Effect of Reflected Waves on the GISAXS Analysis of As-grown Capped Ge Nanodots

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Abstract. Effect of layer structures on the GISAXS intensity analysis of Ge nanodots capped with Si layer has been examined by Born Approximation and Distorted Wave Born Approximation simulations. It is concluded that when the dots are small enough in the out-of-plane directions, most of the structural information are deduced by simple BA. The effect of refracted wave became important when the area of interest is close to the Yoneda line or the analysis deals with small difference in the intermediate q range.

1. Introduction
Grazing incidence small-angle X-ray scattering (GISAXS) has been used to examine the microstructures at the surface region. For the interpretation of the scattering patterns obtained for such surface nanostructures, the dynamical effect [1] [2] has been taken into account as an interesting phenomenon characteristic for GISAXS [3]-[5]. Owing to such developments, it has become a common understanding that a simulation taking into account the dynamical effect is indispensable in the analysis of GISAXS profiles. By publications by Lazzari et al. [6] [7], many structures listed in them are readily available for simulation. On the other hand, it is also clear that how the dynamical effect should be taken into account strongly depends on the nanostructure of individual sample and the detail of the analysis. In the early stage of GISAXS studies, researchers who are more interested in microstructure itself intentionally avoid the condition where dynamical effect is dominant [8]. We have reported[9],[10] that even a simple Born Approximation is sufficient for a simple size and shape analysis of capped nanodot if the height of nanodots are reasonably small. In the present study, effect of reflected wave on the GISAXS profile were examined for as-grown Ge nanodot capped with a thin Si layer.

2. Experimental and simulation procedures

2.1. Sample
The sample examined in the present work contains capped Ge nanodots having no apparent facets. Germanium nanodots were grown on a Si (001) substrate with a gas-source MBE at 783 K, and then capped with a Si layer with a thickness of about 40 nm. Samples used in the present measurements were cut from center part of 2 inch wafers, where uniformity of thickness was confirmed for the growth condition.
2.2. Reflectivity/GISAXS measurements
Specular reflectivity was measured at Beam-line 13XU of SPring8, Japan. GISAXS measurements for the corresponding sample were made at the same beamline using a compact GISAXS apparatus on a multiaxis goniometer and also at BL15A of Photon Factory, Tsukuba, Japan using a conventional SAXS camera in a grazing-incidence geometry [9]. To examine in-plane anisotropy, several directions of incidence were chosen for GISAXS pattern measurements [10].

2.3. Simulations
Roughness and thickness of each layer in the sample were evaluated by using a least-square fit of the reflectivity curve. Then these parameters were used to calculate GISAXS pattern within a framework of DWBA. Since the scattering objects, Ge nanodots, are on a single plane, we assumed here that the scattered waves and these with reflections were coherent.

3. Results
Figure 1 gives a measured and a simulated specular reflectivity of the as-grown sample. The reflectivity curve shows two distinct characters, i.e., a short period oscillation and a slow modulation of the envelope. These two periods correspond to the total thickness of cap and dot layers, and the thickness of dot layer [11]. From a least-square fitting of the intensity, a simulation result suggests that the thickness of the cap Si layer and effective thickness of dot layers are 39.8 nm and 1.98 nm respectively. The roughnesses of the surface and cap/dot interface are estimated to be 0.5 nm and 0.9 nm respectively. These results suggest that the present sample has relatively smooth surface, although the roughness is not as good as a well-defined clean Si surface.

A GISAXS pattern obtained for in-plane angle of 45 degrees between the incident beam and [110] direction of the sample, $\phi$, is shown in Fig. 3. There are two main origins of diffuse scattering in the sample. On is the Ge nanodots which are of our main interests, and the other is surface and interface diffuse scattering. The latter has intensively analysed in the multilayer structures in the last two decades. From the

![Figure 1. Specular reflectivity of the capped Ge nanodots measured at BL13XU of SPring8. Triangular points correspond to a simulation results obtained by least-square fitting.](image)

![Figure 2. GISAXS pattern obtained for $\phi$=45 degree with the angle of incidence $\alpha$=0.35 degree. Two peaks extending towards $q_z$ direction are from Ge nanodots. Rectangular shadow in the figure is the specular beam stop. The line of spots at $q_y$=0 extending towards larger $q_y$ from the specular beam stop is a diffuse part of Kiessig fringe of the layer structure.](image)
present GISAXS profiles, we may conclude that the GISAXS from Ge nanodots appearing between the specular beam stop and \(|q_y| = 0.2\) and the diffuse scattering from surface/interface roughness, appearing at \(q_y = 0\) and above the specular beam stop, are well separated. Therefore, we may regard that the scattering intensity at the larger \(q_y\) corresponds to the scattering from Ge nanodots. As shown in the previous report [12], effect of correction term from DWBA should be small at the large \(q_y\). Figure 3 gives an example of GISAXS pattern and corresponding two-dimensional intensity profiles obtained by Born approximations. For example, the angle of incidence, \(\alpha\), used in the present measurements lies between 0.35 and 0.48 degree. Comparing the simulation results with these conditions, the correction term should be less than 1% considering the reflectivity at the angle. Then the size and shape of the nanodots were successfully reconstructed using these fitting methods. The shape parameters obtained from these fitting was \(R_y = 8.8\) nm in the base diameter and 2.3 nm in height, with a flat dome shape. These results agreed with direct observation using cross-sectional transmission electron microscope. Therefore, it is concluded that for size and shape analysis where we may use the scattering intensity at the region rather far from the Yoneda region to avoid dynamical effect, analysis under Born approximation gives enough information. This condition depends strongly on the size of nanodots. If the height of nanodots is large, then the characteristic \(q_z\) to be used for the analysis becomes too close to the critical angle, and dynamical effect need to be taken into account.

In the preceding analysis, we justified BA analysis from reflectivity curve shown in Fig.1 [9][10]. To make the analysis more quantitative, a DWBA simulation on the same sample was made. For the present sample, a simple layer model was used. For capped single layer of nanodots, GISAXS scattering amplitude is given in a form of [13]:

\[
A(k'_z, k_z^f, q_{||}) = T_f(k'_z)T_f(k_z^f)A(k_z^f - k_z^i, q_{||}) + R_fT_fA(k_z^f + k_z^i, q_{||}) + T_fR_fA(-k_z^f - k_z^j, q_{||}) + R_fR_fA(-k_z^f + k_z^j, q_{||})
\]

with \(k_z^i, k_z^j, q_{||}\) are the wave vector of incoming/outgoing perpendicular and in-plane wave, \(T\) and \(R\) are the transmission and reflection coefficients determined for the incoming/outgoing waves from recursion equation obtained for the reflectivity shown in Fig.1. From equation (1), the form factor of the scattering potential, namely, the shape of the nanodot, and the Fresnel coefficients as a function of
angle are necessary for DWBA simulation of GISAXS patterns. As shown in Fig. 3, the form factor is obtained from BA analysis of the scattering intensity. GISAXS profile calculated for the present nanodot sample is calculated by:

\[ I(q) = A(k^i_z, k^f_z, q_{//}) \cdot A(k^i_z, k^f_z, q_{//})^* \]  

where \( A^* \) is the complex conjugate of \( A \).

In the present simulation, we assumed that the four terms appearing in equation (1) are coherent, i.e., the amplitude can be simply squared after summation to give the intensity. When the sample has some thick layer of scattering bodies, such as nanopores distributing over a relatively thick layer, the scattering occurred near the center of the layer and that reflected at the layer/substrate interface may not be necessarily coherent because the distance between the scattering body and the interface is large. For such cases some decoupling assumption is necessary [14]. In the present case, however, only one layer of nanodot are formed on a well-defined flat substrate. That is, the interface which gives the reflection in the DWBA analysis is adjoining to the dots. The four waves in equation 1 are expected to be therefore almost coherent.

Figure 4 gives the amplitude of the coefficients, corresponding to the factors to be multiplied to the form factor in equation 1 as a function of total moment transfer in \( q_z \) direction. The transmission part corresponding to the first term is the strongest in the all \( q_z \) region. In contrast, the transmission-reflection (\( T_R \)) wave is quite strong near the Yoneda line, but decrease rapidly with \( q_z \). Therefore, the effect of reflected beam, or DWBA correction term, is not negligible only in the region close to the Yoneda line. For example, the third term decreased by a factor of about 30 at the \( q_z \) just 0.2 nm\(^{-1}\) above the Yoneda line. Then, relative magnitude of the correction term to the total intensity is only 0.1%. Therefore, it is concluded that the effect of reflected beam (DWBA correction) is small in the present case and analysis based on Born approximation is sufficient when we can avoid the area in \( q_z \) space about 0.2 nm\(^{-1}\) within the Yoneda line. These considerations depend on the sample structures and the incident angle. Therefore, DWBA analysis might be still necessary to confirm the validity of the approximation.

GISAXS pattern calculated from these parameters described above is shown in Fig. 6, with two principal waves that contributes to the scattering intensity. For simplicity, the calculation is made for a single nanodot having a size that agrees with the average one obtained by experiment, and with the thickness and roughness of the cap layer given from the reflectivity simulation. It is seen that the GISAXS pattern shown in Fig.5(a) is well explained by the transmission(Born) term of Fig.5(b), except the region close to Yoneda line.

When the four waves in equation (1) are coherent, it is worthwhile looking at the relationship of the phases between the waves. Figure 6 gives the phase of the amplitude two waves, i.e., transmission wave(\( T_T \)) and transmission-reflection (\( T_R \)) waves shown in Fig. 5, which give principal contribution.
near the Yoneda line. The phase just on the Yoneda line does not show clear difference, because of the
sign in the form factors in eq.(1). It also suggests that the intensity enhances just on Yoneda, and
modulation occurs at the region very close to Yoneda lines. This might be important for a sample
where a strong peak or line appears near the Yoneda line.

**Figure 5.** GISAXS intensity from single dome nanodot capped with a Si layer. (a) Total
intensity with coherence assumed, (b) transmission wave ($T_t,T_t$) intensity and (c)
Reflection-Transmission ($T_t,R_t$) waves. (b) and (c) gives principal contribution to GISAXS
intensity near the Yoneda regions. It is also seen that GISAXS pattern shown in (a) is
explained by the transmission term shown in (b). In contrast, $R_t,T_t$ term gives strong
contribution only at the Yoneda line.

**Figure 6.** Argument of the complex amplitude of the waves for the first (a) and
third (b) terms of equation (1) calculated for the
GISAXS pattern shown in Fig. 5(b) and (c). It is seen
that the pattern is reversed for the third term, as
suggested from equation (1), implying that the
phase pattern is reversed on reflection. The low $q_z$
region below $q_z=0.35\text{nm}^{-1}$
has no phase information
because no scattering
wave transmits here.
4. Conclusions
Reflectivity/GISAXS measurements were performed at SPring8 and Photon factory for a Ge nanodot sample grown on Si (001) substrate and capped with Si layer by gas-source MBE. From a reflectivity analysis, parameters required for DWBA simulation of GISAXS were obtained. Detailed analysis of each wave component gives quantitative contribution of each wave in the GISAXS pattern formation. From Born approximation, size and shape of nanodots are evaluated. By using the form factor obtained by the BA and the layer structure information obtained by reflectivity, DWBA simulation was made.

It is concluded that Born approximation is sufficient to evaluate the microstructure of the present nanodot sample. However, it might not be the case for self-organized polymer films where incident angle is generally much smaller and structure is larger. In conclusion, a combined analysis of GISAXS/reflectivity is therefore a useful tool to reconstruct buried nanostructure near the surface, in particular to examine the validity of analysis.

References
[1] Sinha S.K., Sirota E.B., Garoff S.and Stanley H.B. 1988, Phys. Rev. B 38 2297.
[2] Rauscher M. 1999 J.Appl. Phys. 86 6763.
[3] Renaud G., Lazzari R., Revenant C., et al., 2003 Science 300 1416.
[4] Paniago R., Metzger H., Rauscher M., Kovats, Z., Schultze J. Eisele I. and Ferrer S. 2000 J.Appl. Cryst., 33 433.
[5] Stangl J., Holy V. and Bauer G. 2004 Rev. Mod. Phys. 76 725.
[6] Razzari L 2002 J. Appl. Cryst. 35 406.
[7] http://www.insp.jussieu.fr/axe2/Oxydes/IsGISAXS/isgisaxs.htm
[8] Babonneau, D. Naudon, A., Thiaudiere D., and Lequien S. 1999 J.Appl. Cryst.,32 226.
[9] Okuda H., Ochiai S., Amemiya Y. and Ito K 2002 Appl. Phys. Lett. 81 2358.
[10] Ogawa T., Niwa H. Okuda H.and Ochiai S., 2005 Materials Science Forum, 475-479 1097.
[11] Tolan M., 'X-ray Scattering from Soft-Matter Thin Films', Springer Verlag Berlin1999.
[12] Okuda H., Ochiai S., Ohtaka M., Ichitubo T., Matsubara E., Usami N., Nakajima K., Sasaki S. and Sakata O., 2007 Trans. Mater. Res. Soc. Japan, 32 275.
[13] Rauscher M., Salditt T. and Spohn H., 1995 Phys. Rev B 52 16855
[14] Lee, B., Yoon J. Oh W., Hwang Y.T. Heo K., Jin K.S., Kin J. Kim K-W. and Ree M.H. 2005 Macromolecules 38, 3395.