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Ab initio based atomic scattering amplitudes and {002} electron structure factors of $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$ quantum wells

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Abstract. The atomic scattering amplitudes of the various atoms of the systems $\text{Ga}_{1-x}\text{In}_x\text{As}$, $\text{GaAs}_{1-x}\text{N}_x$ and $\text{InAs}_{1-x}\text{N}_x$ are calculated using the density functional theory (DFT) approach. The scattering amplitudes of N, Ga, As and In in the model systems are compared with the frequently used Doyle and Turner values. Deviation from the latter values is found for small scattering vectors ($s < 0.3 \text{ \AA}^{-1}$) and for these scattering vectors dependence on the orientation of the scattering vector and the chemical environment is reported. We suggest a parametrization of these modified scattering amplitudes (MASAs) for small scattering vectors ($s < 1.0 \text{ \AA}^{-1}$). The MASAs are exploited within zero pressure classical Metropolis Monte Carlo (MC), finite temperature calculations to investigate the effect of quantum well size on the electron {002} structure factor (SF) of $\text{Ga}_{1-x}\text{In}_x\text{As}$ quantum wells.

1. Introduction

Accurate chemical composition analysis of technologically important III-V nanostructures is of importance. It is known that minute compositional variations can induce large changes in the photoluminescence properties of these materials. Using transmission electron microscopy (TEM), chemical composition quantification can be carried out with methods based on the composition sensitive {002} reflection [1] obtained from the atomic positions and the atomic scattering amplitudes (ASAs) of the atoms of the system. The most commonly used set of ASAs is that based on the relativistic Hartree-Fock (HF) calculations of Doyle and Turner (DT) on isolated atoms [2]. Using density functional theory (DFT) calculations on the binary crystals, Rosenauer et al. [3] demonstrated the importance of using DFT to obtain more accurate modified atomic scattering amplitudes (MASAs). The latter set of parameters is more accurate since it accounts for charge redistribution in the system as a result of bonding. However, the MASAs that have so far been used are those obtained from DFT calculations based on the binary zinc-blende crystals. The effect on the MASAs of the static atomic displacements present in the ternary and quaternary systems has never been investigated. This effect should become very important in technologically important $\text{Ga}_{1-x}\text{In}_x\text{As}_{1-y}\text{N}_y$ where the insertion of nitrogen can introduce atomic displacements of more than 15% of the GaAs bond length. The study of the effect of static displacements on the structure factor (SF) [3-5] often ignores this important issue. An accurate composition analysis based on the structure factor should incorporate these effects at all levels. Another issue of interest is the system size dependence of the {002} electron structure factor of nanostructured III-V semiconductors.

In this work we have performed DFT calculations on a series of $\text{Ga}_{1-x}\text{In}_x\text{As}$, $\text{GaAs}_{1-x}\text{N}_x$, $\text{InAs}_{1-x}\text{N}_x$ and the binary counterparts, all in the zinc-blende phase, with unrelaxed and relaxed atomic coordinates. Within the formulation of ref. [3] we have calculated the MASA of Ga, In, As and N in various chemical environments. The MASAs of all these atoms are compiled as a function of the magnitude of the scattering vector $s = \sin \theta / \lambda$ (where θ is the scattering angle and λ is the wavelength) and compared with the results of Doyle and Turner [2]. Then special attention is devoted to the relaxation of the multi quantum wells (MQWs) and the calculation of the {002} structure factors.

2. Theoretical considerations

The DFT calculations of the MASAs are performed using the all-electron full-potential-linearized augmented-plane-wave (LAPW) DFT code WIEN2k [6]. The MASAs are calculated using the method developed in ref [3]. These MASAs will be used to evaluate the SFs of the strained MQWs. QWs with In fractions of 20, 40, 60 and 80% are considered. InGaAs/GaAs QWs with In content as high as 80% have not been synthesized due to a dislocation-induced transition from two-dimensional (2D) to 3D growth mechanism for increasing well size. In fractions as high as 80% were considered in our simulations to investigate the effect of the inherent high strain present in such systems on the use of the electron structure factor for chemical composition quantification. The InGaAs/GaAs heterostructures have been relaxed as follows. The unit cell used in our simulation consists of a slab of InGaAs QW of thickness t_w on a GaAs barrier of thickness t_b . While the lateral widths along the [100] and [010] directions of the box are kept fixed in order to simulate a pseudomorphic growth condition on the GaAs barrier, the atomic positions and the length $L = t_w + t_b$ of the simulation box in the growth direction (i.e. along the [001] direction) is relaxed using a zero pressure Metropolis Monte Carlo Scheme. Periodic boundary conditions were considered in all three dimensions, whereby if an atom exited the box at one end, its periodic image in the other end is retained. We use the Tersoff potential [7] that was recently parametrized by us [8] using accurate ab initio energies and the experimental elastic constants of the binary crystals.

3. MASA

DFT calculations were performed on systems containing Ga, In, As and N either as binary crystals (GaAs, InAs, GaN and InN all in the zinc-blende phase) or on supercells made of 64 atoms mixed ternary $\text{III}_{1-x}\text{III}_x\text{V}$ or $\text{V}_{1-x}\text{V}_x\text{III}$ crystals. The mixed systems were relaxed using the DFT approach and the average MASA of each atom type was calculated. In Fig. 1 we plot the MASAs for various reflections and for all the model systems as a function of the magnitude of the scattering vector s . An acceleration voltage of $U=200$ kV is used. The spread of the MASAs demonstrate their chemical environment sensitivity. As Fig. 1 shows, for Ga, As and In, the HF calculations accurately reproduces the DFT results for $s > 0.3 \text{ \AA}^{-1}$. For smaller s -values deviations (towards larger values) from the HF calculations become quite important. The agreement for N is poor, thus discrediting the use of the DT ASAs on nitrogen containing systems. However the agreement of the HF results with our DFT calculations for scattering vectors larger than $s = 0.3 \text{ \AA}^{-1}$ encourages the use of the DT results for higher order reflections like the {220} and {004} reflections. Another important aspect of the DFT result is the orientation sensitivity at small scattering vectors which is exemplified by {111} spots. We have also found that for small scattering vectors, atomic relaxation increases the MASA (see Fig.2).

In order to account for all these effects on the MASA, we performed a fit of the MASAs of Ga, In, As and N as a function of s . For these atoms we consider the binary zinc-blende GaAs, InAs, GaN, InN and the relaxed ternary $\text{Ga}_{1-x}\text{In}_x\text{As}$, $\text{GaAs}_{1-x}\text{N}_x$ and $\text{InAs}_{1-x}\text{N}_x$ systems. In analogy with the work of Doyle and Turner [2], who needed parameter tables for economic data storage, we retained $f(s)$ (in nm) as the sum of four exponential functions as follows:

$$f(s) = \sum_{i=1}^4 a_{\alpha i} e^{-b_{\alpha i} s^2} \quad (1)$$

where a_{ai} and b_{ai} are fitting parameters and α is the atom type as reproduced in Table 1.

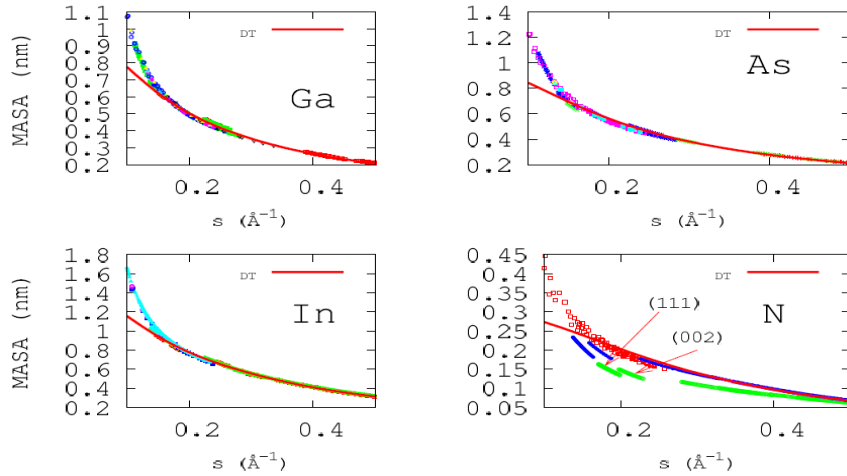


Figure 1. Electron atomic form factors as a function of s compared with the Doyle and Turner (DT) results.

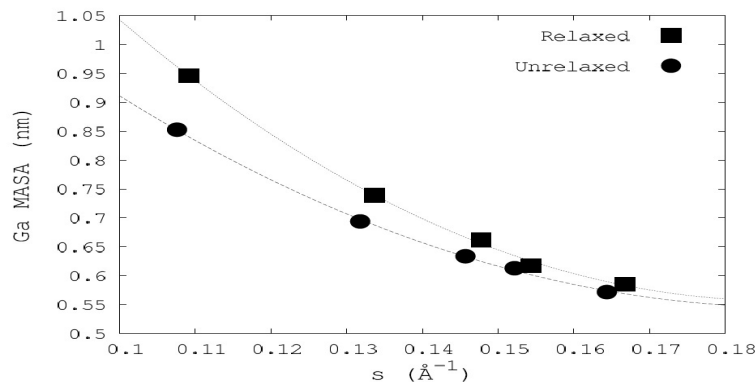


Figure 2. The MASA of Ga for relaxed and unrelaxed $\text{Ga}_{24}\text{In}_8\text{As}_{32}$ system (64 atoms) showing the effect of atomic relaxation on the MASA.

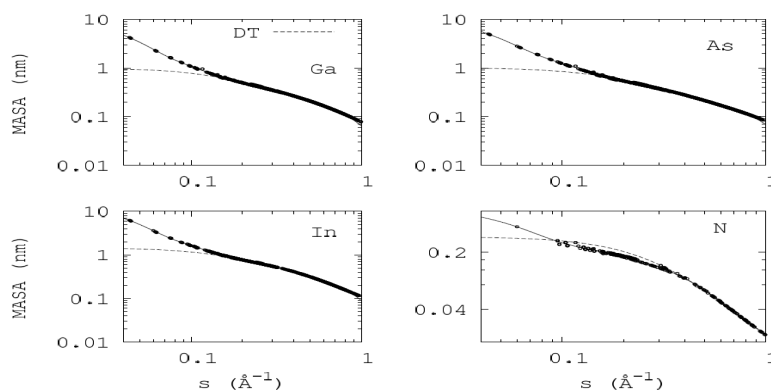


Figure 3. Fitted MASAs for Ga, In, As and N in zinc-blende binary and ternary materials compared with the Doyle and Turner results (DT). The points correspond to DFT calculations for several binary and ternary alloys and the full lines are the fitted expressions.

Figure 3 compares the fitted functions with the DT results for Ga, In, As and N. Table 1 gives the parameters of the fit, which yield very accurate form factors for small scattering vectors. The coefficients a_{ai} must be corrected by the factor $(1 + eU/m_0c^2)$ for the appropriate electron energy.

Table 1. The parameters of the fit of the MASAs of Ga, In, As and N.

Atom α	a_1	b_1	a_2	b_2	a_3	b_3	a_4	b_4
Ga	5.5541	632.8870	1.0827	119.4029	0.2713	14.5702	0.2060	1.4976
In	8.0200	626.1903	1.5765	118.4147	0.4236	13.4353	0.3083	1.5710
As	6.6263	628.0908	1.2804	116.9245	0.3183	13.7264	0.2030	1.4090
N	0.3121	295.5509	0.0544	38.6426	0.1071	6.2606	0.03323	0.8909

4. $\text{In}_x\text{Ga}_{1-x}\text{As}$ / GaAs multi quantum well simulations

Typical layer by layer changes in the bond lengths of the atoms in a MQW are shown in Fig. 4 for a ~10nm wide $\text{In}_{0.4}\text{Ga}_{0.6}\text{As}$ / GaAs quantum well. The pair correlation function shows a bimodal bond length distribution with Ga-As bonds at about 2.4 Å and In-As bonds at about 2.6 Å. The Ga-As bond length is seen to shrink within the well.

In the absence of static displacements and at low temperatures we can write the modulus square of the 'average' {002} SF as:

$$S_0^{002}(x) = |(1-x)f_{\text{Ga}} + xf_{\text{In}} - f_{\text{As}}|^2 \quad (2)$$

It is commonplace to use the SF of the (200) diffraction spot for composition analysis. In $\text{Ga}_{1-x}\text{In}_x\text{As}$ heterostructures the (200) TEM dark field measurement is based on the intensity of the (200) electron SF [9]. For sufficiently thin specimens and off-zone axis imaging conditions, this intensity is proportional to the modulus square of the SF. If we neglect static atomic displacements, strain effects and temperature effects, this intensity can be written as the modulus square of the 'average' {002} SF. Any deviation from this dependence could result from one or more of these effects. Because of tetragonal distortion the (200) and (002) spots become inequivalent. Typical profiles of the modulus square of the (002) and (200) SF across a MQW are depicted in Fig. 5 together with the prediction of Eq. (2). Note that the deviations from a constant value within the InGaAs MQWs are caused by the statistical fluctuations of the In-concentration. The 'average' SF deviates slightly from the calculations. The figure shows the GaAs value at 0.04 nm², an interface region of about 1 nm thick for large x , where the SF is lower than that of the GaAs region, and the well region in which the intensity is found to be lower than that of the barrier for the 20% In systems. The low intensity interface region is often seen in quantum wells of $\text{Ga}_{1-x}\text{In}_x\text{As}$ and dilute nitrogen $\text{Ga}_{1-x}\text{In}_x\text{As}_{1-y}\text{N}_y$ [10-12] and has been attributed to the lower In content at these interfaces. Our calculations confirm this.

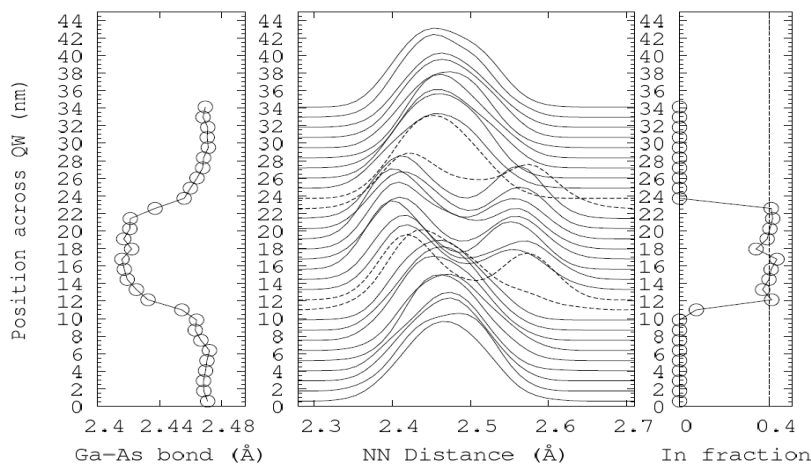


Figure 4. Left: Variation of the Ga-As bond length across the well of a multi-well $\text{Ga}_{1-x}\text{In}_x\text{As}$ system: Nearest neighbour pair correlation showing the bimodal bond length distribution within the well. Ga-As bond length shows up at about 2.4 Å and that of In-As at about 2.6 Å. Right: Variation of In content for the 40% In containing $\text{Ga}_{1-x}\text{In}_x\text{As}$ quantum well.

The quantum wells considered in this work are highly strained, the strain level being dependent on the size of both the QW and the barrier. In a recent work we showed that as the well size increases beyond 5 nm for the In contents considered in this work, continuum elasticity theory fails to explain the nature

of the strained state of the QWs [13]. Such large wells were shown to be characterized by a strained interface layer of about 2-4 monolayers thick and a strain-relaxed interior. We have analysed the effect of the strain on the (002) structure factor. For the large QWs, in which strain relaxation occurs, the real and imaginary parts of the (002) SF are found to be sensitive to the strain level even though the modulus square is only weakly sensitive as Figs. 5 shows.

The dependence of the square modulus of the (002) vs. (002) structure factors on the In fraction and well size is shown in Fig. 6. While the well sizes vary from 12 monolayers (MLs) to 40 MLs, the sum of the well and the barrier sizes is fixed at 120 MLs (about 34 nm). Comparison with the prediction of Eq. (2) shows that for thin layers the use of this expression for In content quantification based on the (002) spot can lead to an overestimation of the In fraction especially for high In containing QWs meanwhile the use of the (200) spot may lead to a slight underestimation of this fraction. The reason for the deviation of the (200) spot from the 'average' profile is the fact that the in-plane lattice parameter is constrained at the GaAs bulk value and the MASAs used for the (200) SF calculation are those obtained at this lattice parameter. Strain and/or static atomic displacements are responsible for the deviation of the (002) intensities from the average values.

5. Summary

We have computed modified atomic scattering amplitudes of $\text{Ga}_{1-x}\text{In}_x\text{As}$, $\text{GaAs}_{1-x}\text{N}_x$ and $\text{InAs}_{1-x}\text{N}_x$ ternary systems. For small scattering vectors ($s < 0.3 \text{ \AA}^{-1}$), major deviations from Hartree-Fock calculations are recorded in addition to remarkable chemical environment sensitivity. For such vectors orientation sensitivity of the reflection spots are highlighted and atomic relaxation is also found to increase the MASA. These findings call for caution to be taken when using various sets of MASAs for quantification purposes. We have investigated the effect of well size on the (002) and (200) structure factors of multi quantum wells using the Metropolis Monte Carlo approach. The (002) and (200) SFs of the MQWs differ from each other and both show deviation from the 'average' values. We have found that for thin layers, the 'average' intensity underestimates the (002) SF. This means that quantifications of the In content exploiting the (002) spot will overestimate the In fraction in thin GaInAs MQWs.

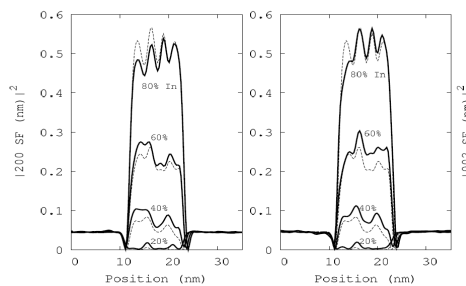


Figure 5. Variation of the modulus square of the (200) and (002) SFs of the MQW along the growth direction.

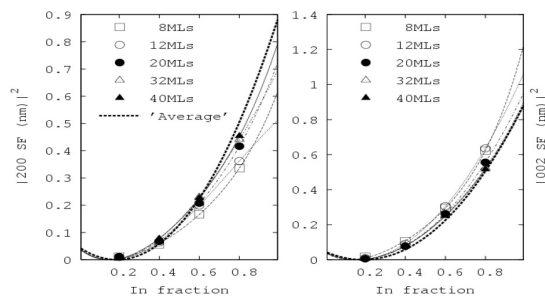


Figure 6. Variation of the square modulus of the (200) and (002) SFs with In content for various well sizes ranging from 12 to 40 MLs.

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References

- [1] Rosenauer A, Fischer U, Gerthsen D and Forster A 1988 *Ultramicroscopy* **72**, 121
- [2] Doyle PA and Turner PS 1968 *Acta Cryst. A* **24**, 390
- [3] Rosenauer A, Schowalter M, Glas F and Lamoen D 2005 *Phys. Rev. B* **72**, 085326
- [4] Glas F 2005 *Philos. Mag.* **11**, 2055
- [5] Volz K, Rubel O, Torunski T, Baranovskii SD and Stolz W 2006 *Appl. Phys. Lett.* **88**, 081910

- [6] Blaha P, Schwarz K, Madsen GKH, Kvasnicka D and Luitz J: *WIEN2k, An Augmented Plane Wave + Local Orbitals Program for Calculating Crystal Properties* (Karlheinz Schwarz, Techn. Universität Wien, Austria), 2001 ISBN 3-9501031-1-2.
- [7] Tersoff J 1986 *Phys. Rev. Lett.* **56**, 632
- [8] Titantah JT, Lamoen D, Schowalter M and Rosenauer A 2007 *J. Appl. Phys.* **101**, 123508
- [9] Bithell EG and Stobbs WM 1989 *Philos. Mag. A* **60**, 39
- [10] Herrera M, González D, Hopkinson M, Gutiérrez M, Navaretti P, Liu HY and Garcíá R 2005 *J. Appl. Phys.* **97**, 073705
- [11] Largeau L, Bondoux C and Patriarche G 2001 *Appl. Phys. Lett.* **79**, 1795
- [12] Luna E, Trampert A, Pavelescu EM and Pessa M 2007 *New Journal Phys.* **9**, 405
- [13] Titantah JT, Lamoen D, Schowalter M and Rosenauer A 2008 *Phys. Rev. B* **78**, 165326