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High-resolution x-ray diffraction from multilayered self-assembled Ge dots

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We have studied multilayers of self-assembled Ge-rich dots embedded in silicon grown by molecular-beam epitaxy using high resolution x-ray reciprocal space mapping and reflectivity. The Si spacer thicknesses between the dot arrays were in the range of 10–40 nm, the typical dot size was about 150 nm for the diameter and 7 nm for the height. The measured reciprocal space maps were simulated using statistical kinematical x-ray-diffraction theory, and a good agreement between experimental and simulated data has been achieved. From the measurements, the in-plane strain in the dot lattice was determined. We derived the degree of the vertical correlation of the dot positions (“stacking”) and a lateral ordering of the dots in a square array with main axes parallel to the $\langle 100 \rangle$ directions, with an array lattice constant of about 500 nm. [S0163-1829(97)05623-3]

I. INTRODUCTION

Silicon-based heterostructures like Si/SiGe have attracted a great deal of attention in the last decade, since they offer the possibility of confining either electrons or holes in a material system which is highly compatible with Si technology.¹ The properties of two-dimensional electron and hole gases in Si/SiGe have been studied thoroughly, and are used in semiconductor devices.^{2,3} Recently, using lateral patterning techniques, Si-based structures were also realized in which the carriers are confined in two (quantum wires) (Refs. 4 and 5) or in three dimensions (quantum dots), with the motivation of achieving superior electronic and optoelectronic properties.

A large lattice mismatch offers an interesting approach for the fabrication of quantum dots, taking advantage of self-assembling mechanisms.^{6–9} Under suitable growth conditions, elastically strained Ge layers do not relax plastically via the introduction of misfit dislocations, but an elastic relaxation takes place via the formation of strained Ge islands. Islands (dots) occur when the thickness of the epilayers exceeds a critical value and the growth mode changes from two-dimensional strained-layer growth (Frank–van der Merwe growth) to a three-dimensional one (Stranski–Krastanow growth).¹⁰ In the Si–Ge system, the transition of this crossover in the growth mode has been studied using photoluminescence and transmission electron microscopy (TEM),^{8,11–13} atomic force microscopy (AFM),¹³ and scanning tunneling microscopy.¹⁴ In contrast to reactive ion-etched lateral structures, these self-organized nanostructures do not exhibit surface damage.

The introduction of nanometer-scale regions with differ-

ent and appropriate chemical composition, i.e., Ge quantum dots in a Si matrix, provides high confinement energies up to several 100 meV, particularly for holes, and thus the prospect of room-temperature performance of devices like resonant-tunneling diodes.¹⁵ In the III–V material system, extremely low threshold lasers have been realized by the alternating growth of GaAs and strained InAs, which yields layers of InAs dots embedded in GaAs.^{16,17}

In order to achieve a high spatial density of dots, which is a requirement for any technological application, dot multilayers were grown. In such a multilayer, the layers containing Ge islands are overgrown with a Si spacer layer, and this sequence is repeated several times. As shown by Tersoff and co-workers,⁹ such multilayers exhibit interesting properties such as enhanced vertical and horizontal spatial correlation of the dot positions, and furthermore an improved size homogeneity as required for electronic devices. These favorable phenomena are driven by the strain fields associated with the buried islands.

Since the self-assemblance is mismatch driven, the structures are often very close to or even beyond the two-dimensional critical thickness, and the probability of the nucleation of dislocations in multilayers is considerable. Therefore, a structural characterization by a nondestructive technique like x-ray diffraction is desirable. Moreover, x-ray diffraction yields information on the strain status of dots and their vertical and horizontal correlations. Whereas reactive ion-etched lateral structures have been investigated thoroughly with respect to their structural properties,^{18,19} similar information on self-assembled dot structures is rather sparse²⁰ and no quantitative information was given. Previously, grazing-incidence diffraction and reflectivity experi-

TABLE I. Structural parameters of the samples.

Sample	A	B	C	D	Source
Spacer thickness D (Å)	100	200	300	400	nominal values
Dot distance L (Å)			6000 ± 800	5000 ± 500	diffraction
Distance dispersion σ_l (Å)			2000 ± 700	1500 ± 600	diffraction
Vert. correl. length ξ_{corr} (Å)	$500 \pm 200^*$	$1000 \pm 200^*$	$> 3000^\times$	$> 4000^\times$	*reflectivity, $^\times$ diffraction
In-plane strain ε_{xx}			$(1.4 \pm 0.1) \times 10^{-3}$	$(1.5 \pm 0.5) \times 10^{-3}$	diffraction
Elastic strength P	$(0.5 \pm 0.5) \times 10^{-3}$	$(0.5 \pm 0.5) \times 10^{-3}$	$(1.5 \pm 0.5) \times 10^{-3}$	$(0.5 \pm 0.5) \times 10^{-3}$	diffraction

ments on uncovered single-dot layers of Ge grown on (001)-oriented Si were reported by Williams *et al.*,²¹ with the intention of determining the critical thickness for coherent Ge islanding. Steinfort *et al.*²² studied the elastic relaxation in a single layer of Ge hut clusters using grazing incidence for the determination of the gradient of the in-plane strain.

In this paper, we report on high-resolution x-ray reciprocal space mapping and x-ray reflectivity measurements of self-assembled Ge quantum-dot multilayer structures, and analyze the coherent and diffuse scattering. Information on the strain distribution and on vertical and lateral ordering is obtained from a comparison of the experimental data with calculations based on statistical kinematical diffraction theory.^{23,24}

The structure of the paper is as follows: In Sec. II we outline the growth of the SiGe/Si dot multilayers. A short overview of the experimental results is presented in Sec. III. In Sec. IV, we formulate the theoretical description of x-ray scattering from the dot arrays using kinematical diffraction theory. In Sec. V, the in-plane strain fields are determined from a comparison between the experimental reciprocal space maps of diffuse scattering and the calculated ones. We determined the average distance between neighboring dots, and the degree of vertical correlation of the Ge dots in the multilayers was estimated.

II. EXPERIMENT

Multilayer samples were grown on (001)-oriented Si substrates using solid source molecular-beam epitaxy (MBE) in a Riber Siva-32 MBE at the Walter Schottky Institut. The optimum growth conditions for achieving a narrow size and height distribution of the Ge dots in the multilayers were determined previously^{13,25} by a systematic variation of the growth temperature, the growth rate, and the thickness of the deposited Ge layers. For the structural analysis, atomic force microscopy and transmission electron microscopy were used.²⁵ Based on the previous results, a growth temperature of 670 °C and a Ge-growth rate of 0.075 Å/s were chosen for the samples investigated in this paper. The samples contain $N = 19$ periods of nominally 5.5-ML Ge separated by Si spacers with thicknesses in the range of 10–40 nm. Basic structural parameters of the samples are summarized in Table I. From the cross-section TEM data,²⁵ it follows that the dot diameter as well as the dot height increase somewhat toward the free surface. Typical mean values are 160 nm for the dot diameter and 7 nm for the dot height. The Ge concentration in the dots was estimated from photoluminescence and Raman-scattering experiments,²⁶ and a value of 70% is used in the simulations.

X-ray-diffraction measurements were performed with a diffractometer equipped with a four-crystal Ge monochromator using a conventional x-ray tube and the Cu $K\alpha_1$ line. For the reciprocal space maps a two-crystal Ge(220) analyzer was used, with an angular resolution of 12 arcsec. For each sample, we measured reciprocal space maps in symmetrical 004 and asymmetrical 113, 224, and 404 diffraction geometries (low angles of incidence). In addition, we performed linear scans around 206 as well as specular and nonspecular reflectivity measurements (2θ -scans) at the Optics beamline of the synchrotron source ESRF, Grenoble.

III. EXPERIMENTAL RESULTS

In Fig. 1, we present a double-crystal 004 rocking curve (points) of sample C as well as a simulation using dynamical diffraction theory (line), which neglects the formation of Ge islands, but takes only the Ge wetting layer into account. The rocking curve reflects the laterally averaged superlattice structure of the sample. On the curve, superlattice satellites up to the eighth order are visible, which serve as a qualitative measure of the overall structural quality of the self-assembled quantum-dot sample. From the fitted curve a period of 29.4 nm and an average Ge-content of 2.04% are obtained, depending both on the structural and geometrical parameters of the wetting layer and the Ge dots and their coverage. Since the thickness of the wetting layer decreases toward the free surface, the measured superlattice satellites are broader than in the simulated curve.

In Figs. 2 and 3, we show the measured 004, 224, and 404 reciprocal space maps of two samples (C and D; see Table I)

FIG. 1. Measured (004) double-crystal rocking curve of sample C (points), and its dynamical simulation (line).

FIG. 2. Measured (left-hand side) and simulated (right-hand side) reciprocal space maps around the 004 (a), 224 (b), and 404 (c) reciprocal-lattice points for sample C. Only the diffuse parts of the reciprocal space maps are simulated. In (a), *S* denotes the substrate peak, and *SL*₀ and *SL*₁ are the superlattice satellites of the zeroth and first order, respectively. *A* denotes the analyzer streak.

FIG. 3. Analogous to Fig. 2 for sample D.

along with their numerical simulations. The increase in intensity between neighboring contours is $10^{1/4}$ in all the maps. The measured maps (Figs. 2 and 3, left-hand sides) exhibit sharp peaks at $q_x=0$ denoted S , SL_0 , and $SL_{\pm 1}$. The horizontal widths of these maxima are determined by the diffractometer resolution, and they represent the coherent contribution to the scattered intensity. The peak labeled S corresponds to the diffraction from the substrate, and SL_n is the n th satellite peak of the laterally averaged superlattice. The distance between the satellites equals $2\pi/D$ (D is the superlattice period), and the spacing between SL_0 and the substrate peak is proportional to the relative difference of the vertical lattice constant of the substrate and the averaged lattice constant of the superlattice. The streak labeled A (analyzer streak) is an artifact caused by the tails of the acceptance curve of the crystal analyzer.

IV. THEORETICAL DESCRIPTION

Self-assembled quantum dots (SQD's) give rise to diffuse x-ray scattering accompanying the coherent diffraction or reflection signal of x rays from the whole sample. The coherently scattered wave depends only on the laterally averaged sample structure. Performing this averaging, we replace the dot array by an effective layer, whose thickness equals the mean dot height, and whose scattering factor (proportional to the x-ray crystal polarizability) is an average of the scattering factors of the dot and the crystal matrix (weighted by the coverage of the dot array).

The diffuse scattering is caused by the difference in the scattering factors of Ge and Si and the elastic deformation field in the Si matrix surrounding the dots. In the case of x-ray reflection (i.e., when the scattering vector is very small), only the first mechanism contributes. If the scattering vector $\mathbf{K}_{\text{out}} - \mathbf{K}_{\text{in}}$ ($\mathbf{K}_{\text{in,out}}$ are the wave vectors of the incident and scattered beams, respectively) is close to a vector $\mathbf{h}$ of the reciprocal lattice, the scattered intensity is sensitive to the $\mathbf{h}$ th Fourier component of the crystal polarizability $\chi(\mathbf{r})$. In this case, the diffuse scattering mainly originates in the deformation field both outside and inside the dots.

The information on the dot structure is contained only in the diffuse component of the scattered intensity accompanying the coherent intensity. Therefore, we did not include the coherently scattered intensity in the simulations. In the following two subsections we first formulate a description of the strain fields and the statistical properties of a SQD ensemble, and then deal with the simulation of the diffuse x-ray scattering.

A. Morphology and strain fields

The starting point is the shape function $\Omega(\mathbf{r})$ of a single quantum dot, which is unity in the dot and zero outside. The $\mathbf{h}$ th Fourier component of the polarizability $\chi(\mathbf{r})$ of a SQD sample can be written as

$$\chi_h(\mathbf{r}) = \sum_j [\Omega(\mathbf{r} - \mathbf{R}_j) \chi_{hD} + (1 - \Omega(\mathbf{r} - \mathbf{R}_j)) \chi_{hL}],$$

where χ_{hD} and χ_{hL} are the polarizability components of the dot material and the surrounding matrix, respectively, and the positions of the dot centers are $\mathbf{R}_j$. The polarizability

χ_{hD} of the dot structure is a linear function of the chemical composition x of the $\text{Si}_{1-x}\text{Ge}_x$ dot lattice. Here we neglect the thin two-dimensional wetting layers, since they do not contribute to the diffuse x-ray scattering.

Because of the lattice mismatch between Si and Ge, the crystal lattices of the dots and their surroundings are deformed. The calculation of the displacement field in and around a dot is a complicated problem. In a rigorous treatment, one has to take into account the difference in the elastic constants inside and outside the dot, and the elastic anisotropy as well as the elastic relaxation of internal stresses at the free surface. For our purposes, it is suitable to neglect the anisotropy and the difference in the elastic constants, and we express the deformation field $\mathbf{t}(\mathbf{r}; Z)$ in the surrounding of a single dot (centered at $\mathbf{r} = \mathbf{0}$) as a superposition of the deformation fields $\mathbf{t}_0(\mathbf{r} - \mathbf{r}'; Z) d^3\mathbf{r}'$ of elementary point defects²⁷ in the interior points $\mathbf{r}'$:

$$\mathbf{t}(\mathbf{r}; Z) = \int d^3\mathbf{r}' \mathbf{t}_0(\mathbf{r} - \mathbf{r}'; Z) \Omega(\mathbf{r}'). \quad (1)$$

The deformation field $\mathbf{t}(\mathbf{r}; Z)$ depends not only on the relative position $\mathbf{r}$ of the observation point with respect to the dot center, but also on the depth Z of the dot center below the free surface (due to the elastic relaxation at the surface). The deformation field of an isotropic elementary point defect in an isotropic semi-infinite elastic continuum has been calculated previously.^{27,28} For our calculations, we will use the function $\varphi(\mathbf{r}; Z) = \mathbf{h} \cdot \mathbf{t}(\mathbf{r}; Z)$ and its Fourier transform

$$\varphi^{\text{FT}}(\mathbf{q}; Z) = \int d^3\mathbf{r} \varphi(\mathbf{r}; Z) e^{i\mathbf{q} \cdot \mathbf{r}}.$$

In the following, the superscript FT will denote the Fourier transform.

Using the results in Ref. 28 after some manipulation we obtain

$$\varphi^{\text{FT}}(\mathbf{q}; Z) = iP \left[\frac{\mathbf{h} \cdot \mathbf{q}}{q^2} - \Phi(\mathbf{q}) \exp(-q_{\parallel} Z - iq_z Z) \right] \Omega^{\text{FT}}(\mathbf{q}). \quad (2)$$

q_z is the vertical component and $q_{\parallel} = \sqrt{q_x^2 + q_y^2}$ the in-plane component of the reciprocal space vector $\mathbf{q}$. The real-space coordinates x and y are parallel to the sample surface, and they are parallel and perpendicular to the scattering plane, respectively. $\Omega^{\text{FT}}(\mathbf{q})$ is the Fourier transform of the dot shape function, and the function $\Phi(\mathbf{q})$ expresses the elastic relaxation at the free surface,²⁸

$$\Phi(\mathbf{q}) = \frac{1}{2} \left(\frac{1}{q_{\parallel} + iq_z} - \frac{3 - 4\nu}{q_{\parallel} - iq_z} \right) \left(\frac{\mathbf{h}_{\parallel} \cdot \mathbf{q}_{\parallel}}{q_{\parallel}} + ih_z \right) + \frac{q_{\parallel}}{(q_{\parallel} - iq)} \left(\frac{\mathbf{h}_{\parallel} \cdot \mathbf{q}_{\parallel}}{q_{\parallel}} - ih_z \right),$$

where $\nu = 0.22$ is the Poisson number.

P is the elastic "strength" of a dot, which expresses the difference of the lattice constants of the $\text{Si}_{1-x}\text{Ge}_x$ dot lattice and the surrounding Si matrix. If we denote the lattice mismatch of Ge and Si as $\delta = (a_{\text{Ge}} - a_{\text{Si}})/a_{\text{Si}}$, and x is the Ge content in the dots, P can be expressed as²⁴

$$P = x \delta \frac{1 + \nu}{1 - \nu}. \quad (3)$$

As we show in Sec. V, the elastic strength P following from Eq. (3) is unrealistically large, and a probable reason for this discrepancy will be discussed.

In deriving Eq. (2), we assumed that the contribution to the elastic relaxation, which depends on the depth below the surface, is equivalent for all points inside the dot. This is valid if the dot height is much smaller than the depth Z , and, therefore, it is not fulfilled for the dots lying in the topmost layers.

Numerical calculation^{27,29} of the deformation field in the dots confirmed that it is reasonable to assume a homogeneous deformation of the dot volume, i.e., the elements of the strain tensor $\hat{\epsilon}$ are constant. Nonhomogeneities of the strain occur only in a small region close to the dot surface. In our calculations, we defined the elements of the strain tensor $\hat{\epsilon}$ with respect to the unstrained Si matrix, thus $\epsilon_{xx} = 0$ corresponds to a fully pseudomorphic structure.

In our samples, the dots are situated on Ge wetting layers in the depths $Z_n = nD$ below the sample surface, where D is the superlattice period and $n = 0, \dots, N-1$. The lateral positions of the dots are described by the correlation function

$$w(\mathbf{R}, \mathbf{R}') = \langle c(\mathbf{R})c(\mathbf{R}') \rangle.$$

$c(\mathbf{R})$ equals unity if the point $\mathbf{R} = (\mathbf{X}, Z)$ is occupied by a dot center, and zero otherwise. The average $c = \langle c(\mathbf{R}) \rangle$ is the normalized dot density. We assume that the dot distribution is statistically homogeneous (i.e., the *statistical* properties are invariant under arbitrary translations parallel to the surface), so that the correlation function is

$$w(\mathbf{X}, Z_n; \mathbf{X}', Z_m) = w(\mathbf{X} - \mathbf{X}'; Z_n, Z_m) \equiv w_{nm}(\mathbf{X} - \mathbf{X}').$$

The Fourier transform of the correlation function is

$$\begin{aligned} w^{\text{FT}}(\mathbf{q}) &= \int d^3\mathbf{r} w(\mathbf{r}) e^{i\mathbf{q} \cdot \mathbf{r}} \\ &= \text{const} \sum_{n=1}^N \sum_{m=1}^N \exp[iq_z(Z_n - Z_m)] \\ &\quad \times \int d^2\mathbf{X} w_{nm}(\mathbf{X}) e^{i\mathbf{q}_{\parallel} \cdot \mathbf{X}}. \end{aligned}$$

The correlation function of the dot distribution in a depth Z_n and a particular direction can be obtained using the theory of stationary random processes.³⁰ If we denote $S(l)$ as the probability of finding a neighboring dot in the distance l , we obtain

$$w_{nn}^{\text{FT}}(q) = \int_{-\infty}^{\infty} dx w_{nn}(x) e^{iqx} = c \left[1 + 2 \text{Re} \left(\frac{S^{\text{FT}}(q)}{1 - S^{\text{FT}}(q)} \right) \right],$$

$q \neq 0.$

From this expression, it follows that $w_{nn}^{\text{FT}}(q)$ has a maximum when $S^{\text{FT}}(q)$ is close to unity. For $q=0$ this function exhibits a δ -like peak. For a nearly periodical distribution of the dots with an average dot spacing L , the characteristic function $S^{\text{FT}}(q)$ [i.e., the Fourier transform of $S(l)$] would be

approximately $\exp(iqL)$ and the maxima of $w_{nn}^{\text{FT}}(q)$ would have the coordinates $q_p = 2\pi p/L$, where p is an integer. The width of these maxima depends on the statistical dispersion σ_l of $S(l)$.

The dot distribution in two orthogonal directions is assumed statistically independent, thus

$$w_{nn}(\mathbf{X} - \mathbf{X}') = w_{nn}(X - X') w_{nn}(Y - Y').$$

We assume that the distributions of the dots in different depths is statistically equivalent, therefore w_{nn} becomes independent of n , and we denote w_{nn} as $w_{\parallel}$. In a real structure, this assumption is not completely fulfilled, since the in-plane correlation of the dot positions in a multilayer was found to increase toward the free surface.⁸ Therefore, this theoretical description yields only effective (averaged) parameters of the lateral dot correlation.

It is well established⁹ that the positions of the dots in different depths are correlated. The degree of correlation depends, e.g., on the thickness of the spacer layers between the dot layers. We denote the vertical correlation length as ξ_{corr} . Then the correlation function w_{nm} can be postulated in the form

$$\begin{aligned} w_{nm}(\mathbf{X} - \mathbf{X}') &= w_{\parallel}(\mathbf{X} - \mathbf{X}') \exp\left(-\frac{|Z_n - Z_m|}{\xi_{\text{corr}}}\right) \\ &\quad + c^2 \left[1 - \exp\left(-\frac{|Z_n - Z_m|}{\xi_{\text{corr}}}\right) \right]. \end{aligned} \quad (4)$$

If $\xi_{\text{corr}} \rightarrow \infty$, the dots are fully correlated vertically and $w_{nm} = w_{\parallel}$; for $\xi_{\text{corr}} = 0$ we obtain $w_{nm} = w_{\parallel} \delta_{nm} + c^2(1 - \delta_{nm})$. The second term on the right-hand side of Eq. (4) does not depend on the lateral coordinates and, therefore, it will only contribute to the central δ -like peak in $w_{nm}^{\text{FT}}(\mathbf{q}_{\parallel})$.

B. Diffuse x-ray scattering

Due to the small volume of a dot, the diffusely scattered intensity is weak and, therefore, can be described by means of kinematical diffraction theory. In the case of x-ray reflection, however, this approach cannot be used since it neglects x-ray refraction. In this case, a semikinematical theory is applicable (the distorted-wave Born approximation³¹). Both theoretical approaches result in very similar formulas yielding the distribution of the scattered intensity $I(\mathbf{q})$ in reciprocal space. The vector $\mathbf{q}$ is defined as $\mathbf{q} = \mathbf{K}_{\text{out}} - \mathbf{K}_{\text{in}} - \mathbf{h}$.

The expression for $I(\mathbf{q})$ in the case of diffraction can also be used for x-ray reflection, if the following three amendments are made: (1) $\mathbf{h}$ is set to zero. (2) $\mathbf{q}$ is replaced by the corresponding vector *inside* the crystal matrix, which accounts for the refraction at the crystal surface. (3) The scattered intensity is multiplied by the factor $|T_1 T_2|^2$, where $T_{1,2}$ are the Fresnel transmittivities of the crystal surface corresponding to the primary and scattered beams, respectively.³¹

The distribution of diffusely scattered intensity $I(\mathbf{q})$ is calculated by means of the statistical scattering theory of Dederichs²³ and Krivoglaз,²⁴ which yields the following expression for a crystal with statistically distributed defects:

$$\begin{aligned}
I(\mathbf{q}) &= C \sum_{\mathbf{R}} \sum_{\mathbf{R}'} w(\mathbf{R}, \mathbf{R}') F(\mathbf{q}; Z) F^*(\mathbf{q}; Z') e^{i\mathbf{q} \cdot (\mathbf{R} - \mathbf{R}')} \\
&\sim C \sum_{n=1}^N \sum_{m=1}^N w_{nm}^{\text{FT}}(\mathbf{q}_{\parallel}) F(\mathbf{q}; Z_n) F^*(\mathbf{q}; Z_m) e^{iq_z(Z_n - Z_m)},
\end{aligned} \quad (5)$$

where the constant C contains the static Debye-Waller factor, among other parameters. The function F is

$$\begin{aligned}
F(\mathbf{q}; Z) &= \int d^3\mathbf{r} [\chi_{hL}(e^{i\mathbf{h} \cdot \mathbf{t}(\mathbf{r}; Z)} - 1) \\
&\quad + \Omega(\mathbf{r})(\chi_{hD} - \chi_{hL})e^{i\mathbf{h} \cdot \mathbf{t}(\mathbf{r}; Z)}] e^{i\mathbf{q} \cdot \mathbf{r}}.
\end{aligned}$$

This formula can be simplified if we assume that the deformation field is weak outside the dots. Then, the first term in the square brackets can be replaced by $i\chi_{hL}\mathbf{h} \cdot \mathbf{t}(\mathbf{r}; Z)$. Obviously, this approximation is valid for larger $|\mathbf{r}|$, and therefore for smaller $|\mathbf{q}|$ and for smaller dot strengths. After some rearrangement, for small $|\mathbf{q}|$ we obtain

$$\begin{aligned}
F(\mathbf{q}; Z) &\approx \chi_{hD}\Omega^{\text{FT}}(\mathbf{q} + \mathbf{h}\hat{\epsilon}) - \chi_{hL}\Omega^{\text{FT}}(\mathbf{q} + \mathbf{h}\hat{\epsilon}) \\
&\quad + i\chi_{hL}\varphi^{\text{FT}}(\mathbf{q}; Z).
\end{aligned} \quad (6)$$

The diffusely scattered intensity is therefore a coherent superposition of three terms. The first term on the right-hand side of Eq. (6) expresses the wave scattered by the dot volume. Because of the deformation of the dot lattice, its maximum is shifted by $-\mathbf{h}\hat{\epsilon}$, and the shape of the intensity distribution is affected by the dot shape (due to the Fourier transform of the dot shape function Ω). The second term in Eq. (6) arises from the diffuse scattering from the corresponding holes in the deformed crystal matrix due to the Babinet principle, which is well known from classical optics. The maximum of this contribution again is centered at the point $\mathbf{q}_{\text{max}} = -\mathbf{h}\hat{\epsilon}$, and its phase is opposite to that of the first term. The third term in Eq. (6) describes the diffuse scattering from the deformation field around the dot. In the literature, the first two terms are called defect-core scattering and the third one corresponds to the Huang scattering.^{23,32} It is worthwhile to note that, in the case of x-ray reflection, the expression of $F(\mathbf{q}; Z)$ substantially simplifies:

$$F(\mathbf{q}; Z) = (\chi_{0D} - \chi_{0L})\Omega^{\text{FT}}(\mathbf{q}).$$

Inserting Eq. (2) into Eq. (6), we find that the function F can be written as

$$F(\mathbf{q}; Z) = F_1(\mathbf{q}) + F_2(\mathbf{q})\exp[-(q_{\parallel} + iq_z)Z], \quad (7)$$

where

$$F_1(\mathbf{q}) = (\chi_{hD} - \chi_{hL})\Omega^{\text{FT}}(\mathbf{q} + \mathbf{h}\hat{\epsilon}) - \chi_{hL}\Omega^{\text{FT}}(\mathbf{q})P \frac{\mathbf{h} \cdot \mathbf{q}}{q^2}$$

and

$$F_2(\mathbf{q}) = \chi_{hL}\Omega^{\text{FT}}(\mathbf{q})P\Phi(\mathbf{q}).$$

The term F_2 expresses the surface relaxation of the deformation field of the dot and, in most cases, it represents only a

small correction to F_1 , since we assume that the dot height is much smaller than the depths Z_n . From Eq. (7) it follows that the contribution of the surface stress relaxation is restricted to a region in reciprocal space with small $q_{\parallel}$, i.e., very close to the q_z axis. Its value will be discussed later. In addition, the relaxation term decreases with increasing layer thickness, and it will be shown that this term can completely be neglected for SQD arrays with thicker spacer layers.

Now, we insert Eq. (7) and the correlation function of the dot positions [Eq. (4)] into Eq. (5), and finally we obtain

$$\begin{aligned}
I(\mathbf{q}) &= \text{const } w_{\parallel}^{\text{FT}}(\mathbf{q}_{\parallel}) [|F_1|^2 H(iq_z, -iq_z; \xi_{\text{corr}}) \\
&\quad + |F_2|^2 H(-q_{\parallel}, -q_{\parallel}; \xi_{\text{corr}}) \\
&\quad + 2 \text{Re}(F_1 F_2^* H(iq_z, -q_{\parallel}; \xi_{\text{corr}}))],
\end{aligned} \quad (8)$$

where H is defined as

$$H(a, b; \xi_{\text{corr}}) = \sum_{n=1}^N \sum_{m=1}^N \exp(aZ_n + bZ_m) \exp\left(-\frac{|Z_n - Z_m|}{\xi_{\text{corr}}}\right).$$

The final formula Eq. (8) has a simple physical meaning. If the dots are fully correlated vertically ($\xi_{\text{corr}} \rightarrow \infty$), and if we neglect the surface stress relaxation ($F_2 = 0$), the q_z dependence of the scattered intensity is mainly determined by the structure factor of the superlattice $H(iq_z, -iq_z; \infty)$. Thus, in the reciprocal space map, the diffusely scattered intensity is concentrated in “stripes” parallel to q_x axis at the same positions $q_z = (2\pi n)/D$ as the coherent superlattice satellite maxima, where n is an integer. In the opposite limiting case, i.e., if the dots are completely uncorrelated vertically, then $H(iq_z, -iq_z; 0) = N$ holds. In this case, the q_z dependence of the scattered intensity is determined by $|F_1|^2$, i.e., it is rather broad and it exhibits no stripelike structure.

This behavior of the diffuse scattering is completely equivalent to the diffuse scattering from multilayers with correlated interface roughness. It has been found previously³³ for the case of x-ray reflection that the width of the diffuse intensity stripes depends on the vertical correlation length of the interface roughness.

The distribution of the scattered intensity in the q_x direction is determined by the lateral arrangement of the dots and, for x-ray diffraction, by the deformation fields inside and outside the dots. The former factor gives rise to lateral intensity satellites at the positions $q_x = 2\pi p/L$ ($L = \langle l \rangle$ is the mean dot distance, and p is an integer), where the function $w_{\parallel}^{\text{FT}}(\mathbf{q}_{\parallel})$ has a local maximum. The spacing of these satellites is $2\pi/L$, and their width is proportional to the dispersion of the distance distribution $S(l)$. The shape of these lateral satellites does not depend on the vertical dot correlation.

Since the volume of the deformed region around the dot is much larger than the dot itself, the contribution of the deformation field around the dots to the diffusely scattered intensity [the third term in Eq. (6)] is mainly concentrated in the close vicinity of SL_0 . Consequently, the intensity distribution far from SL_0 is influenced mainly by the core scattering, i.e., by the strain status of the dot lattice. Therefore, it is possible to determine the strain $\hat{\epsilon}$ in the dots from the intensity distribution in this region.

As was stated above, diffuse x-ray reflectivity ($\mathbf{h}=\mathbf{0}$) is completely insensitive to the deformation fields in the structure. Therefore, it is suitable for a determination of the correlation properties of the dot distribution, especially the degree of vertical correlation, as we show later.

We used a conventional laboratory x-ray source for the reciprocal space maps and a diffractometer with a very poor resolution in the q_y direction, i.e., perpendicular to the scattering plane. In order to simulate the measured signal J , we have to integrate the intensity obtained by Eq. (8) with respect to q_y ,

$$J(q_x, q_z) = \text{const} \int_{-K\psi}^{K\psi} dq_y I(\mathbf{q}),$$

where 2ψ is the angular acceptance of the detector perpendicular to the scattering plane. We proved numerically that this integration has nearly no effect on the simulated intensity distributions, and, hence, the simulated signal J can be replaced by $I(q_x, q_y=0, q_z)$.

V. DISCUSSION

Information on the Ge islands is contained in the diffuse peaks accompanying the coherent satellites. In the maps acquired in the symmetrical 004 diffraction [Figs. 2(a) and 3(a)], the diffuse peaks are symmetrical with respect to the q_z axis, whereas in the asymmetrical maps [Figs. 2(b), 2(c), 3(b), and 3(c)] they exhibit a distinct asymmetry. Close to SL_0 (i.e., for small $|\mathbf{q}|$), the diffuse scattering is determined mainly by the deformation field and the scattering factor of the lattice far from the dot [the third term in Eq. (6)]. The maximum of the diffuse intensity around SL_0 is shifted in the positive q_x direction, i.e., in the direction of larger h_x , which corresponds to the *compressive* deformation of the Si crystal lattice between the dots. The diffuse peaks around SL_1 are shifted in the negative q_x direction, i.e., to smaller h_x . Therefore, this portion of diffuse scattering stems from *expanded* regions of the lattice, i.e., from the dot volumes, and it is given mainly by the first two terms in Eq. (6).

From the experiments we determined (1) the average in-plane strain ε_{xx} in the dots, (2) the mean lateral distance of the dots and its statistical dispersion, and (3) the vertical correlation of the dots. Below, we discuss the determination of these parameters and the results.

A. In-plane strain in the dot volumes

As stated above, the in-plane strain ε_{xx} affects the lateral position of the diffuse maxima close to the nonzero order coherent satellites. We calculated the projections of the measured diffuse satellites close to SL_1 around various reciprocal-lattice points on the q_x axis (see Fig. 4 for sample C and Fig. 5 for D). We determined the positions of the maxima $q_{x\text{max}}$. Since, in this region of the reciprocal plane, the role of the third term in Eq. (6) (the contribution of the deformation field around the dots) is not completely negligible, there is no simple correspondence of $q_{x\text{max}}$ with ε_{xx} . In order to estimate the influence of the deformation field outside the dots (and, particularly of the elastic strength P) on $q_{x\text{max}}$, we calculated the dependence of $q_{x\text{max}}$ on ε_{xx} for different values of P . For this calculation, we used Eq. (8),

FIG. 4. Projections of the diffuse peaks around SL_1 on the q_x axis determined from the measured reciprocal space maps [Figs. 2(a)–2(c)], around 004, 113, 224, and 404 for sample C. The positions of the maxima indicated by solid arrows ($q_{x\text{max}}$) are proportional to the in-plane strain ε_{xx} in the dot volume. The dashed arrows show the lateral satellites (less distinct than around SL_0) due to the in-plane ordering of the dot positions.

and found that the calculated position $q_{x\text{max}}$ is nearly independent on the dot size, but it depends substantially on D . An example of the results of these calculation is plotted in Fig. 6 (sample C, around 224). From this plot, we determined possible pairs of values ε_{xx} and P that correspond to the measured value of $q_{x\text{max}}$. We performed this procedure for all nonsymmetrical reciprocal-lattice points (113, 224, and 404) for both samples C and D, and we obtained a triplet of lines in a ε_{xx} vs P plot (Fig. 7). Since ε_{xx} and P must not depend on the diffraction vector, the common intersection point of these lines corresponds to the true values of ε_{xx} and P in a sample. In the case of the samples with thinner spacer layers (samples B and A) the intensity of the SL_1 satellite was too weak for a reliable determination of the satellite maximum. Consequently, it was not possible to determine the in-plane strain ε_{xx} in these samples. Their elastic strength P was roughly estimated by comparing the measured and simulated 004 reciprocal space maps (see below) and we found $P \approx 5 \times 10^{-4}$ for both samples B and A with an uncertainty of about 100%.

FIG. 5. Analogous to Fig. 4 for sample D.

FIG. 6. The lateral position $q_{x\max}$ of the diffuse peak around SL_1 as a function of the in-plane strain ε_{xx} calculated for various elastic strengths P for sample C around the reciprocal-lattice point 224. The horizontal full line indicates the measured value of $q_{x\max}$.

In the case of sample C, we found $\varepsilon_{xx} = (1.4 \pm 0.1) \times 10^{-3}$ and $P = (1.5 \pm 0.5) \times 10^{-3}$ (see also Fig. 7 and Table I), and $\varepsilon_{xx} = (0.9 \pm 0.2) \times 10^{-3}$ and $P = (0.5 \pm 0.5) \times 10^{-3}$ for sample D. The accuracy of ε_{xx} and P was higher for sample C (thinner spacer layers) since, in this case, the distance of the diffuse satellite close to SL_1 from SL_0 is larger, and the influence of the third term in Eq. (6) on $q_{\max}$ is smaller.

The values of P were found to be much smaller than those according to Eq. (3) (this formula yields a value of P approximately ten times larger). The reason for this discrepancy is not completely clear yet; most likely, Eq. (1) is only a rough approximation, so that we determined only an effective value of P . A more exact solution for the deformation field $\mathbf{t}(\mathbf{r}; Z)$ around the dot can only be obtained by means of a numerical approach to the anisotropic elasticity problem (using finite element methods, among others).

We also investigated the lateral positions of the maxima of the nonzero diffuse satellites (orders from -4 to $+2$) of sample D around 206 reciprocal-lattice point using synchro-

FIG. 7. The calculated pairs $[P, \varepsilon_{xx}]$ corresponding to the measured value of $q_{x\max}$ for sample C, and the asymmetrical lattice points 113, 224, and 404. The actual values of P and ε_{xx} correspond to the common intersection point of the full lines.

FIG. 8. The calculated dependence of the strains ε_{xx} and ε_{zz} in the center of a cylindrical GeSi dot on the depth of the dot center below the free surface for a dot radius of $R = 750$ Å, a dot height of $h = 70$ Å, and a Ge concentration $x = 0.7$. Isotropic elastic constants identical for the dot lattice and the surrounding Si matrix were assumed.

tron radiation. We found that these positions do not depend on the satellite order, and they yield (within the error limits) the same value of ε_{xx} .

The vertical component ε_{zz} of the strain tensor cannot be determined from the reciprocal space maps, since the vertical distribution of the intensity in the diffuse satellite maxima is determined mainly by the superlattice structure factor $H(a, b; \xi_{\text{corr}})$. The maxima of this factor (i.e., the superlattice satellites SL_n) depend on the superlattice period D and on the mean vertical lattice parameter of the superlattice, which nearly equals a_{Si} .

To check the order of magnitude of the strains, we calculated the strains ε_{xx} and ε_{zz} in the center of a dot analytically as functions of the depth Z below the surface (see Fig. 8). We used the concept of the elementary point defect,^{27,29} and assumed a cylindrical shape of the dot and identical isotropic elastic constants in the dot volume and in the surrounding crystal matrix. The strains are proportional to $x\delta$, and we found that already for values of Z larger than approximately 1000 Å they no longer depend on Z . For typical dot sizes (radius $R = 750$ Å, dot height $h = 70$ Å, and the Ge content $x = 0.7$) the strain calculation showed that the dot structure is nearly pseudomorphic ($\varepsilon_{xx} = 1.1 \times 10^{-3}$ and $\varepsilon_{zz} = 4.4 \times 10^{-2}$). This value coincides quite well with the values of ε_{xx} following from the analysis of the reciprocal space maps, which are averaged over all 19 periods of the dot multilayer. From the calculations shown in Fig. 8, it is clear that in regions close to the surface the relaxation is much larger than for lower dots. In all samples the multilayer stack is capped with a Ge-dot layer, which results in a corrugated surface. The latter increases the degree of relaxation of dots in the topmost layers, since the calculated data in Fig. 8 hold only for a flat surface.

B. Lateral distribution of the dots

Information on the lateral order of the dots is contained in the Fourier transform $w_{\parallel}^{\text{FT}}(\mathbf{q}_{\parallel})$ of the in-plane correlation function of the dot positions. This function gives rise to

FIG. 9. Projections of the diffuse SL_0 peaks on the q_x axis calculated from the measured reciprocal space maps (Figs. 2 and 3), around the reciprocal-lattice points 004, 113, 224, and 404 for sample C. The positions of the lateral satellites (inversely proportional to the mean distance L of the dots) are indicated by vertical arrows.

maxima (lateral satellites) in the diffuse intensity arranged periodically in the q_x direction. This type of maxima can be found in the diffuse peaks close to SL_0 and (hardly resolved) close to SL_1 . In Figs. 9 (sample C) and 10 (sample D), we plotted the projections of these diffuse peaks on the q_x axis in all measured diffractions. Only one lateral maximum can be seen; the other are suppressed due to the large dispersion σ_l of the dot distances. In the case of symmetrical diffraction, no lateral maximum occurs, most probably due to the smaller projected coherence length of the incident x-ray beam as compared to asymmetric diffraction geometries. Similar maxima can also be resolved in the q_x projections of the SL_1 satellites (Figs. 4 and 7); in this case, however, the scattered intensity is much weaker, and the maxima are less distinct than at SL_0 . In the figures it can be seen that the positions of the lateral satellites are identical around the reciprocal-lattice points 113 and 224, and larger by a factor of approximately $\sqrt{2}$ around 404. For the 113 and 224 reciprocal space maps, the scattering plane was $\{110\}$, while it was $\{100\}$ for 404. Therefore, the difference in the satellite separations indicates that the dots are ordered in a square

FIG. 10. Analogous to Fig. 9, for sample D.

FIG. 11. Measured coherent ($q_x=0$) and incoherent ($q_x \neq 0$) q_z scans around the asymmetrical reciprocal lattice point 206 for sample C. S labels the substrate peak, and the numbers denote the orders of the superlattice satellites.

array, the axes of which are parallel to $\langle 100 \rangle$. AFM observations of self-assembled Ge dots grown at lower temperatures⁹ revealed the same behavior. From the satellite positions we found the mean distance of the dots $L = (6000 \pm 800)$ Å in sample C and $L = (5000 \pm 500)$ Å in sample D, which agree within the limits of experimental accuracy with atomic force microscopy data. In measured maps of the thinner samples (A and B) no distinct lateral maxima in the SL_0 diffuse maxima were resolved, and, therefore, the lateral separation of the dots L could not be determined.

From the width of the satellites, the root-mean-square dispersion σ_l of the dot distances can be estimated. We found $L/\sigma_l = 3 \pm 1$ in both samples, thus, roughly speaking, the dot positions are correlated up to the third-nearest neighbor; i.e., only a short-range ordering of the dots exists.

Since we assumed that the samples are statistically homogeneous, L and σ_l represent only effective values. They neglect the increase of the lateral ordering of the dot positions toward the free sample surface, which has been found by atomic force microscopy measurements.^{8,9} On the other hand, x-ray diffraction is sensitive to the lateral ordering of the dots averaged over the entire multilayer stack. Therefore it yields a statistically reliable mean value of the dot distance. Furthermore it has the advantage that the structural properties of dots buried under a cap layer can be investigated as well.

C. Vertical correlation

Vertical ordering of the dots influences the widths of the diffuse maxima in the q_z direction. These widths can be seen in Fig. 11, where we plotted the linear q_z scans measured around 206 (sample D) for various values of q_x using synchrotron radiation ($\lambda = 1.05$ Å). In these measurements, we used a narrow slit in front of the detector instead of an analyzing crystal, which is the reason for the worse resolution. However, from the figure it is obvious that the widths of the diffuse satellite maxima do not depend on the satellite order, and they are the same as the widths of the coherent maxima (measured for $q_x=0$). The coherent widths equal approximately $2\pi/(ND)$; in our measurement, however, they are

FIG. 12. Measured (points) and simulated (lines) x-ray reflectivity 2θ scans ($\mathbf{h}=\mathbf{0}$) of samples B and A. S denotes the specular peak, where the angle of incidence is equal to the exit angle ($\vartheta_{\text{in}} = \vartheta_{\text{out}}$).

larger due to the inferior resolution. From the measurement, we can estimate the vertical correlation length $\xi_{\text{corr}}/(ND) > 0.5$, and so we can conclude that more than 50% of the dots are vertically correlated.

For the investigation of the vertical correlation of the dot positions, a nonspecular x-ray reflectivity measurement (at $\mathbf{h}=\mathbf{0}$) is advantageous. It is not sensitive to the strain fields, and therefore an analysis of the experimental data is easier. We measured the nonspecular x-ray reflectivity using so-called 2θ scans, i.e., we varied the exit angle ϑ_{out} of the scattered beam with the sample surface by rotating the detector arm at a constant incidence angle ϑ_{in} of the primary beam. The measured and simulated 2θ scans of samples B and A are shown in Fig. 12, and the simulations were performed using Eq. (8) and $\mathbf{h}=\mathbf{0}$. It is obvious that the diffuse peaks are broader in the scan of sample A than in that of sample B. From the fit of the theoretical curves to the experimental data, we determined the vertical correlation lengths $\xi_{\text{corr}} = (1000 \pm 200)$ Å in sample B and $\xi_{\text{corr}} = (500 \pm 200)$ Å in sample A. Both in x-ray diffraction and x-ray reflection, a shorter vertical correlation length results in a broader SL_0 diffuse maximum in the q_z direction. This is also obvious from Fig. 13, where we plot the measured and simulated reciprocal space maps of sample B in the symmetrical diffraction 004.

The measured data indicate a clear tendency of an increase of ξ_{corr} with increasing thickness of the Si spacer layers. The value of ξ_{corr} reflects not only the positional correlation of the dots, it is also affected by a vertical inhomogeneity of the dot sizes. If, for instance, the positions of the dot centers were fully correlated vertically ($\xi_{\text{corr}} \rightarrow \infty$), but the sizes of the dots increased toward the free surface, we would determine a smaller, finite value of ξ_{corr} from the measured diffuse maxima. The cross-sectional TEM data²⁶ revealed a good vertical correlation of the dot positions in all the samples, and a distinct gradient of the dot sizes in the samples with thinner Si spacer layers (samples A and B). Consequently, as for the latter, the value of ξ_{corr} is rather affected by the size gradient.

FIG. 13. Measured (left) and calculated (right) reciprocal space map of sample B around 004. The higher-order diffuse satellites are not visible in the measured map. The width of the diffuse maximum in the q_z direction is broadened due to the reduced vertical correlation length in the dot multilayer.

D. Simulations of the reciprocal space maps

Finally, we simulated the measured reciprocal space maps using values of the superlattice period D , the in-plane strain ε_{xx} , the mean lateral distance of the dots L , its root-mean-square dispersion σ_L , and the correlation length ξ_{corr} of the dot positions as determined above. (The values are summarized in Table I.) The reciprocal space maps were not sufficiently sensitive to the other parameters of the SQD array, namely, the dot diameter R , the dot height h , and the chemical composition x . Therefore, these parameters could not be determined from the measurements, and we took the following mean values for all samples $R = 800$ Å, $h = 70$ Å, and $x = 0.7$. The symmetrical 004 maps have been calculated assuming $\sigma_L = L$; therefore, no lateral maxima in the diffuse peak around SL_0 occurred. As stated above, it was not possible to determine the values of L and ε_{xx} for the samples B and A. Therefore, we restricted ourselves only to the calculation of symmetrical 004 maps of the thinner samples B and A, which were not sensitive to these parameters.

For the simulations we used Eq. (8) and we assumed that the dot spacings are distributed according to a gamma distribution. The order of the gamma distribution is a function of σ_L ; e.g., for the simulation of the symmetrical 004 maps it was taken to be 1. The measured and simulated maps are compared in Figs. 2, 3, and 13. The simulated maps agree qualitatively with the measurements; however, the detailed shape of the diffuse maximum around SL_0 is not fully reproduced. Since this maximum is mainly affected by the third term in Eq. (6), it depends on the deformation field around the dots. This field was estimated using Eqs. (1) and (2). A better correspondence could be achieved, if the actual deformation field obtained from exact numerical calculations could be used. Unfortunately, since the simulations require a three-dimensional elastic deformation field at rather large distances from the dot center in a dense grid of points, this task would need considerable computational resources.

In calculating the diffuse scattering intensity around SL_0 , we made two simplifications, which might be the reason for the differences: In deriving Eq. (6), we replaced the term $\exp[i\mathbf{h} \cdot \mathbf{t}(\mathbf{r}; Z)] - 1$ by $i\mathbf{h} \cdot \mathbf{t}(\mathbf{r}; Z)$, and we assumed that the relaxation term in Eq. (2) does not depend on the dot shape.

FIG. 14. Reciprocal space maps of samples B [(a) and (b)] and D [(c) and (d)] around 004 calculated without [(a) and (c)] and with [(b) and (d)] a surface relaxation of internal stresses. In the case of the thicker spacer sample D, the surface relaxation affects only the closest neighborhood of the q_z axis.

In order to estimate the effect of the first simplification, we calculated the reciprocal space map taking the exact term $\exp(i\mathbf{h} \cdot \mathbf{t}) - 1$ into account, but neglecting the surface relaxation. We found that for elastic strengths P below 10^{-2} , the error introduced by the simplification is negligibly small. For a stronger deformation field, however, this simplification gives rise to a large difference in the calculated intensity.

As follows from Eq. (2), the surface relaxation of internal stresses affects reciprocal space maps in a region not far from the q_z axis. In Fig. 14 we compare the reciprocal space maps of samples D ($D=400$ Å) and B ($D=200$ Å) calculated with and without the surface relaxation. In the case of thicker layers, the relaxation changes the shape of the iso-intensity contours only in points with very small q_x , where the

diffuse intensity is very small compared with the coherently diffracted intensity. Therefore, we completely neglected the surface relaxation in the calculation of the reciprocal space maps of the samples D and C (Figs. 2 and 3, right-hand sides). If the layers are sufficiently thin, the surface relaxation affects a much wider region of the reciprocal plane and, therefore, the surface relaxation cannot be neglected. The calculated reciprocal space map of sample B (Fig. 13) includes the surface relaxation. Therefore, the discrepancy between the measured and simulated maps could be caused by the approximations in the derivation of Eq. (2).²⁸

VI. CONCLUSION

We investigated the structural properties of self-organized Ge islands on silicon using high-resolution x-ray diffraction and kinematical diffraction theory. From the reciprocal space maps measured in symmetrical and asymmetrical diffraction geometries, we determined the in-plane strain in the dots, and we estimated the effective strength of the deformation field around the dots. We found that the dot positions were partially correlated both in the horizontal and vertical directions. The horizontal ordering of the dots gave rise to lateral satellites close to the SL_0 superlattice satellite. From their positions and widths we estimated the mean dot distance and its statistical dispersion. The vertical ordering of the dot positions affects the width of the diffuse intensity maxima in the q_z direction. From the measurements, a distinct tendency toward increasing vertical order with increasing thickness of the Si spacer layers is observable.

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