Origin of unknown x-ray diffraction peaks from incommensurate superlattices

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Unexpected x-ray diffraction peaks have been observed in some thin film modulated structures in which each constituent element has a nonintegral number of atomic layers. The origin of these peaks has not been clearly identified. The positions and intensities of these peaks were analyzed by numerical calculation from a model superlattice. The results indicate that the positions of the anomalous peaks are caused by a new long range periodicity due to the nonintegral number of atomic layers of each constituent element and that the intensities of the anomalous peaks are determined by the interfacial structure between the two different kinds of atomic layers. © *1995 American Institute of Physics.*

Physical properties of a superlattice are closely correlated to the thicknesses of the constituent elements in a period and interfacial atomic configurations between two constituent layers. Structural analysis of a superlattice has been carried out along both the modulation direction and the lateral direction of an interface.[1–7] It is known that the atomic arrangement at the interface is controlled by various factors, such as the surface roughness of the substrate, island growth due to adsorbed atoms, intrinsic surface diffusion of the matrix elements, and thickness fluctuation of the top layers.[8,9] These might lead to interfacial roughness, which is defined as compositional variation from the average smooth interface along the lateral direction. This interfacial roughness cannot create new diffraction peaks. However, if each constituent element of the superlattice is grown at a very uniform deposition rate, each constituent element can have nonintegral number of atomic layers. The atomic configuration of the transition layers of this structure would be a mixture of two elements. The result of the nonintegral number of atomic layers deposited may create a new long range periodicity along the modulation direction of the superlattice. In this paper, such a superlattice is referred to as an incommensurate superlattice as far as synthetic superlattices are concerned. Schuller *et al.*[8] first introduced this kind of superlattice as well-controlled interfacial roughness, and distinguished it from random interfacial roughness. However, the origin of unknown x-ray diffraction peaks was not clearly explained. The aim of this paper is to explain the origin of the unusual x-ray diffraction peaks from incommensurate superlattice.

The model internal structures of the incommensurate superlattices are shown in Table I. For clarity, some terms are defined as follows: $\lambda_0 = t_A + t_B$ is the sum of the average deposited thickness of each constituent for the N times repeating deposition of two elements A and B. $t_{A(B)} = d_{A(B)}(m_{A(B)} + p_{A(B)})$, where $d_{A(B)}$ is the lattice spacings of the element, $A(B)$, along the modulation direction, and $m_{A(B)}$ and $p_{A(B)}$ are the integral and fractional parts of atomic layers in the deposited constituent $A(B)$, respectively. When 3.2 layers of A atoms and 5 layers of B atoms deposit uniformly as shown in the second column of Table I, a new translational symmetry, $5\lambda_0$, forms along modulation direction. The composition of the transition layers changes layer by layer, however, there is a mirror plane at half of $5\lambda_0$. The new period totally depends on the fraction of each constituent. Other examples in Table I are similar superlattices which create a new period of $5\lambda_0$.

The x-ray diffraction profile of the above structures can be calculated using the principal diffraction equation. The effect of the new periodicity due to the fractional part of the constituent can be calculated by

$$I(q) = \left| \sum_{j=1}^{M} f_j e^{iqd_j} \right|^2, \tag{1}$$

where M is the total number of atomic planes along the modulation direction. $q = 4\pi \sin \theta / \lambda_x$ is the scattering vector, f_j and d_j are the atomic scattering factor in the jth plane of atoms and the lattice spacing between jth and $(j-1)$th planes, respectively. In this model calculation, f_j of a transition layer is considered to be $p_A^t f_A + (1 - p_A^t) f_B$, where p_A^t is the fraction occupied by constituent A in the jth transition layer. It is assumed that the atomic configuration along the lateral direction on the transition layer is uniform. To see the effect of the compositional modulation and the new translational symmetry, the lattice spacings of the constituents are set to be equal, $\bar{d} = d_A = d_B = 2.246$ Å. The atomic scattering factors of the constituents are $f_A = 21.13$ and $f_B = 36.0$.

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TABLE I. The structure of the incommensurate superlattices which form new translational symmetry $5\lambda_0$. T_i's represent transition layers. AB_i's and BA_i's indicate the transition layers between A/B and B/A, respectively. $A_i(B_i)$'s represent only constituent $A(B)$ layers.

Layer	$(m_A+p_A)/(m_B+p_B)$ 3.2/5.0	3.4/5.0	3.6/5.0	3.8/5.0
A_1	3A	3A	3A	3A
$T_1(AB_1)$	$0.2A+0.8B$	$0.4A+0.6B$	$0.6A+0.4B$	$0.8A+0.2B$
B_1	4B	4B	4B	4B
$T_2(BA_1)$	$0.8A+0.2B$	$0.6A+0.4B$	$0.4A+0.6B$	$0.2A+0.8B$
A_2	2A	2A	3A	3A
$T_3(AB_2)$	$0.4A+0.6B$	$0.8A+0.2B$	$0.2A+0.8B$	$0.6A+0.4B$
B_2	4B	4B	4B	4B
$T_4(BA_2)$	$0.6A+0.4B$	$0.2A+0.8B$	$0.8A+0.2B$	$0.4A+0.6B$
A_3	2A	3A	2A	3A
$T_5(AB_3)$	$0.6A+0.4B$	$0.2A+0.8B$	$0.8A+0.2B$	$0.4A+0.6B$
B_3	4B	4B	4B	4B
$T_6(BA_3)$	$0.4A+0.6B$	$0.8A+0.2B$	$0.2A+0.8B$	$0.6A+0.4B$
A_4	2A	2A	3A	3A
$T_7(AB_4)$	$0.8A+0.2B$	$0.6A+0.4B$	$0.4A+0.6B$	$0.2A+0.8B$
B_4	4B	4B	4B	4B
$T_8(BA_4)$	$0.2A+0.8B$	$0.4A+0.6B$	$0.6A+0.4B$	$0.8A+0.2B$
A_5	3A	3A	3A	3A
B_5	5B	5B	5B	5B

Diffraction patterns of two different kinds of structures are shown in Fig. 1. Figure 1(a) is an illustration of a deposition with an integral number of atomic planes whose modulation wavelength is $8\bar{d}$. In this case, the satellite peaks, which are denoted as s^{+1}, s^{-1}, ..., due to composition modulation, appear on both sides of the main peak indicated by "m". The position of the satellite peaks can be written by

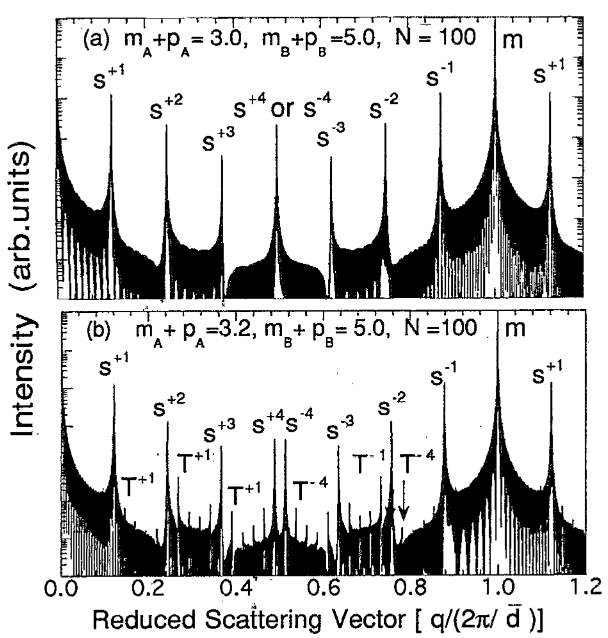

FIG. 1. Calculated x-ray diffraction profiles for the commensurate superlattice in (a), and incommensurate superlattices in (b). The vertical scale is an arbitrary unit in log scale. The symbol, m, indicates the main peak caused by the average lattice spacing, $\bar{d}$.

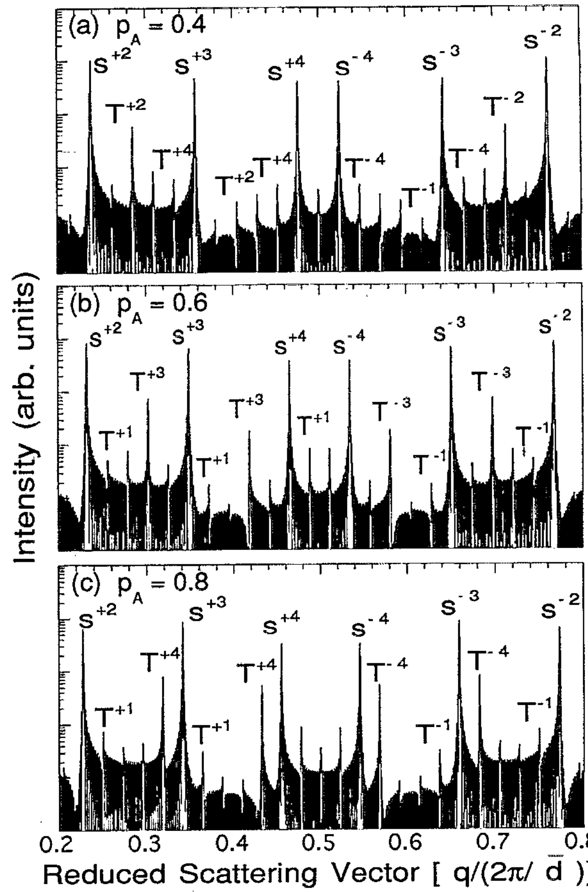

FIG. 2. Calculated x-ray diffraction profiles for an incommensurate superlattice with $(m_A+p_A)/(m_B+p_B)$, and $N=100$. The fraction, p_A, was changed to 0.4, 0.6, and 0.8, and p_B as zero.

$$q=\frac{2\pi n}{\bar{d}}\pm\frac{2\pi n'}{\lambda_0}, \tag{2}$$

where n and n' are the integers, and λ_0 is the compositional wavelength. In contrast, Fig. 1(b) is the diffraction pattern of a deposition with a nonintegral number of atomic planes as well as of the formation of new translation symmetry, $5\lambda_0$. This incommensurate superlattice has three different kinds of periodic atomic arrangements, $\bar{d}$, $8.2\bar{d}$, and $41\bar{d}$. From this structure, two kinds of satellite peaks arise and those positions are as follows:

$$q=\frac{2\pi n}{\bar{d}}\pm\frac{2\pi n'}{\lambda_0}\pm\frac{2\pi n''}{D\lambda_0}, \tag{3}$$

where n'' are integers and $D\lambda_0$ is the new wavelength which is $5\lambda_0$ in this case. The satellites arising from λ_0 modulation are indicated by the symbol, s^{-1}, s^{+1},..., and those from $5\lambda_0$ by the symbol, T^{-1}, T^{+1},... . The peaks of the incommensurate superlattice, $T^{n''}$, are not anomalous ones, but satellites due to the new translation symmetry, $5\lambda_0$.

The two different kinds of satellites can be anticipated from an incommensurate superlattice using Eq. (3). The intensities of the satellites depend fully on the internal structure of the incommensurate superlattice. Figure 2 illustrates the changes in the positions and intensities of two kinds of satellite peaks, $s^{n'}$ and $T^{n''}$, due to the fractional changes of the constituent A, p_A. The intensities of $T^{n''}$ are changed remarkably by changing the fraction of the constituents at the interfaces. The positions of the satellites are also changed by changing the fraction, p_A. From this result, the fraction of the constituents in an incommensurate superlattice can be inferred from the satellite position and intensity. As shown in Fig. 2, the intensities of the satellites, $T^{n''}$, rely on the fraction of the constituents. Further, the distance between s^{+4} and s^{-4} also depends on the fraction of the constituents. Using Eq. (3), this distance can be estimated as $\Delta q = 2\pi (p_A + p_B)/\lambda_0$ with $p_B = 0$ for this case.

In summary, we have shown the satellite peaks which can arise from not only uniform incommensurability of the superlattice but also from a new translational symmetry formed by long range incommensurability under well controlled deposition. The positions and intensities of these peaks can give information of the interface structure.

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