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The role of layer width in valley mixing and cross-interface recombination in GaAs–AlAs superlattices

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Abstract. We have carried out pseudopotential calculations of the effect of mixing in semiconductor superlattices between bulk states derived from the conduction band minimum at the centre of the bulk Brillouin zone and those of the X minima at the zone edges. We present a systematic account of (i) the strength of this coupling in GaAs–AlAs superlattices as a function of the layer widths, (ii) the variation as a function of layer width of the magnitude of the energy gap that occurs when the levels derived from different bulk valleys cross, (iii) the change of the oscillator strength of the transition across the superlattice gap as a function of the separation of the interacting levels, and (iv) the magnitude of the effect of strain on valley mixing.

1. Introduction

The electronic structure of semiconductor superlattices formed by growing a sequence of thin layers of alternating materials such as GaAs and AlAs is normally modelled in the framework of the particle-in-a-box picture. In this model, it is assumed that the bulk momentum associated with the Bloch states of the constituent semiconductors remains a good quantum number. The effect of the change of the crystal potential experienced at the interfaces by the bulk waves is represented by a simple potential step (band offset) associated with the bulk state in question. The effect of this barrier is to cause reflections and confinement without destroying the periodic part of the bulk Bloch function. Hence the bulk wave functions are merely modulated by a smooth envelope function and shifted in energy.

However, the symmetry of the bulk crystal potential is broken in the superlattice. Consequently states derived from different parts of the bulk Brillouin zone may interact (for a review of band structure effects in superlattices see, for example, Jaros 1990). This interaction or coupling of levels from different conduction band valleys is usually small but can be observed, particularly when the interacting bulk levels lie close in energy. For example, it is possible to use hydrostatic pressure to alter the separation of the states derived from the principal ($k = 0$) and secondary (X) minima in GaAs–AlAs superlattices and observe luminescence as a function of the level separation. The optical transition from the X-like levels to the valence states at the top of the valence band at $k = 0$ that are forbidden in a perfect semiconductor become allowed as a result of this momentum mixing. As we move the levels towards the crossing, the changes of the strength of the luminescence signal span several orders of magnitude since they represent

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a change from forbidden to allowed transitions. The magnitude and width of the effect of valley mixing is a clear microscopic signature of the level crossing. Furthermore, it has been demonstrated that it can be used as a sensitive tool for characterization of the microscopic quality of the interface which is reflected in the ordering and strength of the individual X-related transitions.

Numerous theoretical and experimental studies of the effect of valley mixing (Ando and Akera (1989), and references therein) have been made. However, these studies have been concerned with individual aspects of the ordering of the mixed states and with the strength of the mixing in specific GaAs–AlAs structures. Accordingly, in the present study we set out to provide a systematic account of the variation of the strength of the valley mixing as a function of the layer width over the range of widths where the coupling is significant. We also present a systematic account of the magnitude and rate of change of the optical transition probability across the superlattice gap in the vicinity of the pressure-induced level crossing as a function of the layer width. These results are then used to analyse a set of typical data and provide general guidelines for assessment of experimental results concerning cross-interface recombination. Finally, we address the question of the magnitude of the contribution to momentum mixing associated with the presence of strain. So far the effect of strain has either been ignored altogether or considered only to the extent to which it shifts the relevant energy levels.

2. The strength of valley mixing versus layer widths

The calculations reported here have been carried out using the pseudopotential scheme in which the microscopic potentials of the constituent semiconductors are represented by empirical potentials fitted to reproduce the bulk band structure near the band edges. The wave functions of the superlattice are then computed as a linear combination of the bulk Bloch states defined unambiguously by the periodic boundary condition (see, for example, Gell *et al* 1987). The expansion coefficients $A_{n,k}$ determine the degree of participation in the superlattice wave function of the bulk states associated with the corresponding bulk reduced wave vector k and bulk band n. This facilitates a direct determination of the degree to which, for example, states of bulk X character contribute to a given superlattice state. This method has been used extensively in the past; a detailed account of it can be found in the literature and will not be repeated here.

The relative alignment of the bulk states derived from the principal and secondary (X) minima of the conduction band of GaAs is shown in figure 1. This figure also illustrates the relative position in energy of the wells confining states of X character and those of the states associated with bulk zone centre. This alignment can be changed in several ways, for example by changing the Al composition in the barrier. This changes the band offset and consequently the separation of the individual valleys. The most attractive way of altering this separation is, however, that achieved by applying hydrostatic pressure. The pressure pushes the states from the principal valley up in energy and closer to the X valleys. This means that the separation of the valleys can be altered in a continuous fashion and without switching samples. It is an important advantage since the quality of the sample is always difficult to assess. In the pseudopotential calculation the effect of pressure is modelled so as to account accurately for the observed pressure coefficients of the ground state. The empirical potentials and band offsets are adjusted accordingly as described in detail by Gell *et al* (1987).

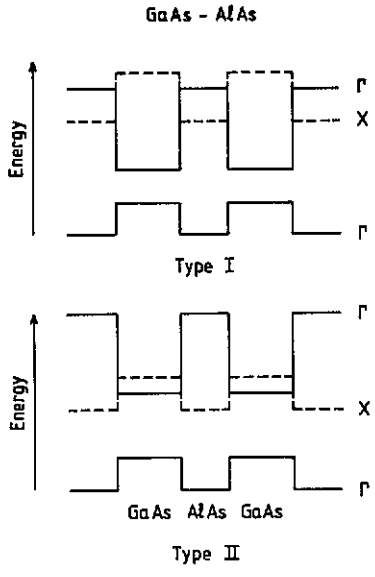

Figure 1. The band offsets for GaAs–AlAs. Upper diagram: type I structure in which both electrons and holes at the band edge are confined in GaAs. Lower diagram: type II structure which occurs when the principal valley of GaAs is driven up in energy by the application of hydrostatic pressure. The position of the wells confining electrons derived from the bulk X valleys is also indicated. The superlattice growth direction is [001].

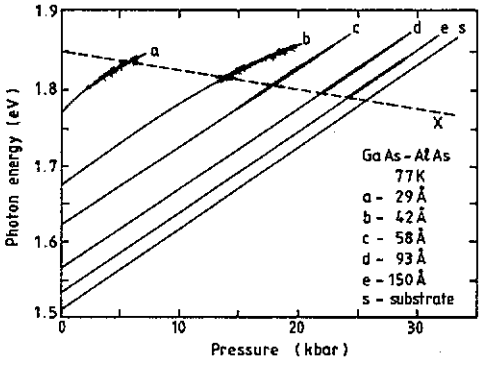

Figure 2. The energy of the direct gap luminescence as a function of hydrostatic pressure. A fit to the data reported by Masumoto *et al* (1989) is shown for isolated GaAs quantum wells a, b, c and d whose widths are indicated in the inset. s is the reference GaAs substrate luminescence. The shaded line marks the onset of the observed drop of the luminescence signal. It ends at the point where the luminescence has been reduced to 0.1% of its original value. The line marked X indicates the theoretical position of the X minimum in the AlAs barrier.

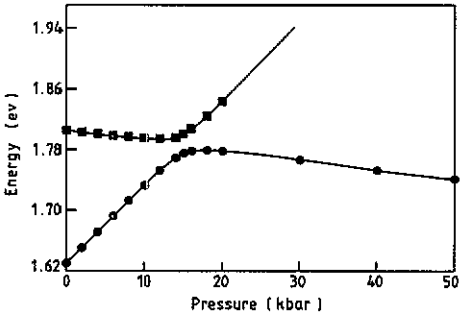

Figure 3. The variation of the lowest levels derived from the principal conduction band valley of GaAs and (the upper level) from the X valley as a function of hydrostatic pressure. The strength of their interaction is translated into the magnitude of the gap at the crossing. All energies are measured with respect to the top of the valence band of GaAs.

A typical set of level-crossing data (Masumoto *et al* 1989) from such a pressure experiment is shown in figure 2. It illustrates, first of all, the movement of the levels under pressure. The data are unique in that they cover a systematically chosen set of samples with different layer widths. These results also illustrate the difficulties encoun-

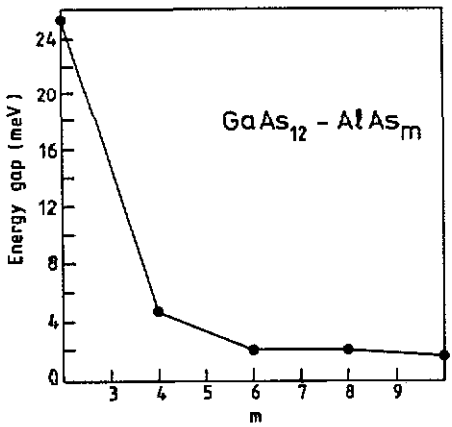

Figure 4. The variation of the magnitude of the gap between the interacting levels of Γ and X character at the crossing point as a function of barrier width.

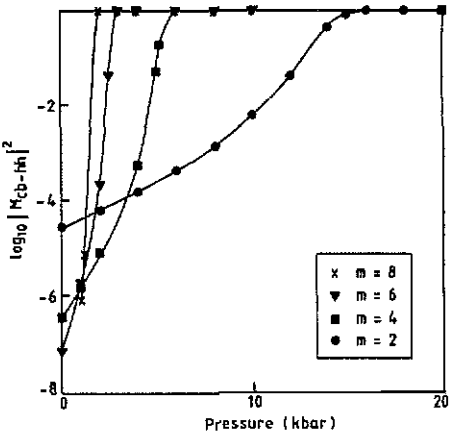

Figure 5. Pressure dependence of the strength of the optical matrix element M between the lowest X-like conduction level (cb) and the state representing the heavy hole (hh) state at the top of the valence band. The figure shows the rate with which the matrix element M vanishes away from the crossing. m indicates the number of monolayers of AlAs. The thickness of the GaAs layer is fixed at 12 monolayers.

tered when it comes to the interpretation of such data. The shaded region begins at the onset of the drop in the direct gap luminescence intensity and ends at the point where the intensity is reduced to 0.1% of its original value. One can see, for example, that the onset of the decrease does not occur quite where the idealized theory would predict it (i.e. in an intrinsic system with sharp interfaces the drop should commence within a few milli-electron volts of the level crossing). This highlights the importance of theoretical understanding of the properties of the crossing energy levels.

In figure 3 we show the effect of level crossing on the electronic structure obtained in our calculation. The gap shown in figure 3 between the crossing levels is a measure of the strength of the coupling between the two states. This can be best visualized when one considers this crossing as an interaction between two states degenerate in energy. The gap is then equal to twice the matrix element (integral over all space) of the superlattice crystal potential between the wave functions of the two interacting levels. It follows that the simplest way to indicate the strength of the coupling versus layer width is to plot the magnitude of the gap versus layer width. This is done in figure 4. We can see that the magnitude of the coupling is a sensitive function of the layer width. It has been shown that in structures with an odd number of monolayers the valley mixing is forbidden in first order. This is the result of the zone-folded X states and the states from the principal valley belonging to different symmetry representations. However, this theoretical result is valid only for perfect interfaces. Interface imperfections can lift this selection rule and the observed finite coupling strength in such systems is a direct measure of the departure from perfect interfaces or from the desired layer width.

We can see in figure 4 that the magnitude of the calculated gap rapidly decreases with increasing barrier width and falls below 2 meV where it cannot be followed further because it becomes comparable with the error with which we can calculate this quantity.

It must be said that we are referring here to the relative error, i.e. the error with which we can distinguish one structure from the others. The absolute error is difficult to establish but we expect it to be not more than 2 meV.

Although our aim here is to identify the rate of change of the absolute magnitude of the coupling across the bulk Brillouin zone in perfect structures, it should be pointed out that the effect of interface imperfections exerts considerable influence on both the strength of the valley mixing and on level ordering. Such effects are observable as spectral features in luminescence and can be used for characterization of interface quality. We have given a detailed account of such effects in a typical system elsewhere (Morrison *et al* 1990).

3. The optical spectra of mixed states

The physical make-up of the conduction band wave function at and in the vicinity of the crossing as a superposition of states derived from the two coupled valleys is well understood from earlier detailed studies of individual structures (see, e.g., Gell *et al* 1987). Here we concentrate on a systematic study of four sequential structures with an increasing barrier width. The result is shown in figure 5. We can see that the range of energies over which the crossing is observable varies by an order of magnitude depending on which of the four structures we look at. Since the pressure change of one kbar corresponds to a change in energy of the central valley level by about 11 meV, the optical width can be as large as several tens of meV.

We can now examine the experimental results of figure 2 in the light of our theoretical findings. The results presented in figures 4 and 5 show that the variation in the position of the onset of the drop in luminescence intensity shown in figure 2 is much larger than the uncertainty that might be associated with the width of the region where the mixing is at its strongest. With the exception of the smallest well all experimental data are on wider wells and barriers. This means that the overlap in real space of the states derived from the two bulk valleys is drastically reduced compared with that in the cases considered in our calculations and the magnitude and width of the crossing in perfect material must be much smaller than the natural linewidth. The onset variation apparent from the data must therefore be attributed to the uncertainty in the interface definition. A glance at the data in figure 2 also suggests that the rate of decay of the luminescence intensity is not related to the intrinsic properties of the level crossing.

4. The effect of strain

Having established the overall behaviour of the valley mixing in the absence of strain we can now address the question concerning the effect of strain upon the magnitude of the mixing. The role of strain in the analysis of the optical spectra of GaAs–AlAs has been confined to the assessment of the shifts of individual levels due to the difference in the lattice constant of GaAs (substrate) and AlAs layers. No attempt has been made to examine its effect on momentum mixing. However, when the strain is very small as is the case in GaAs–AlAs superlattices grown on GaAs substrates, one must expect its effect upon the mixing to be negligibly small. This is indeed recovered in our calculations.

Assuming that we now have a representative result for the valley mixing in perfect GaAs–AlAs superlattices, it is interesting to compare this result with other structures

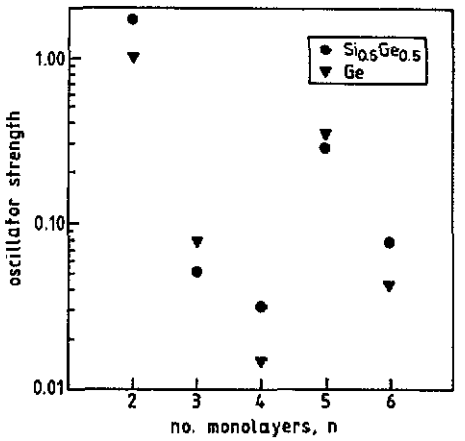

Figure 6. The oscillator strength of the optical transition across the principal gap of the Si_nGe_n superlatice grown either on Ge or on $Si_{0.5}Ge_{0.5}$ substrates.

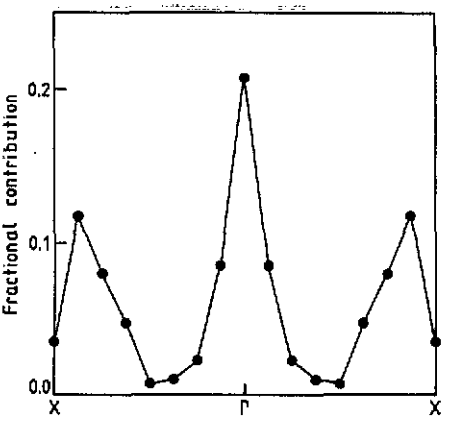

Figure 7. The magnitude of the modulus squared of the coefficients representing the contribution to the wave function of the superlattice state at the bottom of the conduction band of the individual bulk states lying on the ⟨001⟩ cubic axis in the bulk Brillouin zone of Si. The contributions are normalized so that the sum of them is equal to 1. X indicates the bulk zone-edge point.

(e.g. Si–Ge superlattices) where the effect of strain upon X-like levels has been studied extensively though from a different point of view (Pollack 1990). We refer the reader to earlier publications where technical details concerning models of the electronic structure of Si–Ge superlattices can be found (see Jaros 1990). Let us begin by looking at the role of strain distribution. For example, it is possible to show that optical transition probabilities for the jump from the bottom conduction band (mixed) state to the top valence band state in Si–Ge superlattices do not reflect the way in which strain is distributed in the system. This is clear from figure 6 where we show the oscillator strength of the lowest direct transition in Si–Ge superlattices grown on Ge and $Si_{0.5}Ge_{0.5}$ substrates. The oscillator strength varies only slightly with the choice of substate, which determines whether strain is distributed equally in both layers or (in the case of Ge substrate) is found only in Si layers. This is consistent with the view that the mixing is most effective at the interface region where the crystal potential change is at its highest. This trend has also been recovered in all studies of valley mixing in unstrained structures and does not reveal any properties peculiar to the presence of strain.

It is, however, more instructive to look at the absolute strength of the oscillator strength in Si–Ge superlattices. If we assume, for the sake of simplicity, that perturbation theory is approximately valid for the estimate of the magnitude of the coupling between individual bulk states from well separated valleys, we can expect the coupling between the states at the bulk zone edge and centre to be inversely proportional to the square of the energy separation between them. From a simple picture of the band structure of silicon (e.g. Jaros 1989) it is seen that the separation between the conduction band X minimum and that at the centre of the Brillouin zone is much larger (about 3 eV). It follows from the oscillator strengths reported in figure 6 that the coupling of the states across the zone by the difference in the microscopic potentials in the adjacent layers of

Si and Ge is stronger than that in GaAs–AlAs. Indeed, a quantitative analysis of the origin of the strength of the optical transition in Si–Ge shows that the contribution of $A_{n,0}$ from the lowest conduction band minimum relative to that at the zone-edge minimum (figure 7) is incomparably larger than that in GaAs–AlAs. Furthermore, this large $A_{n,0}$ accounts for only a small fraction (around 20%) of the total oscillator strength of the transition across the gap. This is because there is a considerable mixing of X-like bulk states that participate in the expansion of the superlattice wave function. This is also apparent from figure 7. The magnitude of $A_{n,0}$ shown in figure 6 can only be achieved in GaAs–AlAs superlattices for an X-like wavefunction associated with a configuration in which the states from the two valleys are nearly degenerate in energy! Also the contribution of states lying further from the zone centre as well as from the secondary zone edge minima are not significant in structures with the well width considered in figure 4. We must conclude that the effect of valley mixing in strained-layer Si–Ge superlattices is significantly stronger. The corresponding valley mixing in strained III–V heterostructures (e.g. GaAs–$Ga_{1-x}In_xAs$) is therefore expected to be a much stronger effect than in GaAs–AlAs.

Acknowledgments

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