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Author(s)	Shimura, Takayoshi; Umeno, Masataka; Takahashi, Isao et al.
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Comment on "Observation of a Distributed Epitaxial Oxide in Thermally Grown SiO$_2$ on Si(001)"

In a recent Letter, Munkholm, Brennan, Comin, and Ortega reported the results of an x-ray diffraction study of thermal oxide layers grown on Si(001) substrates [1]. They observed the same oscillations in the scattered intensity of the $11l$ rod as we had already reported [2], and have also observed the broad scattering peak which coincides with the scattering from the bulk silicon rod at $(1, 1, 0.45)$. They interpreted the broad peak as the actual scattering from the ordered oxide in the SiO$_2$ layer, which is responsible for the modulation of the crystal truncation rod (CTR), since the integrated intensity of the broad peak increases roughly linearly with oxide film thickness.

However, we do not agree with their interpretation that the broad peak is the scattering from well-oriented columnar grains of an epitaxial ordered oxide. The broad peak may be observed if the columnar grains exist in the amorphous thermal oxide film. But the oscillation of the CTR cannot be explained by their structural model. As seen in Fig. 2 of their Letter, the intensity of the broad scattering is a factor of 10 weaker than the intensity of the rod. The jump in intensity of the rod at $l = 0.45$ is almost a factor of 25. It is clear that the modulation of CTR scattering is not due to the broad scattering, even if the interference effect exists between the CTR and the broad peak. A glance at Fig. 2 of their Letter gives us the impression that the intensity modulation of the top of the sharp scattering is due to the broad scattering, since the intensity is given in logarithmic scale. However, it should be noted that the intensity of the narrow scattering, subtracted from the intensity of the broad peak, is modulated with l value, which we had already reported [2,3].

If the broad peak is due to some diffraction effect of the epitaxial ordered oxide, it is a significant result that provides us with further information about the atomic structure of the thermal oxide layer. However, we cannot completely exclude the possibility that the broad scattering is due to the tail of the resolution function, because they considered that the small horizontal and vertical divergence of an incident beam resulted in very high scattering resolution, even without a diffracted beam analyzer crystal. Their argument is untrue, since the resolution function is much correlated with the detector collimation [4–7]. Indeed, it is not important for the l scan, but it is one of the substantial parameters for the radial scans and rocking curves, because the resolution function passes the rod, when the intensity profile perpendicular to the rod is made. The situation of their case is very complicated, since the rod has the intensity modulation with the jump of a factor of a few decades and is tilted from the line perpendicular to the diffraction plane.

In addition, the characteristic properties of the broad peak make us doubtful of the origin. The intensity ratio of the broad peak to the sharp one is almost constant in the thickness oscillation of Fig. 2 of their Letter. The intensity of the broad peak increases with that of the sharp peak. The width of the broad peak decreases with the period of the thickness oscillation. These facts seem to indicate that the broad peak is due to the tail of the resolution function.

To summarize, we feel that the results of the x-ray diffraction study presented in [1] do not provide sufficient evidence of an epitaxial ordered oxide distributed throughout thermally grown oxide films on Si(001).

Takayoshi Shimura,[1] Masataka Umeno,[1] Isao Takahashi,[2] and Jimpei Harada[3]
 [1]Department of Material and Life Science
  Graduate School of Osaka University
  Suita 565, Japan
 [2]School of Science
  Kwansei Gakuin University
  Uegahara, Nishinomiya 662, Japan
 [3]X-Ray Laboratory
  Rigaku Corporation
  3-9-12 Matsubara-cho
  Akishima, Tokyo 196, Japan

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