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New Challenges for Delta-Like Confinement of Impurities in GaAs

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We review some aspects of delta-like confinement of isolectronic and dopant impurities in (001) GaAs. The nucleation, the redistribution and the relaxation are the important processes for the structural perfection and hence for the resulting electronic properties for both the generation of quantum dots and wires by selforganized MBE growth and the high-density incorporation up to one monolayer. In detail we discuss the fabrication and properties of InAs quantum dots and wires of Si doping wires in GaAs, and of Si-delta-doped GaAs with highly improved free-carrier density. The most important issue of our experiments is that the impurities species have to be deposited not continuously but in pulses in order to ensure ordered incorporation on lattice sites along step edges.

1. INTRODUCTION

The confinement of the distribution of impurity atoms in semiconductors to one lattice plane, called delta-doping, leads to a variety of novel electronic phenomena, depending on the nature of the impurity (i. e. isoelectronic or dopant species) and on its density (i. e. small fraction of or complete lattice plane). The ongoing challenge is (i) to confine the impurity distribution to one lattice plane, (ii) to control the lateral distribution of the impurity atoms within the lattice plane, and (iii) to increase the density of the impurity atoms to a completed lattice plane. We review recent progress in laterally ordered impurity incorporation to create semiconductor quantum wire (QWR) and quantum dot (QD) structures and in enhancing the fraction of lattice sites occupied by impurity species towards the limit of one monolayer. We use GaAs as prototype host lattice and we employ elemental-source MBE. The discussion will concentrate on In isoelectronic impurity sheets in GaAs on Si delta-doping of GaAs and as the most thoroughly studied dopant impurity.

2. PROBLEMS OF MONOLAYER IMPURITY INCORPORATION IN GaAs

The impurity species confined to one lattice plane have atomic radii that differ from those of the GaAs host lattice, hence elastic strain in the vicinity of their location is generated. Three distinct MBE growth sequences are required to obtain a delta-like confinement of impurities in GaAs [1]. First, a buffer layer has to generate an atomically flat starting surface (terrace). Second, the flux of the constituents of the host material is interrupted and the growth surface is exposed to the impurity flux. Changes in surface reconstruction occuring during impurity deposition can be detected by RHEED [2]. The third growth stage involves termination of the impurity flux and restoration of the matrix flux so that the impurity atoms are overgrown. Ideally the arrangement of the surface with the impurity atoms on lattice sites should not change during overgrowth. However, very often diffusion and surface segregation of the impurity atoms occur, the extent of which depends on impurity species, impurity density and growth conditions [3]. The effect of segregation causing an asymmetric broadening of the impurity distribution has for long prevented the formation of perfectly confined impurity sheets in the host lattice. The redistribution of the impurity atoms during overgrowth can furthermore result in a compensation of the actual free-carrier concentration by atomic exchange on either donor or acceptor sites [4].

3. ISOLECTRONIC In IMPURITIES IN GaAs CONFINED TO ONE (001) LATTICE PLANE

After successful preparation of the well-ordered and atomically flat starting (001) surface (terrace), the crucial point in the second stage is the suppression of 3D growth during deposition of the impurity layer [5, 6]. The key parameters are deposition of the impurity species in pulses interrupted by annealing periods and well-adjusted modulations of the substrate temperature. The growth interruption allows the In atoms to arrange on the appropriate lattice sites, and the temperature modulations minimize adverse effects on the morphology of the growth front, in particular during overgrowth with GaAs. During overgrowth of InAs with GaAs a considerable In fraction segregates, forming a "floating" layer on the GaAs growth surface, which is gradually dissolved in the overgrowing GaAs, thus preventing the formation of a well defined abrupt InAs/GaAs interface [5]. Neglecting this segregation has led to erroneous relations between the energy shift of the excitonic luminescence and the width of the InAs insertion. We have developed two strategies to minimize the adverse effect of In segregation at the GaAs-on-InAs interface, (i) the flash-off of the In floating layer prior to continuous GaAs overgrowth [5], and (ii) the suppression of any In segregation by a high As_4 overpressure [6]. In both methods the structural perfection of the GaAs overlayer is crucial for the electronic properties of the InAs monolayer in the GaAs matrix.

The applied growth procedures result in a high degree of structural perfection of strained InAs insertions in the GaAs matrix, as evidenced by high-resolution X-ray diffraction (XRD) [7] and electron microscopy (HREM) [8]. The strain is entirely accomodated by the elastic tetragonal distortion of the InAs unit cell. The critical thickness of homogeneously strained (001) InAs in the GaAs matrix is 2.6 ML. Beyond 2.6 ML a partial relaxation of the InAs film through 3D growth and the formation of misfit dislocations begins [5]. In order to get more quantitative information on the actual distribution of the In atoms within the respective (001) lattice planes of the GaAs matrix, we have also undertaken X-ray standing wave experiments [9]. In terms of the electronic properties the isoelectronic (001) InAs lattice planes in the GaAs matrix act as a normal quantum well despite the fact that the binding energy as well as the spatial extent of the localized excitons are bulk-like. The optical response of the systems is determined by the 2D nature of the plane potential [10].

When the InAs coverage of the (001) plane is reduced, coherent InAs islands of monolayer heights can form InAs quantum dots (QD) or quantum wires (QWR) in the GaAs matrix. The formation of periodic QD arrays can be promoted by the deposition at the step edges of terraced (001) GaAs surfaces [11]. The specific terrace configuration of a vicinal (001) GaAs surface tilted towards [100] consists of two staircases with steps running along the [110] and [$1\bar{1}0$] directions. This regular terrace distribution evolves during MBE using the step-flow growth mode and can be monitored by RHEED. The InAs QD structures of the present study comprising 10 periods of fractional (0.3 ML) InAs monolayers separated by 100 ML GaAs in [001] direction were grown by the flash-off technique.

In samples misoriented by 3.2° and 6.4° towards [100] the average In coverage of the (001) terraces is 0.3 ML, as revealed by high-resolution XRD [11]. The mismatch of 7.26 % is totally accomodated by biaxial elastic compression in the (001) plane. In the 3.2°-misoriented sample the lateral periodicity of the InAs QD array in the (001) plane was found to be 110 Å compared to a value of 100 Å deduced from the misorientation angle [12]. Lattice images obtained by HREM and image simulations of the InAs insertions in the GaAs matrix confirmed the existence of isolated InAs cluster with a periodicity, which coincides with the mean terrace width derived from the tilt angle. The interfaces between InAs and GaAs are free from any defects [11]. In terms of electronic properties the existence of isolated InAs QD in the crystalline GaAs matrix manifests itself in the distinct optical properties [11]. Here, a genuine zero-dimensional (0D) system in the sense that the center-of-mass motion of the exciton is localized by the InAs cluster was realized. The spontaneous emission is governed by a decay rate characteristic of atomic dipole transitions.

A wire-like alignment of elastically strained InAs insertion in the GaAs matrix was achieved on (110) GaAs [13]. This orientation has a strong tendency for step bunching during epitaxial growth. Using the method of arsenic overpressure to suppress In segregation we were able to fabricate InAs QWR arrays with a period of 300 Å in GaAs. The striking feature of these (110) InAs QWR arrays is the existence of pronounced lateral piezoelectric fields which give rise to localized electric fields in the GaAs matrix.

4. DOPING IMPURITIES IN GaAs CONFINED TO ONE (001) LATTICE PLANE

In addition to the problem of broadening of the impurity profile, the mechanism of saturation of the free-carrier concentration is an important issue in the case of Si-delta-doping of GaAs. The monotonic relation between electron concentration, as measured by the Hall effect, and dopant density, as deduced from the impurity flux and measured by SIMS, saturates below 10^{13} cm^{-2}. Several factors contribute to the observed saturation behavior: (i) decrease of Si_{Ga} donors, (ii) compensation by Si_{As} acceptors or Si-X acceptor complexes, and (iii) Si_{Ga}-Si_{As} nearest neighbour pair formation [4]. In addition, the effect of DX centers has to be taken into account. Several models were proposed to account for the observed asymmetric broadening of the impurity distribution, including classical surface segregation [14], solubility limited segregation [15], and Fermi-level induced segregation [16].

Based on our finding that Si nucleation on (001) GaAs takes place via the formation of Si nanoclusters [17], we developed a concept to control the site occupancy of the Si atoms even at coverages approaching 1 ML [18]. This concept is based on the ordered incorporation of Si along step edges of vicinal (001) GaAs. The most important improvement is that Si is deposited not continously but in pulses interrupted by annealing periods. This allows the Si atoms to migrate to the step edges and to form ordered stripes, as monitored by RHEED. The ordered impurity arrangement is then capped by GaAs using a lower growth temperature in order not to deteriorate the impurity ordering on the (001) terraces.

We first discuss the fabrication of Si doping wires in GaAs [19]. The laterally ordered incorporation of impurity sheets along step edges requires well ordered vicinal surfaces with straight ledge profiles and sufficient migration lengths of the impurity atoms on the terraces. We have used (001) GaAs tilted by 2° towards the [110] direction [≙ (111) Ga plane] as starting plane which produces 80 Å wide terraces. At the substrate temperature $T_{crit.}$ the transition from a 2D nucleation and step propagation mechanism to the desired step flow mechanism occurs which is evidenced by the change from an oscillating to a constant RHEED intensity. The existence of well-ordered step arrays on the vicinal (001) GaAs manifests itself in a clear splitting of the RHEED streaks into inclined slashes. During Si deposition in pulses of 90 s duration and 180 s interruption using a flux of $1\text{x}10^{11}$ $cm^{-2}s^{-1}$ (≙ $1.7\text{x}10^{-4}$ ML s^{-1}), the RHEED intensity decreases nonlinearly with the Si coverage and exhibits a certain recovery when the Si flux is interrupted. When reaching a coverage of 0.041 ML the intensity increases and a (3x2) reconstruction develops. The twofold periodicity in [110] direction is due to the formation of areas with dimerized Si atoms on Ga sites. The threefold periodicity in [$\bar{1}10$] direction is produced by an ordered array of Si dimers in (3x2) units which consist of two Si dimers and one missing dimer per unit mesh. The fractional-order spots in the RHEED pattern thus appear after a certain completion of (3x2) units along the step edge and the attachment of (3x2) units in a second row.

Our conclusion of a wire-like incorporation of the Si atoms at step edges is thus based on the following observations. First, the RHEED intensity during Si deposition decreases nonlinearly. Second, the Si induced reconstruction and the final intensity increase appear at a coverage which is correlated with the number of sites at the step edge. The arrangement of migrating Si atoms as dimers at the step edges generates kinks. The kink density increases sublinearly with the coverage because with increasing density a rising number of dimers become direct neighbours. This accounts for the observed sublinear intensity decrease. In addition, strain effects are important. The completion of a stripe of (3x2) units along the step edge and the attachment of (3x2) units in a second stripe leads to the threefold and twofold periodicity observed in the $[\bar{1}10]$ and the [110] direction, respectively. The abrupt change in reconstruction and strain accompanying this process accounts for the final intensity increase. The RHEED intensity behavior during Si deposition becomes very different when the terraces on the vicinal (001)GaAs surface are not smooth or when the terrace width becomes larger than the Si migration length (i. e. at small misorientation angle or for Si deposition without flux interruption) [19].

After the wire-like incorporation of the impurity atoms along step edges this ordered arrangement has to be stabilized during overgrowth with the GaAs host material when the impurity atoms are re-bonded to become part of a 3d solid. Several tens of lattice planes of GaAs have to be deposited at temperatures below T_{crit} so that the growth front morphology of the stepped surface is not adversely affected. Under these conditions the Si segregation is reduced. The conclusion of a selforganization of Si incorporation along step edges drawn from the RHEED studies is confirmed by results obtained by Raman scattering [20]. The wire-like Si incorporation in the 2° towards (111)Ga misoriented sample induces a distinct polarization asymmetry in the Raman scattering intensity of collective intersubband plasmon-phonon modes arising from the δ-doping layer.

We now turn to the fabrication of high-density Si doping sheets in GaAs which relies on the ordered incorporation of the impurity atoms along step edges during pulsed deposition [18]. Our results of conductivity and Hall effect measurements on delta-doped GaAs:Si layers with 2D impurity concentrations ranging from $1x10^{13}$ to $4x10^{14}$ cm^{-2}, which corresponds to a Si coverage of 0.017 to 0.61 ML, when we assume all Si atoms to be confined to one (001) Ga plane with $6.26x10^{14}$ atoms cm^{-2} clearly show that the pulsed delta-doping technique produces much higher free-carrier concentrations than the continuous delta-doping method. In contrast to the results obtained with continuous delta-doping at substrate temperatures $400 < T_s < 590$ °C [3, 4], there is no rapid decrease in carrier density beyond 10^{13} cm^{-2}. A slight reduction of the fraction of Si atoms incorporated as donors is observed beyond $5x10^{13}$ atoms cm^{-2}. The maximum free-carrier density of $8x10^{13}$ cm^{-2} achieved with $4x10^{14}$ Si atoms cm^{-2} is about one order of magnitude higher than with conventional delta-doping.

Deeper insight into the incorporation mechanism leading to these excellent results is obtained from the concomittant RHEED studies. For a Si coverage up to 0.12 ML (≙ $7.5x10^{13}$ atoms cm^{-2}) the change in reconstruction as well as the appearance of a (local) intensity minimum after reaching a certain Si coverage and the intensity recovery behavior after growth interruption indicate the ordering process of the Si atoms on the terraces and their arrangement as dimers in (3x2) units along the step edges. The intensity increase after the minimum is due to an abrupt change in strain and in the structure factor after completion of the first stripe of (3x2) units. Our assumption of confinement of the attached Si atoms in the (001)Ga plane is consistent with the high donor activity of 0.8 found in this concentration range. When we increase the Si coverage above 0.12 ML the specular beam intensity decreases continuously due to a deterioration of the (3x2) structure. A clear decrease of the half-order spot intensity occurs at Si coverages above 0.2 ML. Finally, facetting and third-order spots are observed above 0.6 ML Si, which indicates the incorporation of larger quantities of Si atoms in a second layer, i. e. in the As plane. The 90° rotation of the dangling bond direction when going from the Ga to the As plane accounts for the transition from the (3x2) to a distorted (1x3) structure.

In marked contrast to these findings for pulsed delta-doping, asymmetric third-order spots in the RHEED pattern develop already at a Si coverage of 0.2 ML for continuous delta-doping at 590 °C [18]. This clearly indicates the onset of Si incorporation on As sites at much lower coverage in this growth mode which is directly reflected in the differences of the electrical properties. Whereas for continuous deposition of $8x10^{13}$ Si atoms cm^{-2} a donor activation of only 0.06 is found, this value increases by a factor of 10 through pulsed Si deposition. As a result, the Si incorporation mechanism in delta-doping changes dramatically when the dopant atoms are supplied not continuously but in pulses to the growth surface. The ordered incorporation of Si at 590 °C monitored by RHEED yields a sheet donor concentration as high as $8x10^{13}$ cm^{-2} with the supply of $4x10^{14}$ Si atoms cm^{-2}. The observed Hall mobility of 1000 cm^2 V^{-1} s^{-1} evidences the structural perfection of these GaAs-Si-GaAs heterostructures, which was confirmed by X-ray diffraction experiments.

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