MANIFESTATION OF SPATIAL ORDERING OF QUANTUM DOTS IN MULTILAYERED SiGe NANOSTRUCTURES IN X-RAY DIFFRACTION PATTERNS

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By analyzing the intensity distribution of diffusely scattered X-rays in the reciprocal space, information concerning a transformation of two-dimensional structures in multilayered SiGe specimens into three-dimensional ones has been obtained. Weakly correlated quantum dots which contribute to the formation of lateral satellites have been shown to affect insignificantly the formation of a coherent satellite structure. Nevertheless, their influence on the distribution of deformation fields in superlattice layers remains substantial. It has been illustrated by applying the method of two-dimensional reciprocal space mapping of the X-ray intensity distribution to periodic Si/SiGe superlattices with different thicknesses of Ge layers (4 and 7 monolayers (ML)) and to periodically distributed SiGe dots buried in silicon.

1. Introduction

A self-induced structure involving a plenty of two-dimensional small islands (quantum dots) can emerge in a stressed epitaxial superlattice on the growth surface of a layer provided the Stranski—Krastanow growth mode [1]. As the number of superlattice layers increases, the size distribution of dots becomes more homogeneous, and the ordering of dots improves [2,3]. Quantum dots form a two-dimensional almost periodical set, the axes of which coincide with the directions of the minimal value of the elastic modulus of a crystal; for example, these are {100} directions for the diamond structure.

A self-induced epitaxial growth of stressed semiconductor nanostructures has been the subject of extensive researches for the last decade. Among those systems, self-induced Ge-rich islands buried in silicon are of special interest because they give rise to the very effective photoluminescence. We note that the stress fields in multilayers promote the vertical ordering (stacking) of islands, while the horizontal (lateral) ordering of the latter is manifested much weaker. The positions of dots at the neighbor interfaces are vertically correlated, and their vertical spatial period is equal to the superlattice constant [4].

To study correlation phenomena, we should select such diffraction methods where correlations between the positions of self-induced quantum dots result in the maxima of the spectral density in the reciprocal space. Lateral correlations between dots' positions are responsible for a number of the intensity maxima in the reciprocal space which are located in parallel to the direction q_x along the specimen's surface (below, we use notations q_x and q_z for the coordinates of the reciprocal space). The shape functions of the lateral maxima around the satellites of the first, SL_1, and zero order, SL_0, of the basic superlattice bring us to some preliminary conclusion concering a stressed state of the system. A horizontal distribution of the intensity around SL_0 with a maximum shifted towards larger q_x reveals

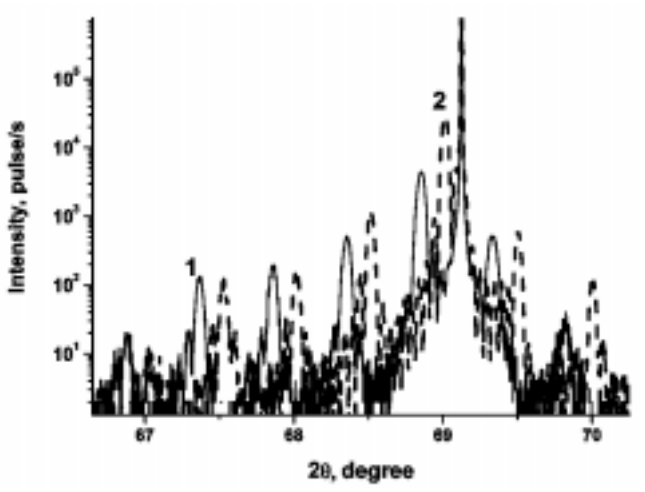


Fig. 1. The curves of reflection 004 ($\omega-2\theta$ scans) from superlattices with (*1*) a 7-ML (with quantum dots) and (*2*) 4-ML germanium layers

the regions stretched and squeezed, mainly, by the silicon matrix, whereas the SL_1 satellites with a maximum shifted towards smaller q_x correspond to elastic relaxation inside Ge-rich regions (dots).

Knowing the distance between those maxima, one can determine the average distance between dots

$$\langle L\rangle = \frac{2\pi}{\Delta q_x}. \tag{1}$$

Two models of the dot arrangement can be used for estimating the structure parameters. In the short-range order (SRO) model, the distance between adjacent dots is random and not correlated. Then, the width of satellites δq_x depends on q_x:

$$\delta\, q_x = \frac{\left(\sigma_{\|} q_x\right)^2}{\langle L\rangle}. \tag{2}$$

Using this dependence, one can determine the mean-square deviation $\sigma_{\|}$ of the distance between dots. In the long-range order (LRO) correlation model, the width of satellites does not depend on q_x, being reciprocally proportional to the size $D_{\|}$ of the lateral coherent regions:

$$\delta\, q_x = \frac{2\,\pi}{D_{\|}}. \tag{3}$$

The position and width of lateral satellites can be measured at any point of the reciprocal lattice.

This work aimed at studying the processes of formation of quantum dots in superlattice structures and at determining the main parameters of dots and the influence of these parameters on the X -ray diffraction spectra, in particular, on the variation of the period of coherent satellites.

2. Experimental Procedure

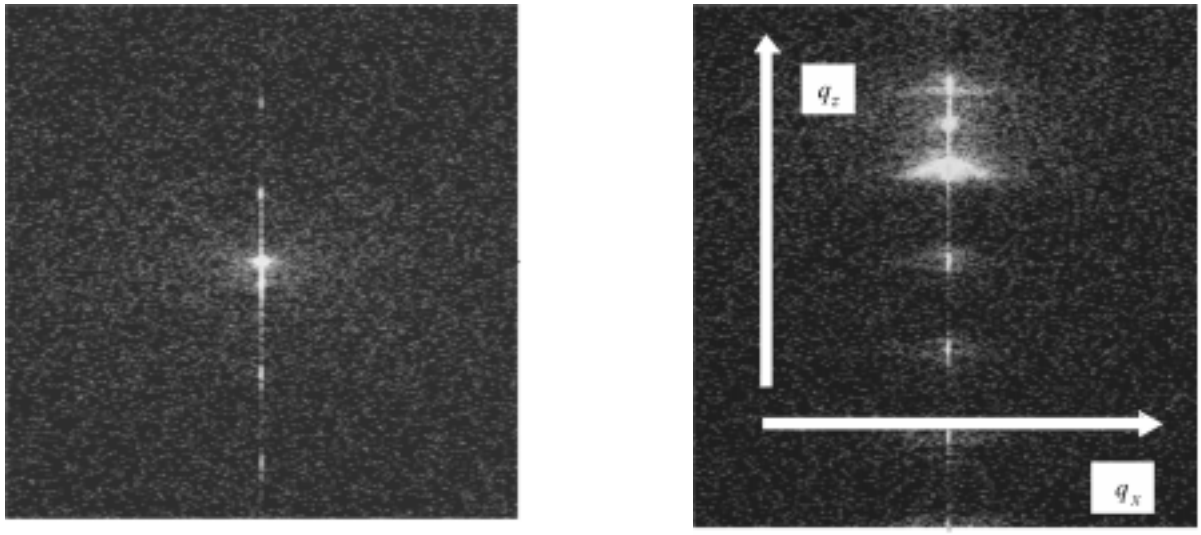
We study 10-period superlattice Si/Ge structures with the thickness of the germanium layer of 4 and 7 ML, which were grown on a Si(100) substrate in a MBE installation. Technological parameters of the specimens are given in the table. The measurement of the Bragg reflection curves for symmetric 400 and asymmetric 224 reflections were carried out using a Philips MRD X-Pert diffractometer. The specimens were scanned within the range of 3° about the exact Bragg angle in the so-called $\omega - 2\theta$ mode. Both the ω- (without an analyzer) and $\omega-2\theta$-scans were registered. In the same mode, the two-dimensional maps of the intensity distribution around the 004 and 224 points of the reciprocal lattice were recorded. The thickness of layers in the multilayered structure was determined with the use of oscillations of the Bragg reflection curves [5].

3. Experimental Results and Their Discussion

The Bragg reflection curves (Fig. 1) recorded in the $\omega - 2\theta$ mode demonstrate a clear picture of both the positive and negative superlattice satellite peaks. The identical periods of satellite intensity oscillations for two structures with different thicknesses of the Ge layer may testify to that these structures are modulated by the wetting and buffer layers with the identical thickness. It is known [1] that if the thickness of a Ge layer exceeds 4 ML, the formation of islands (quantum dots) in the Stranski—Krastanow growth mode begins, so that an excess of Ge (above 4 ML) is spent on their formation. The results presented in Fig. 1 evidence for that the islands do not participate in modulating the superlattice structure. However, as seen from Fig. 1, they influence the distribution of the deformation fields in it.

On the basis of the distance between the peak of the substrate and the zeroth satellite of the superlattice, we determined the average strain over the period (see the table). It is different for different researched structures. Therefore, it was very important to answer the question what is the role of islands in the formation of a superlattice and to obtain information concerning their parameters and the degree of ordering.

The most informative method for researching such structures is to obtain the two-dimensional maps of the



a **b**

Fig. 2. Reciprocal space distribution maps of the diffracted intensity around a symmetric 004 point in the SiGe/Si structures with 4- (*a*) and 7-ML (*b*) Ge layers. $CuK_{\alpha 1}$ emission

X-ray intensity distribution around points of the reciprocal lattice [6, 7]. Fig. 2 shows the patterns of the intensity distribution map for symmetric reflection 004. Scattering by the substrate, the superlattice-induced satellite structure, and the diffuse scattering peaks caused by the ordered structure of quantum dots are clearly distinguished on these maps. Information concerning Ge islands is presented by the diffuse maxima, which accompany the coherent satellite. The diffuse maxima are symmetric with respect to the q_x axis in the map patterns of the symmetric 004 reflection, being however obviously asymmetric in asymmetric patterns (Fig. 3). The lateral diffuse maxima practically do not appear in the case of symmetric diffraction, most likely, owing to a smaller area of the crystal surface that makes a contribution to diffraction, as compared with asymmetric geometry [8, 9].

Therefore, the more detailed information on the distribution of deformation fields in structures with quantum dots can be obtained from the analysis of asymmetric two-dimensional maps of x-ray scattering in the reciprocal space. Let us analyze certain cross-sections of those two-dimensional maps of the intensity distribution, which are parallel to the structure growth plane. Figure 4 shows the corresponding cross-sections near the superlattice satellites of the zeroth and first orders. Near the satellite of the zeroth order SL_0 (at small values of q), the diffusion scattering is determined mainly by the deformation region and by the scattering by those Si lattice regions, which are located far from the islands (quantum dots). The diffusion intensity maximum near SL_0 was shifted, as follows from Fig. 4, in the positive q_x direction, i.e. towards larger values of h_x, which corresponds to the squeezing deformation of the Si crystal lattice in the regions between dots. The squeezing deformation amounted to 5.062×10^{-4}.

Near the first-order satellite SL_1, the diffuse scattering was defined by the deformation and the scattering by the crystal lattices of islands. The maximum of the diffuse intensity near SL_1 was shifted towards smaller values of q_x, i.e. corresponded to a tension of the crystal lattice, which enlarged distances between dots. The average value of this deformation was 1.119×10^{-3}.

Parameters of studed structures

Specimen N	Technological thickness of the layers: Si(in nm)/Ge(in ML)	Exp. value of the superlattice constant, nm	Deformation along q_z averaged over the period	Deformation in a Si layer along q_x	Deformation in a quantum dot along q_x
1	20/7	21.7	$2.32 \cdot 10^{-3}$	$5.10 \cdot 10^{-4}$	$1.12 \cdot 10^{-3}$
2	20/4	21.63	$9.44 \cdot 10^{-4}$	—	—

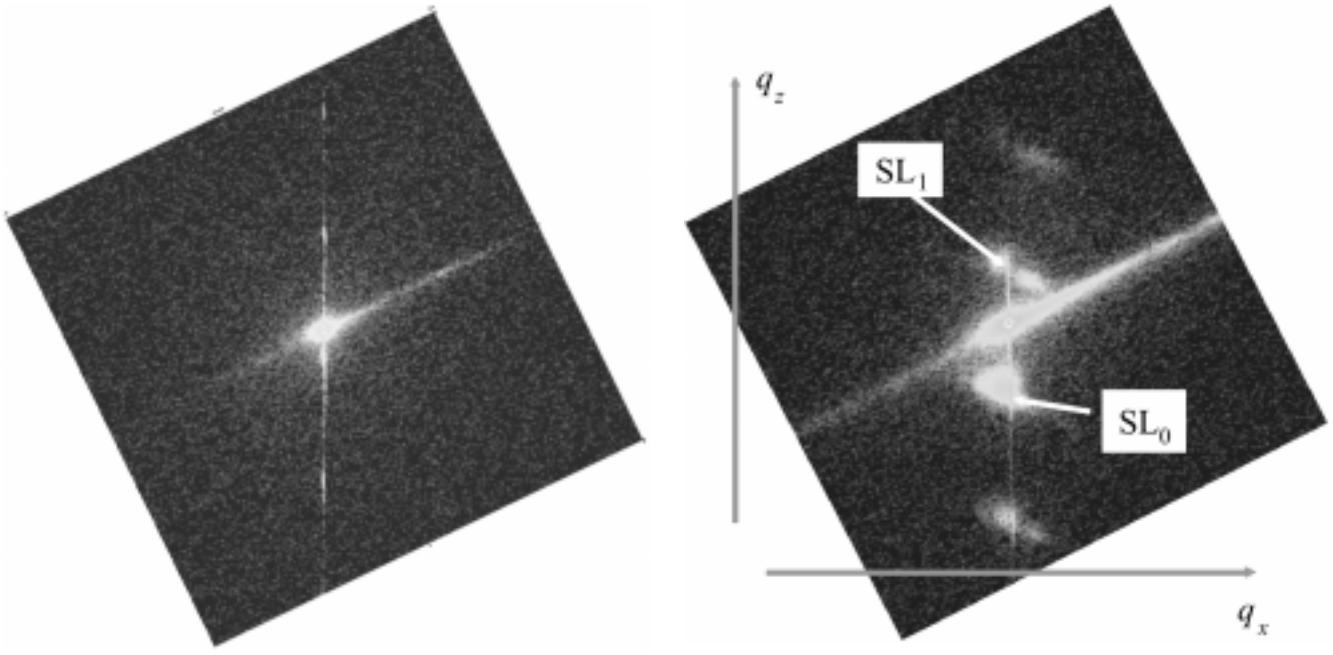


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Fig. 3. The same as in Fig. 2 but for point 224

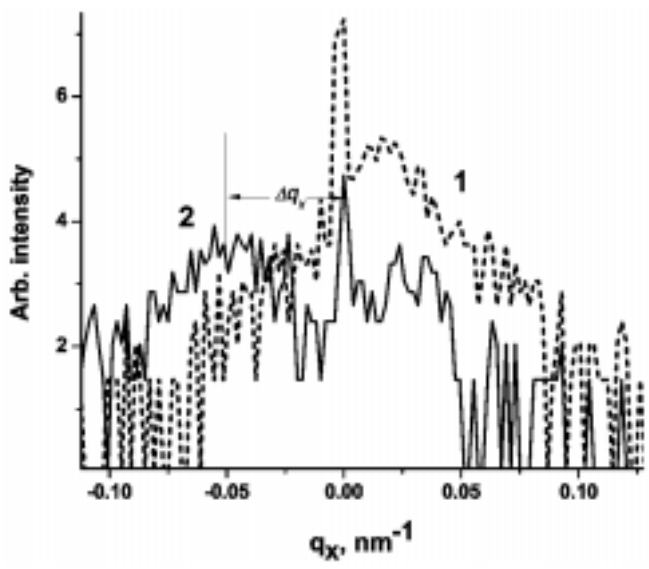

Fig. 4. Projections of the cross-sections of the diffuse scattering peaks around SL_0 (*1*) and SL_1 (*2*) satellites onto the q_x axis (ω scans) calculated from the reciprocal space map for a 10-period SiGe/Si structure around point 224

Knowing the distance Δq_x between lateral maxima, we determined the average distance $\langle L \rangle$ between quantum dots, which was equal in our case to 83.8 nm. The dimension of the dot regions appreciated by formula (3) was 67.5 nm.

Correlations in the lateral distribution of dots in the specimens were very weak. Therefore, only the lateral satellites of the first orders are observed in the x-ray scattering maps and are localized mainly only near coherent superlattice satellites of the zeroth and first orders. The superlattice period is formed only by the wetting layer t_1 and the silicon layer t_2. Poorly correlated dots make a small contribution to the formation of a superlattice structure of coherent lattices. This is manifested only in a redistribution of elastic strain fields, while the superlattice period $T = t_1 + t_2$ preserves its value. This yields that neither the presence of islands in the superlattice nor the dispersion of their heights suppresses the modulation.

4. Conclusions

It has been shown that if the crystal is scanned along the vector of diffraction and the thickness of the germanium layer exceeds 7 ML, then, besides the superlattice maxima, a set of satellites corresponding to quantum dots is observed. The manifestation of correlations in the arrangement of those maxima on the two-dimensional maps in the reciprocal space (in the lateral and normal directions) evidences for a three-dimensional ordering of quantum dots. This is also confirmed by the shape of ω-curves near the superstructural maxima. However,

it should be emphasized that phase correlations in the diffraction vector direction are stronger than those in the growth plane.

It has been established that, due to weaker correlations in the lateral direction, quantum dots do not participate in the formation of coherent superstructure maxima (satellites). It is evidenced for by identical values of the superlattice constants in the structures with the germanium layers of 4 and 7 ML in thickness.

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ПРОЯВ ПРОСТОРОВОГО УПОРЯДКУВАННЯ КВАНТОВИХ ОСТРІВЦІВ У БАГАТОШАРОВИХ НАНОСТРУКТУРАХ SiGe У РЕНТГЕНІВСЬКІЙ ДИФРАКЦІЇ

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Р е з ю м е

З аналізу розподілу інтенсивності дифузно розсіяних рентгенівських променів в оберненому просторі отримано інформацію про перехід від 2D- до 3D-структур у багатошарових зразках SiGe. Показано, що слабоскорельовані квантові точки, що дають внесок у формування латеральних сателітів, мало впливають на формування когерентної сателітної структури. При цьому істотним залишається їх вплив на розподіл полів деформацій у шарах надгратки. Це проілюстровано застосуванням методу двовимірних карт розподілу інтенсивності навколо вузла оберненої гратки до періодичних Si/SiGe-надграток з різною товщиною шару германію (4 або 7 моношарів (МШ)), а також до періодичних SiGe-точок, вбудованих у кремній.