxford University Press, Oxford, 1989).

[3]E. Caruthers and P. J. Lin-Chung, Phys. Rev. B **17**, 2705 (1978).

[4]M. A. Gell, D. Ninno, M. Jaros, and D. C. Herbert, Phys. Rev. B **34**, 2416 (1986).

[5]D. Z. Y. Ting and Y. C. Chang, Phys. Rev. B **36**, 4359 (1987).

[6]K. J. Moore, G. Duggan, P. Dawson, and C. T. Foxon, Phys. Rev. B **38**, 5535 (1988).

[7]M. A. Gell, D. Ninno, M. Jaros, D. J. Wolford, T. F. Keuch, and J. A. Bradley, Phys. Rev. B **35**, 1196 (1987).

[8]A. Ishibashi, Y. Mori, T. Itabashi, and N. Watanabe, J. Appl. Phys. **59**, 2503 (1986).

[9]D. J. Wolford, T. F. Keuch, J. A. Bradley, M. A. Gell, D. Ninno, and M. Jaros, J. Vac. Sci. Technol. B **4**, 1043 (1986).

[10]B. F. Levine, R. J. Malik, J. Walker, K. K. Choi, C. G. Bethe, D. A. Kleinman, and J. Vandenberg, Appl. Phys. Lett. **50**, 273 (1987).

[11]N. C. Jarosik, B. D. McCombe, B. V. Shanabrook, J. Comas, J. Ralston, and G. Wicks, Phys. Rev. Lett. **54**, 1283 (1985).

[12]L. D. L. Brown, D. Ninno, and M. Jaros, Phys. Rev. B **36**, 2935 (1987).