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# Microstructures of Ge/Si Superlattices Growtn at Low Temperature*

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**Abstract** Si/Ge superlattices were grown at low temperature with modified Stranski-Krastanov (SK) MBE method. X-ray specular, off-specular reflectivity, and X-ray transverse scattering measurements were done to characterize the structure of the Ge/Si superlattice. The fitted roughness and the thickness of the Ge-layer indicate that Ge may diffuse into Si-layer and form the inverted trapezium or nano-scaled hut at the Si/Ge interface. The inverted trapezium extends to form islands, which can be estimated from the volume of the Ge-Si alloy in the structure. These islands can be averaged as a Ge-Si alloy sub-layer, the averaged thickness was fitted from the pure X-ray specular reflectivity. The composition of Ge in the SiGe dots was estimated as 15 %—25 % by X-ray specular reflectivity and by the thickness of Ge sub-layer. These results were confirmed by TEM observation.

**Key words** synchrotron X-ray reflectivity and transverse scattering, Si/Ge superlattices, MBE growth at low temperature

## 1 Introduction

Ge/Si superlattices and/or Ge-Si alloys with nano-structures have attracted great attention due to their outstandingly optical and electronic properties[1—5]. The 3-dimensional clusters ("Ge huts"), which are grown by the conventional Stranski-Krastanov (SK) growth mode at high temperature (from 500℃ to 750℃), are usually formed on top of the Si layer[6]. In recent several years, Cheng et al[7] have reported a modified SK growth technique, which produced high quality Si-Ge alloy nano-structures at much lower growth temperatures (from 260℃ to 450℃). Atomic force microscopy and scanning electron microscopy showed that the surface of the Si/Ge superlattices is smoother grown at low temperature than that growth at high temperature. Extended X-ray absorption fine structures studies[8] indicated that the mixing of Si and Ge at the interface of Si/Ge may be the reason why the Si/Ge superlattices growtn at low temperature form the smooth surface. The Ge distribution dependence on the depth in Si crystal has been detected by synchrotron X-ray reflection method[9]. In the present work, the grazing incident X-ray reflectivity and diffuse scattering methods were utilized to probe the surface and interfacial structures in Si/Ge superlattices grown at low temperature. Due to Ge diffusion into Si layer in the Ge/Si interface, the mixing of Si and Ge was proved to form a regular 3-dimensional (3D) structure arrays.

## 2 Experimental details

The sample used in the present work is a 6 period Si/Ge superlattice with the Ge-wetting layer thickness of 38 Å. All films were deposited by MBE and an about 400—500Å Si layer was first deposited as a buffer on a Si (100) wafer. The surface of the Si wafer was deoxidized, and then six periods of Ge/Si bilayers were grown at

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a temperature of 260℃ for Ge and 450℃ for Si, respectively. One period of Ge/Si consists of a Ge layer and a Si layer with thickness of 500Å.

Grazing incidence X-ray scattering measurements were carried out at the diffraction/scattering station on 4B9A beam line in Beijing Synchrotron Radiation Facility (BSRF). The X-ray energy of 8900 eV was picked up by a monochromator inserted in the beam line. Three types measurements were made: a) specular scan with the same detector angle $\beta$ and the sample angle $\alpha$ ($\beta = \alpha$), which records the intensity parallel to the reciprocal space vector $q_z$, as the detector is swept at twice the angular rate of the incidence angle; b) off-specular scan with $\beta = \alpha + \delta$, which records the intensity of diffuse scatter diverged from the sepcular ridge by scanning in the same way as specular scans but with a slight offset in the sample angle $\delta$. c) transvers scan or omega-scan, where the scattering angle $2\theta = \alpha + \beta$ between primary and scattered beams is kept constant and the sample rotates. Since the scattering angle $2\theta$ is very small, the omega scans correspond to the intensity distribution in reciprocal space along the line $q_z \approx$ constant. Specular reflectivity profiles as a function of incident angle were fitted using the Bede REFS MERCURY code. This uses a genetic algorithm to minimize the log absolute deviation between the experimental data and the simulated ones from a structure model according to the sample growth parameters as initial. The structural model can be adjusted during refinements.

## 3 Results and discussion

Fig.1 shows the specular curves for the sample, corrected for the forward diffuse scatter measured in $\delta = 0.1°$ offset longitudinal diffuse scan. The offset off-specular and the best fitted curves with the growth parameters as the initial structural model are also plotted in Fig. 1. More than 20 orders Bragg peaks of the Ge/Si superlattices are observed. The periods and the layer thicknesses of the samples were determined from the refinements. A model of 6 identical bilayers buffered by a Si layer and covered by a thin $SiO_2$ layer was used for the simulation of the specular reflectivity. The parameters following from the best fit are listed in Table 1.

**Table 1. The best fit parameters with the structural model according to the growth parameters. The thickness $t$ and the interface width $\sigma$ are in units of Å. $\rho$ and $\rho_0$ are the mass density of the film and the corresponding bulk materials, respectively.**

| Layers | | Si-buffer | Ge-layer | Si-layer |
|---|---|---|---|---|
| Ge/Si | $t$/Å | 435.5 | 25.6 | 473.1 |
| | $\sigma$/Å | 47.5 | 7.0 | 37.1 |
| | $\rho/\rho_0$ | 1.19 | 0.94 | 1.02 |

These fitted results show that the thickness for each layer is in agreement with respect to that experiments from the growth parameters except for the Ge-layer (the growth thicknesses are 450 Å for Si-buffer, 500Å for Si-sublayer). The fitted thickness of Ge-layers is much less than those of the nominal growth values (38Å). The roughness

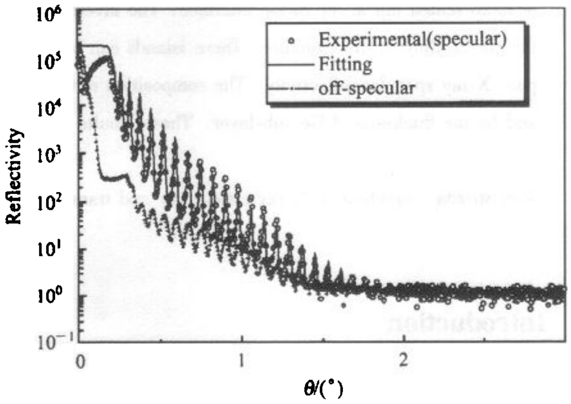

Fig.1. Specular and off-specular scans for Ge/Si superlattices with the thickness of Ge-layer of 38Å. The Kiessig fringes can not identified due to the experimental resolution. More than 20-order superlattice Bragg peaks are observed.

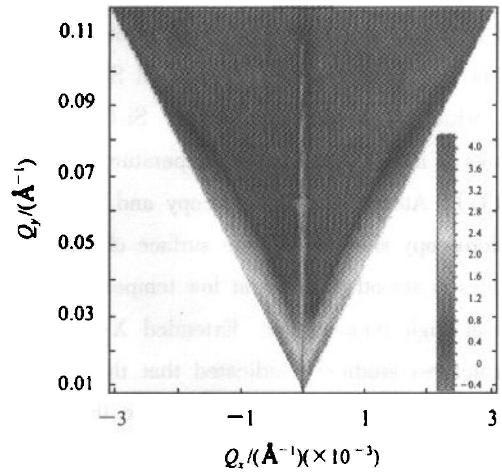

Fig.2. Reciprocal space maps for Si/Ge superlattice growth at low temperature.

or interface width of the Si-buffer and the Si-sublayer of Si/Ge superlattice are very large. The interdiffusion between Si and Ge in the interface of Si and Ge may occur.

To distinguish the true interface roughness from compositional grading, it is essential to measure the diffuse scatter. This is usually done by fixing the detector angle with scanning the sample from grazing incidence to grazing exit. This technique is often called a transverse scan due to its trajectory in reciprocal space (Fig.2). In the reciprocal space map, the intensity is concerned at the narrow center stripe, which represent the specular reflection at different detector angle ($\beta$). The Bragg peak can also be recognized from the map. Uniform weak intensity in Fig.2 represents the diffuse scatter, which tells us that the transverse diffuse scatter is weak. Fig.3 is the diffuse scan for the sample with the detector angles at the Bragg peaks of 1.20° and 1.81°. We note that there is a sharp peak appeared in the diffuse scatter pattern, which correspond to narrow specular peak. There are no clear Yoneda wings in either experimental or simulated curves of Fig.3. The majority of the diffuse scatter through the Bragg peak comes from the buried interfaces. Since the Yoneda wings arise from the enhancement of the electric field amplitude in the surface at the critical angle, Yoneda wings are hidden when the majority of the scattering is from the conformal roughness of the superlattices. The in-plane roughness correlation length ($\xi$), and Hurst fractral parameter ($h$) were obtained by matching two sets of experimental data for each sample to those of the corresponding simulated ones with a fratoral model within the distorted wave Born approximation[10]. The fitted parameters are listed in Table 2. The correlated length $\xi = 1100$Å and the Hurst factor $h_1 = 0.7$, which indicates the roughness is more 2-dimensional. By measuring the transverse scatter along different $\varphi$ angle ($\varphi$ angle is set as 0°, 90°, and 180°, respectively in our experiments), the similar fitted parameters may correspond to the 3-dimensional islands interface roughness. The ratio of the correlated to uncorrelated roughness is 15:85, which indicates that the roughness is less conform.

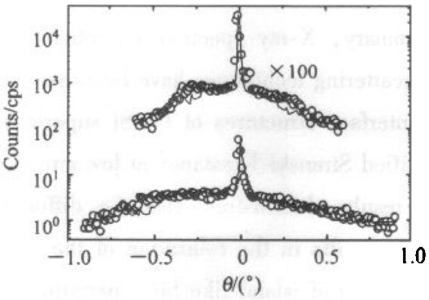

Fig.3. The fitting and experimental curves for the sample scan of the transverse scatter with the detector at two Bragg peaks of 1.20° and 1.81°.

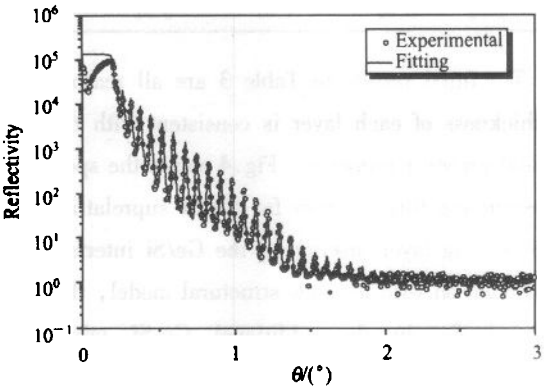

Fig.4. Experimental and fitted spcular reflectivity curves. An abstract Ge-Si alloy layer was inserted in the Ge/Si interface beneath the Ge-layer.

**Table 2. The fitted diffuse scattering parameters. The partial correlated interface is assumed. $\xi$ is the correlated length; $h_1$ is the Hurst factor, and 3-$h_1$ represents the interface roughness dimension; $h_2$ is the correlated fraction of the interface roughness.**

| Sample | $\xi$(Å) | $h_1$ | $h_2$ |
|---|---|---|---|
| Ge/Si | 1100 ± 100 | 0.7 ± 0.1 | 0.15 ± 0.03 |

The fitted interface width makes a contribution to both of the true height-height correlation roughness and the compositional grading as in specular scatters, the scattering vector has no in-plane component. However, the off-specular scatter is small. Thus the large interface width above the Si-layer at the Ge/Si interface (Table 1) is primary due to the compositional grading. The large interface width of Si-layer may correspond to the electron density grading resulting from the regular "inverted huts" profile arraying beneath the Ge-layers[7]. To fit the roughness to be a reasonable value, we assume that there exists a sub-layer of Ge-Si mixing at the Ge/Si interface. The assumed mixing layer works like a composition mixing in specular X-ray reflectivity. The specular X-ray reflectivity is fitted again with a sub-layer Ge-Si mixing inserted beneath Ge-layer at the Ge/Si interface. The fitted results are shown in Table 3.

**Table 3. The best fit parameters with the structural model containing a Ge-Si mixing layer.**

| Layer | | Si-buffer | Si-Ge mixing | Ge-layer | Si-layer |
|---|---|---|---|---|---|
| | $t$(Å) | 423.4 | 34.3 | 22.8 | 443.2 |
| Ge/Si | $\sigma$(Å) | 1.1 | 11.0 | 6.0 | 13.0 |
| | $\rho/\rho_0$ | 0.96 | 1.07 | 0.91 | 0.98 |

The fitted results in Table 3 are all reasonable and the thickness of each layer is consistent with that of the nominal growth parameters. Fig.4 shows the specular and corresponding fitted curves for Ge/Si suprelattice with a Ge-Si mixing layer inserted at the Ge/Si interface. From the simulation with the new structural model, the composition of Ge in the additional Ge-Si mixing layer ($Ge_xSi_{1-x}$) is from 0.15—0.25. As an estimation, we can take $x$ as 0.20. The mixing layer will cost about 11Å Ge for the formation of $Ge_xSi_{1-x}$ sub-layer. Therefore, the total thickness of Ge-layer is 34Å in each period. It is consistent with that of the growth thickness of 38Å. However, the same specular X-ray scatter fitting with different azimuth angles ($\varphi$ = 0°, 90°, and 180°) and the above diffuse scattering results support that the interface has the island-like profiles, i.e., the composition islands.

In contrast to the usual nano-sized Ge "hut clusters" commonly grown on top of Si layers using the convensional Stranski-Krastanow self-organized growth mode, the "groove islands" or "inverted huts" SiGe crystallized solution can be formed beneath the Ge wetting layer and grown insert into the Si layer in Ge/Si superlattices prepared by MBE in modified SK growth mode under low growth temperature[7,8]. The formation of the "inverted huts" depends on the Ge thickness in the Ge/Si superlattices besides the growth temperatures. By a two shell model, EXAFS studies[8] show that the mixing of Ge and Si atoms may take place in a zone of 3 — 5 monolayers around the Ge/Si interface with varying the Ge thickness. Above X-ray scattering results clearly show that the mixing is due to Ge diffusing into Si beneath the Ge layer.

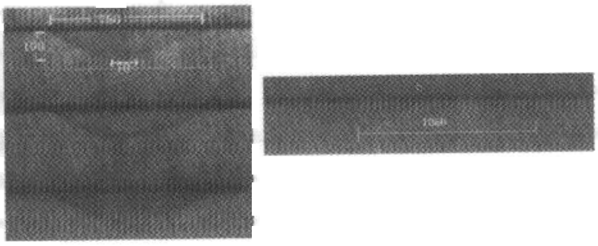

Fig.5. Cross-section transmission electron microscopy pattern for Ge/Si superlattice growtn at low temperature. The superlattice structure is clearly shown in the pattern. Regular arrayed island-like dots are observed (The unit used in the pattern is Å).

Cross-section transmission electron microscopy (TEM) has been done to prove the above X-ray scattering results (see Fig.5). TEM pattem clearly show the superlattice structure. The black thick lines are Ge-layer and the gray area is Si. Regular array of dark-gray islands or strips are seen beneath the Ge-layer in the pattern, which correspond to the Si-Ge alloy structures. The separation of the islands is about 1050Å, which corresponds to the correlation length obtained from the X-ray diffuse scattering results.

## 4 Conclusions

In summary, X-ray specular reflectivity and X-ray transverse scattering techniques have been used to characterize the interface structures of Ge/Si superlattice grown by the modified Stranski-Krastanov at low growth temperature. The results demonstrate that Ge diffused into Si layer, which results in the relaxation of the lattice strain and the formation of island-like SiGe nanostructural dots. The nano-dots are regularly arrayed beneath the Ge layer. Cross-section TEM patterns proved our results.

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# 低温生长的 Ge/Si 超晶格界面结构的 X 射线散射研究

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**摘要**　低温下用 MBE 方法生长了 Ge/Si 超晶格. X 射线反射及横向散射研究表明,Ge 亚层上下表面的粗糙度呈反对称,下表面大的粗糙度来源于 Ge 向 Si 亚层中扩散形成 SiGe 混合组分结构. 这种组分结构可以用一平均成分的 SiGe 合金层加以拟合,从而使得各亚层均有一个合理的粗糙度. 旋转样品进行的 X 射线散射研究表明,这种 SiGe 的混合是各向同性的,这与透射电子显微镜的研究结构相一致.

**关键词**　同步辐射 X 射线镜面反射和散射　Ge/Si 超晶格　低温分子束外延生长

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