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Energy Bands and Stable Structures of Ultra-Thin- Layer Semiconductor Superlattices

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We present the calculated results of the band structures, effective masses and total energies of ultra-thin-layer superlattices consisting of m-layers of GaAs and n-layers of AlAs in a period, $(GaAs)_m(AlAs)_n$, on the basis of the first-principles full-potential linearlized augmented-plane-wave method. The calculated energy bands for cases of (m,n)=(1,1), and (1,3) exhibit indirect character in the band gap, while the conduction bands for cases of (m,n)=(2,2) and (3,1) have the minima at the Γ point.

1. INTRODUCTION

Recent developments in technology of crystal growth have provided us with one of the foundations for the hitherto unconstructed bridge between macroscopic phenomena in real materials and microscopic first principles. Controlability of locations of each atom in the process of crystal growth has been improved in great degree,[1] and we have almost attained the atom-layer by atom-layer growth of crystal using the technique of molecular beam epitaxy or metal organic chemical vapor deposition. This technique enables us to design new materials which exhibit novel physical properties. For example, in semiconductor super-lattice-structures, we are almost in a stage that impurities could be doped at desired places so as to obtain the required transport and optical properties.[2]

This progress in technology, in turn, challenges new problems to theoretical physicists, since empirical approach in calculation of physical properties, which has met great success in ordinary bulk materials, has no guarantee for the calculated quantities in intentionally designed structures or new materials. Computational physics with the aid of supercomputer, however, is a promissing candidate to respond the challenging problem to reveal the origin of the macroscopic phenomena in novel structures or materials from the first principles. Density Functional Theory[3] implemented on computers provides theoretical values of the ground state properties such as lattice constant, cohesive energy or bulk modulus for various kinds of materials[4] without any spurious adjustable parameters. The deviation of the theoretical value from the experimental value for lattice constant is generally less than 1%. The aim of the present paper is thus to apply this theoretical method in computational physics to the novel structures, and to predict their characteristic features.

We have developed full-potential linearlized augmented-plane-wave (FLAPW) method which gives electronic structures and total energies of any materials from the first principles. There is no ambiguity in calculational procedure of ground state properties: We solve an effective Schrodinger equation (Kohn-Sham equation)[5] exactly within the local density functional approximation. The calculation of energy-band gap, however, requires the description of the excited states. No conclusive formulation has been prepared from the theoretical point of view for the description of the excited states. Therefore, we introduce the new scheme of self-interaction correction (SIC) in calculation of energy-band gaps. This new scheme has already

applied by the present authors[6], and satisfactory agreement between theoretical and experimental values is achieved for bulk materials such as diamond, Si, GaAs and AlAs. With this firm theoretical framework, we study, in the present paper, electronic and atomic structures of ultra-thin-layer superlattices consisting of m-layers of GaAs and n-layers of AlAS ($(GaAs)_m(AlAs)_n$) in one period. We present the calculated results of the band structures , effective masses and total energies of this technologically important superlattices.

2. CALCULATIONAL METHODS

Since the detailed description of the calculational method in the present paper has been described elsewhere[6], only the brief sketch of the method is given in this section.

The total energy, E, of the condensed matter in which electrons interact with each other is given as a functional of electron charge density $n_e(r)$:

$$E = T[n_e] - \sum_i Z_i \int \frac{n_e(r)}{|r-R_i|} dr + \frac{1}{2} \iint dr\, dr' \frac{n_e(r)\, n_e(r')}{|r-r'|} + E_{nuc-nuc} + E_{xc}[n_e] \qquad (1)$$

where $T[n_e]$ is the kinetic energy, and the 2'nd and 3'rd terms are electrostatic energy between nucleus Z_i and electrons, and Hartree energy among electrons, respectively. Further, $E_{nuc-nuc}$ is the electrostatic energy between nuclei, and finally $E_{xc}[n_e]$ denotes exchange-correlation energy of interacting electrons. We use local-density approximation for the expression of E_{xc}.[7] An effective one-electron equation (Kohn-Sham equation) can be introduced in density-functional scheme:

$$\left\{ -\frac{\nabla^2}{2} - \sum_i \frac{Z_i}{|r-R_i|} + \int \frac{n_e(r')}{|r-r'|} + \frac{\delta E_{xc}}{\delta n_e} \right\} \psi_i(r) = \varepsilon_i \psi_i(r) \qquad (2)$$

and

$$n_e(r) = \sum_{i:\,occupied} |\psi_i(r)|^2 \qquad (3)$$

Since equation (2) includes the electron charge density $n_e(r)$ which is, in turn, calculated through equation (3), it is reqired to solve (2) and (3) selfconsistently. When a set of basis functions is introduced, the differential equation (2) is converted to a secular equation. The energy-band structure is obtained as eigenvalues of this secular equation, and the electron charge density is calculated from the eigenvectors and the basis functions. Selfconsistent electron-charge density is obtained by the iterative calculation until input charge agrees with the output charge. We emphasize that all the calculation is rigorous within the framework of density functional theory, and free from adjustable parameters.

The total energy and the eigenvalues of ocuupied states obtained in the local density approximation has been proved to be reliable enough for various kinds of materials.[4] The locations in energies of conduction bands (absolute eigenvalues of unoccupied states), however, are unable to be obtained in the local density approximation with satisfactory agreement with the experiments, although the overall structures of the conduction bands within this approximation agrees well with the experiments. This is because the local density approximation overestimats the Coulomb energy for the electrons at the occupied states.[8] Thus, we introduced self interaction correction to the local density approximation (SIC-LDA) . This correction gives a satisfactory agreement with the experimental values for several materials[6]

3. Results and Discussion

A) Band structures and effective masses of $(GaAs)_m(AlAs)_n$

We now present the calculated results for the superlattices, $(GaAS)_m(AlAs)_n$, which is grown

along (001) direction. The corresponding Brillouine zones of original zinc-blend structure and the superlattice structures are shown in Fig. 1. Due to the long period along (001) direction, the original Brillouine zone of zinc-blend structure is folded into smaller zones in cases of the superlattices.

The calculated band-structures of $(GaAs)_1(AlAs)_1$, $(GaAs)_2(AlAs)_2$, $(GaAs)_1(AlAs)_3$ and $(GaAs)_3(AlAs)_1$ are presented in Fig. 2, Fig. 3, Fig. 4 and Fig.5. The value of the conduction band minima measured from the top of the valence bands for these superlattices are also shown in Table. 1 and Fig. 6. When the widths of AlAs layers becomes thin, the electron wavefunctions spreads in the space over not only in GaAs layers but also in AlAs layers. The folding effect of the Brillouine zones results in the mixing up of the wavefunctions of several conduction band minima.

The most striking feature of the conduction bands of ultra-thin-layer superlattice, $(GaAs)_m(AlAs)_n$, is thus the appearence of the indirect character in the energy-gap, which is in sharp contrast with the ordinary multi-quantum wells of GaAs and AlAs. This indirect character obtained from the first-principles calculation in the present paper is consistent with recent experimental results on photoluminescense.[9]

In Table 2, the calculated effective masses are presented for $(GaAs)_m(AlAs)_n$. The effective masses of the conduction bands is comparable with those in GaAs. This means that the high mobility of GaAs is preserved in ultra-thin-layer superlattice structures if sharp interfaces are formed with the excellent fabrication technique.

B) Total energy and stability

The total energy can be calculated with equation (1) when selfconsistent electron charge density is obtained from the band-structure calculation. We have performed the total energy calculation for GaAs, AlAs, and $(AlAs)_1(GaAs)_1$ as a step toward the general understanding of the stability of ultra-thin-layer superlattice structures. Preliminary calculations shows that the total energy of $(AlAs)_1(GaAs)_1$ is lower than the averaged total energy of GaAs and AlAs. This suggests that the superlattice structures are stable, and is consistent with the recent observation of long-range order in $Al_xGa_{1-x}As$.[10]

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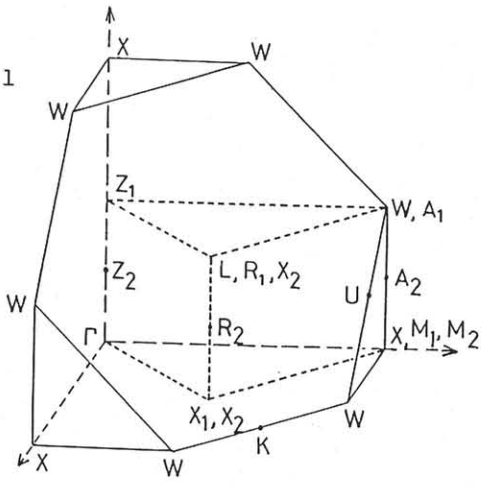
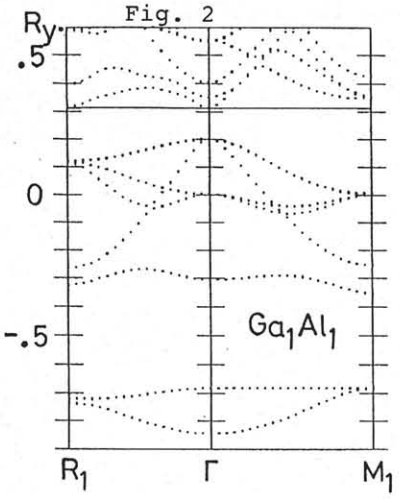
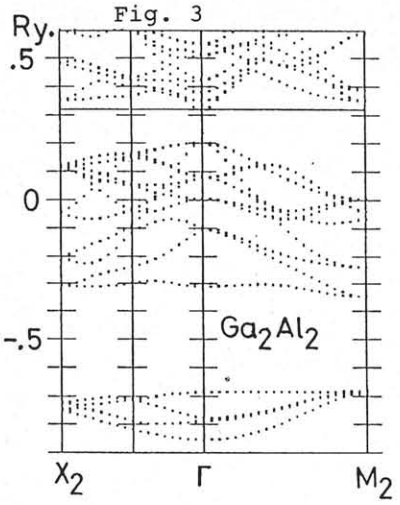
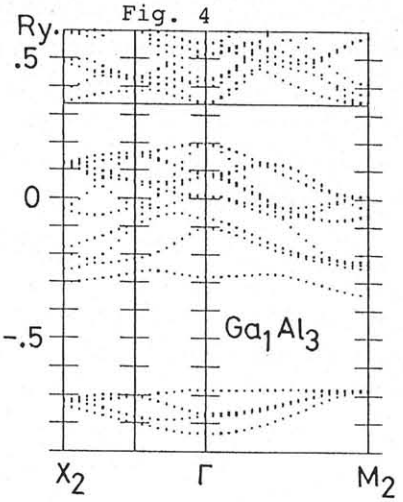
Fig. 1









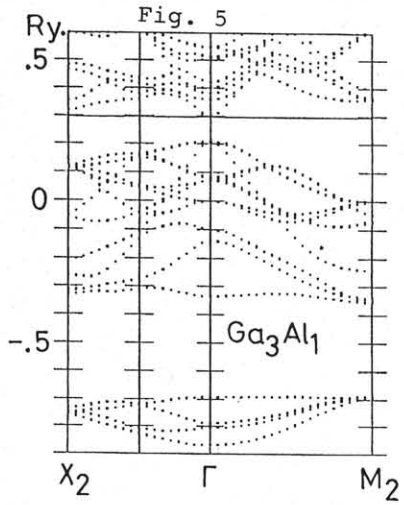
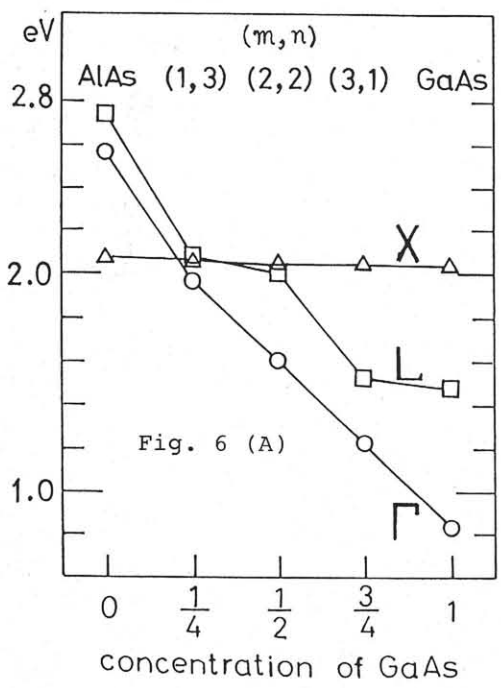


Fig. 2-5. Dispersion relations for superlattices $(GaAs)_m(AlAs)_n$

Table 2. Effective masses of conduction band at Γ point

m n	$m^*(110)$	$m^*(001)$
1 1	0.081	0.125
2 2	0.078	0.080
1 3	0.101	0.202
3 1	0.060	0.061

(110)=Γ-M_1 or Γ-M_2 direction



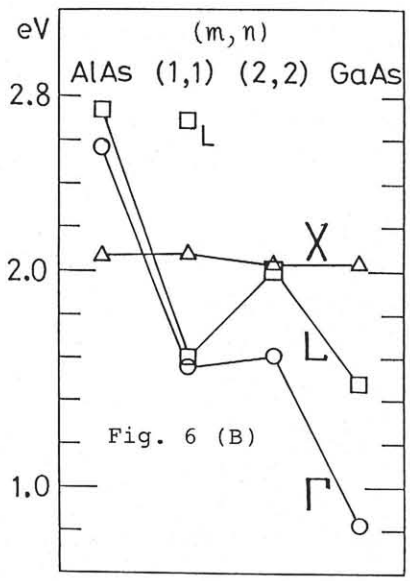


Table 1. Conduction band minima measured from valence band top

m n	Γ	L	X
1 1	1.56	1.61	2.07
2 2	1.61	2.01	2.03
1 3	1.97	2.08	2.06
3 1	1.23	1.53	2.04