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The Dynamic Theory of X-ray Diffraction by the One-dimensional Ideal Superlattice. II. Calculation of Structure Factors for Some Superlattice Models

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Abstract

The superlattice unit-cell structure factors are calculated for the trapeziform model, which may be used for the epitaxically prepared one-dimensional superlattice crystals if the interdiffusion is taken into account. The particular cases of this model are: (1) the rectangular model, which describes the superlattice if the interdiffusion is negligible; (2) the triangular model, which describes the superlattice if the interdiffusion takes place all over the layer thickness. The change of satellite intensities during the interdiffusion is discussed. The results obtained may be used to interpret the X-ray diffraction patterns produced by superlattices.

1. Introduction

The characteristic feature of X-ray diffraction by superlattices (SL) or modulated structures is the presence of satellites around the principal diffraction maximum, the location of which is defined by an average over the SL period lattice parameters. At $z_0 \ll \bar{\Lambda}$ (z_0 and $\bar{\Lambda}$ being the SL period and the mean extinction length of the crystal, respectively) the angular separation between satellites is inversely proportional to the SL period, and their widths and intensities depend on the real structure and the SL thickness. In order to interpret the experimental data obtained from the X-ray reflection curves, various models have been proposed for the SL.

Firstly, Kochendörfer (1939) and Daniel & Lipson (1943, 1944) theoretically considered the sinusoidal modulations of the lattice parameter and structure factor in some alloys to explain the appearance of satellites around the principal maximum. Though their results qualitatively explained the experimental data, there have also been some essential discrepancies.

Hargreaves (1951) considered the SL square-wave model but because of approximations the results turned out to be the same as those for the sinusoidal model.

Then various theoretical models have been proposed for longitudinal and transverse modulations (Balli & Zakharova, 1954; Tiedema, Bouman & Burgers, 1957; Biederman, 1960; Guinier, 1955), which were extensively reviewed by de Fontaine (1966) and Korekawa (1967). All the above-mentioned models relate to the lattice parameter and structure factor modulations in binary and ternary alloys.

Esaki & Tsu (1970) proposed and realized the idea of obtaining the SL by heterojunctions using molecular beam epitaxy. For such a SL Segmüller & Blakeslee (1973) proposed the sinusoidal model including the second harmonic.

In all the above-mentioned works the derivation of the SL reflection amplitude formula was carried out by the use of the solution for sinusoidal modulation, when expanding in a Fourier series the periodic function of the chosen SL. The result is a sum of the infinite series of Bessel functions. Because of a rapid convergence of the series the authors usually considered only the few first terms. Of course, such a procedure gives very approximate results if the series is a slowly converging one. The direct calculation of the reflecting amplitude for the transverse modulation of rectangular and triangular forms was performed by Böhm (1975). In his consideration the lattice parameter was taken as being unchangeable and only the alteration of the atom positions was taken into account. Therefore, the formulae obtained by Böhm are valid only for certain types of modulations in antiferroelectrics and for specific types of twinning, and they are not applicable for the rectangular model of SL based on the heterojunctions of GaAs–AlAs type. For such types of SL Segmüller & Blakeslee (1973) obtained a formula for the reflection amplitude, but they did not make any detailed analysis to enable a direct comparison with the experiment.

Kolpakov, Khapachev, Kouznetzov & Kouz'min (1977) proposed a trapeziform model for the SL based on heterojunctions, the interdiffusion of the components between the neighboring layers being taken into account. However, the expression obtained by

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them for the reflection amplitude is not applicable for practical analysis, and it is difficult to form any conclusions concerning the form of satellites.

The SL model consisting of ideal layers with equal thicknesses and having equal shifts of crystalline lattices is considered by Vardanyan & Manoukyan (1982). With such a model the overlapping stacking-fault defects and antiphase boundaries are described.

In the present paper, based on the theory developed in paper I (Vardanyan, Manoukyan & Petrosyan, 1985) the structure factor for the trapeziform SL cell model is calculated. From the formula obtained the structure factors for the rectangular and triangular SL cell models are found. The formulae are of a simple form and useful for the analysis. The designations are the same as those in paper I (hereafter referred to as I).

2. Trapeziform model

The artificial SL crystals (of GaAs–AlAs type) may be approximately described by the trapeziform model, in which the diffusion between alternating layers is taken into account. It is supposed that the layer thickness is less than the critical thickness for misfit dislocation production (Matthews & Blakeslee, 1974). For such a model the deviation function from the Bragg angle is of the form (see Fig. 1)

$$s(z)=\begin{cases} s_1 & \text{if } z<z_1 \\ s_1+\dfrac{\Delta s}{z_d}(z-z_1) & \text{if } z_1<z<z_1+z_d \\ s_2 & \text{if } z_1+z_d<z<z_1+z_2+z_d \\ s_2-\dfrac{\Delta s}{z_d}(z-z_1-z_2-z_d) & \\ & \text{if } z_1+z_2+z_d<z_1+z_2+2z_d, \end{cases} \tag{1}$$

where

$$z_0=z_1+z_2+2z_d \tag{2}$$

is the SL period, z_d is the transition-layer thickness and the quantity

$$\Delta s=s_2-s_1=2k\sin\bar{\theta}_B(\Delta d/\bar{d}) \tag{3}$$

characterizes the degree of a heterojunction misfit, $\Delta d=d_2-d_1$ is the difference between the interplanar spacings, $\bar{d}$ is the mean interplanar spacing and $k=1/\lambda$ is the wave number in vacuum.

The average over the SL period value of $s(z)$ is

$$\begin{aligned}\bar{s}&=(1/z_0)\int_0^{z_0}s(z)\,\mathrm{d}z\\ &=[s_1(z_1+z_d)+s_2(z_2+z_d)]/z_0.\end{aligned} \tag{4}$$

The diffraction maxima directions are defined from equation (I-32):

$$\bar{s}_m=m/z_0\quad (m=0;\pm1;\pm2;\ldots). \tag{5}$$

The angular separations between the mth maximum and the directions $s_1=0$ and $s_2=0$ are found from (3) and (4):

$$s_{1m}=p_{1m}\Delta s, \tag{6a}$$

$$s_{2m}=p_{2m}\Delta s, \tag{6b}$$

where

$$p_{1m}=(m-\varepsilon_2-\varepsilon_d)/\varepsilon_0, \tag{7a}$$

$$p_{2m}=(m+\varepsilon_1+\varepsilon_d)/\varepsilon_0 \tag{7b}$$

and

$$\varepsilon_j=\Delta s z_j\quad (j=0;1;2;d). \tag{8}$$

Note that $p_{1m}(\varepsilon_1+\varepsilon_d)+p_{2m}(\varepsilon_2+\varepsilon_d)=m$.

Since we consider the short-period SL, *i.e.* $z_0\ll\bar{\Lambda}$, the Fourier components of the SL susceptibility may be represented in the form [equation (I-37)]

$$|\chi_{hm}|=M_m|\bar{\chi}_h|, \tag{9}$$

where $\bar{\chi}_h$ is the mean value of the Fourier components of the crystal susceptibility. The coefficient M_m, which we call the SL cell structure factor, is defined by (I-31) and (I-35):

$$M_m=\left|(1/z_0)\int_0^{z_0}\exp\left[-2i\pi\int_0^z s(z)\,\mathrm{d}z\right]\mathrm{d}z\right|_{\bar{s}=\bar{s}_m}, \tag{10}$$

where the term of order $\Delta\chi_h/\bar{\chi}_h$ is neglected.

Substituting (1) into (10) and taking the integral (Gradstein & Rizjik, 1971) we find

$$\begin{aligned}M_m=\Bigg|&\frac{\sin(\pi p_{1m}\varepsilon_1)}{\pi p_{1m}\varepsilon_0}+(-1)^m\frac{\sin(\pi p_{2m}\varepsilon_2)}{\pi p_{2m}\varepsilon_0}\\ &-F_{1m}-(-1)^m F_{2m}\Bigg|,\end{aligned} \tag{11}$$

where

$$\begin{aligned}F_{jm}=(\varepsilon_d^{1/2}/\varepsilon_0)[&\sin(\pi p_{jm}\varepsilon_j)U_{3/2}(2q_{jm}^2,0)\\ &-(-1)^j\cos(\pi p_{jm}\varepsilon_j)U_{1/2}(2q_{jm}^2,0)],\end{aligned} \tag{12}$$

$$q_{jm}=(\pi\varepsilon_d)^{1/2}p_{jm} \tag{13}$$

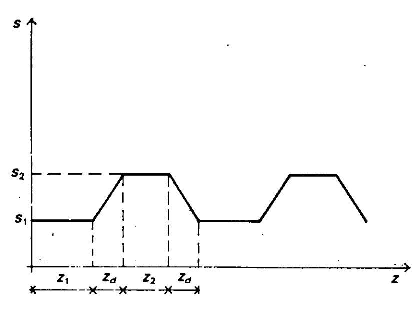

Fig. 1. A trapeziform SL model. z_1 and z_2 are the thicknesses of the undiffused layers with interplanar distances d_1 and d_2, respectively. z_d is the transition-layer thickness.

and

$$U_\nu(2x, 0) = \sum_{k=0}^{\infty} \frac{(-1)^k x^{\nu+2k}}{\Gamma(\nu+2k+1)} \tag{14}$$

is the two-variable Lommel function (Watson, 1944).

The first and second terms in (11) are the contributions of the undiffused parts of sublayers with thicknesses z_1 and z_2 and the third and fourth ones are those for the transition layer with thickness z_d.

Let us consider the particular cases of the trapeziform SL model.

3. Rectangular model

Taking into account that the interdiffusion of SL components takes place very slowly (Chang & Koma, 1976), at the early stage after the preparation of SL, one may set $z_d = 0$ and from (7*a*), (7*b*) and (11) one obtains

$$p_{1m} = (m - \varepsilon_2)/(\varepsilon_1 + \varepsilon_2) \tag{15a}$$

$$p_{2m} = (m + \varepsilon_1)/(\varepsilon_1 + \varepsilon_2) \tag{15b}$$

and

$$M_m = \frac{1}{\pi\varepsilon_0}\left|\frac{\sin(\pi p_{1m}\varepsilon_1)}{p_{1m}p_{2m}}\right|. \tag{16}$$

Note that $\sin(\pi p_{1m}\varepsilon_1) = (-1)^m \sin(\pi p_{2m}\varepsilon_2)$.

It is seen from (16) that, when the superperiodicity is absent, *i.e.* at $\Delta s \to 0$, all the satellites disappear except the principal maximum $M_0 = 1$.

For the principal maximum from (16) we obtain

$$M_0 = |\sin y/y|, \tag{17}$$

where

$$y = \pi\varepsilon_1\varepsilon_2/(\varepsilon_1 + \varepsilon_2). \tag{18}$$

The structure factor M_0 is an oscillating one and is significant at $y \lesssim 1$, *i.e.* when

$$\Delta s \lesssim 1/z_1 + 1/z_2. \tag{19}$$

Condition (19) means that the reflection curves of two sublayers with z_1 and z_2 thicknesses and d_1 and d_2 interplanar spacings, respectively, overlap each other. The structure factors of other maxima in that case are relatively small.

If the misfit is great,

$$\Delta s \gg 1/z_1 + 1/z_2, \tag{20}$$

i.e. when the reflection curves of two sublayers are separated, the structure factor M_m becomes appreciable for those m at which $|s_{1m}| \gg |s_{2m}|$, or *vice versa*.

If $|s_{1m}| \ll |s_{2m}|$, then

$$M_m = \frac{z_1}{z_1 + z_2}\left|\frac{\sin(\pi p_{1m}\varepsilon_1)}{\pi p_{1m}\varepsilon_1}\right| \tag{21a}$$

and if $|s_{2m}| \gg |s_{1m}|$, then

$$M_m = \frac{z_2}{z_1 + z_2}\left|\frac{\sin(\pi p_{2m}\varepsilon_2)}{\pi p_{2m}\varepsilon_2}\right|. \tag{21b}$$

Thus, when (20) is satisfied the diffraction maxima are located around the directions $s_1 = 0$ and $s_2 = 0$, *i.e.* there are, in fact, two SLs, for which diffraction maxima directions do not overlap each other.

If $\varepsilon_1 = n_1$ or $\varepsilon_2 = n_2$ (n_1 and n_2 integers), then from (15*a*), (15*b*) we obtain $s_{2(-n_1)} = 0$ or $s_{1n_2} = 0$, and from (16),

$$M_{-n_1} = z_2/(z_1 + z_2) \tag{22a}$$

$$M_{n_2} = z_1/(z_1 + z_2). \tag{22b}$$

4. Square-wave model

If the sublayer thicknesses are equal ($z_1 = z_2 = z_0/2$), then from (16)

$$M_m = \begin{cases} \dfrac{\varepsilon_0}{\pi}\left|\dfrac{\sin(\pi\varepsilon_0/4)}{m^2 - \varepsilon_0^2/4}\right| & \text{for even } m \\[2ex] \dfrac{\varepsilon_0}{\pi}\left|\dfrac{\cos(\pi\varepsilon_0/4)}{m^2 - \varepsilon_0^2/4}\right| & \text{for odd } m. \end{cases} \tag{23}$$

At $\varepsilon_0 \ll 1$, from (23) we obtain

$$M_m = \begin{cases} 1 - (\pi\varepsilon_0)^2/96 & \text{for } m = 0 \\ \varepsilon_0^2/4m^2 & \text{for even } m \neq 0 \\ \varepsilon_0/\pi m^2 & \text{for odd } m. \end{cases} \tag{24}$$

It is seen from (24) that at a small misfit the principal maximum only is appreciable.

With the increase of ε_0 the principal maximum oscillation decreases:

$$M_0 = \sin(\pi\varepsilon_0/4)/(\pi\varepsilon_0/4). \tag{25}$$

At $\varepsilon_0 = 2m$ only the mth maximum is appreciable

$$M_m = 0{\cdot}5, \tag{26}$$

which means that at the angles of incidence $\bar{s}_m = \pm|m|/z_0$ there will be maxima of equal intensity, because in the case of the square-wave model the diffraction pattern is symmetrical with respect to direction $\bar{s} = 0$.

5. Triangular model

If the SL layers are thin, owing to the interdiffusion of SL components, different materials will be overlapped throughout the layer. One may describe such an SL either by the sinusoidal model or by the symmetrical triangular model. The SL structure factor for the sinusoidal model is defined by Daniel & Lipson (1943, 1944):

$$M_m = |J_m(\varepsilon_0/2)|, \tag{27}$$

where J_m is the Bessel function, and

$$\varepsilon_0 = \Delta s z_0 = 2hu_0, \tag{28}$$

h is the reciprocal vector, u_0 the amplitude of modulation.

The symmetrical triangular model becomes the particular case of the trapeziform model if one sets $z_1 = z_2 = 0$, then the SL period is equal to $z_0 = 2z_d$.

From (11)-(13),

$$M_m = \frac{1}{(2\varepsilon_0)^{1/2}} \left| U_{1/2}(2q_+^2, 0) - (-1)^m U_{1/2}(2q_-^2, 0) \right|, \tag{29}$$

where

$$q_\pm = (m \pm \varepsilon_0/2)(\pi/2\varepsilon_0)^{1/2}. \tag{30}$$

In (29) one has to take into account that

$$U_{1/2}[(-x)^2, 0] = -U_{1/2}(x^2, 0).$$

For the principal maximum

$$M_0 = (2/\varepsilon_0)^{1/2} U_{1/2}(\pi\varepsilon_0/4, 0). \tag{31}$$

At $\varepsilon_0 \ll 1$, from (31),

$$M_0 \simeq 1 - \pi^2 \varepsilon_0^2/240. \tag{32}$$

For the satellites $m \neq 0$, at $\varepsilon_0 \ll 1$ we use the asymptotic formula for $U_\nu(x, 0)$ at $x \gg 1$ (Watson, 1944):

$$U_\nu(x, 0) \simeq \cos(x/2 - \pi\nu/2) + \sum_{k=0}^{\infty} \frac{(-1)^k}{\Gamma(\nu - 1 - 2k)(x/2)^{2k+2-\nu}}$$

yielding

$$M_m = \begin{cases} 2\varepsilon_0/\pi^2 m^3 & \text{for even } m \neq 0 \\ 3\varepsilon_0^2/\pi^2 m^4 & \text{for odd } m. \end{cases} \tag{33}$$

For several satellites $M_m(\varepsilon_0)$ dependence on different SL models is shown in Fig. 2.

6. The change of SL cell structure factors during the interdiffusion

The rectangular model describes the SL at the early stage after being prepared. Owing to the interdiffusion the layer boundaries successively become smooth (Chang & Koma, 1976).

Let us designate the initial thicknesses of sublayers by z_{10} and z_{20}. As a result of the interdiffusion the transition layer of thickness z_d is formed:

$$z_{j0} = z_j + z_d \quad (j = 1, 2), \tag{34}$$

where z_j are the thicknesses of undiffused parts of sublayers. The SL period z_0 and the mean values $\bar{\chi}_h$, $\bar{d}$ and $\bar{s}$ are unchanged. The change of SL cell structure factors during the interdiffusion is shown in

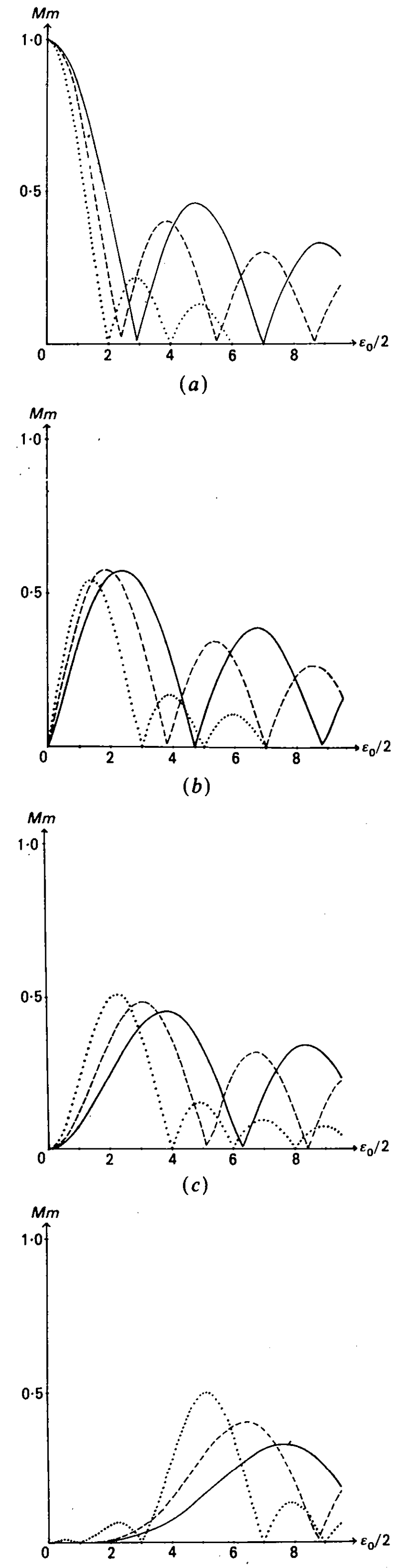

Fig. 2. ε_0 dependence of M_m. Solid curve is for the triangular model, dashed curve is for the sinusoidal model, dotted curve is for the square-wave model. (*a*) $m = 0$; (*b*) $m = 1$; (*c*) $m = 2$; (*d*) $m = 5$.

Fig. 3. The plots are drawn for $\varepsilon_1 = \varepsilon_2 = \varepsilon$. In this case

$$\varepsilon_0 = 2(\varepsilon + \varepsilon_d), \tag{35}$$

where ε_j are defined by (8).

The values $\varepsilon_d = 0$ and $\varepsilon_d = \varepsilon_0/2$ correspond to the square-wave and triangular models, respectively.

During the interdiffusion, at relatively small ε_0 the SL structure factors vary monotonically, while at large ε_0 the variation is an oscillating one.

At small thicknesses of the transition layer ($z_d \ll z_j$), *i.e.* at the beginning of the interdiffusion process, one may neglect the X-ray diffraction on this layer and make use of the approximation $s_{jm} z_d \ll 1$, *i.e.* $q_{jm} \ll 1$. Keeping only the first term in the Lommel function expansion [(14)], from (11) and (12) we obtain

$$M^t_m = M^r_m[1 - (\pi^2/6)\varepsilon_d^2 p_{1m} p_{2m}], \tag{36}$$

where M^r_m is the structure factor of the rectangular model with sublayer thicknesses z_{10} and z_{20} [(16)], and M^t_m is that for the trapeziform model. p_{1m} and p_{2m} are given by (15*a*) and (15*b*).

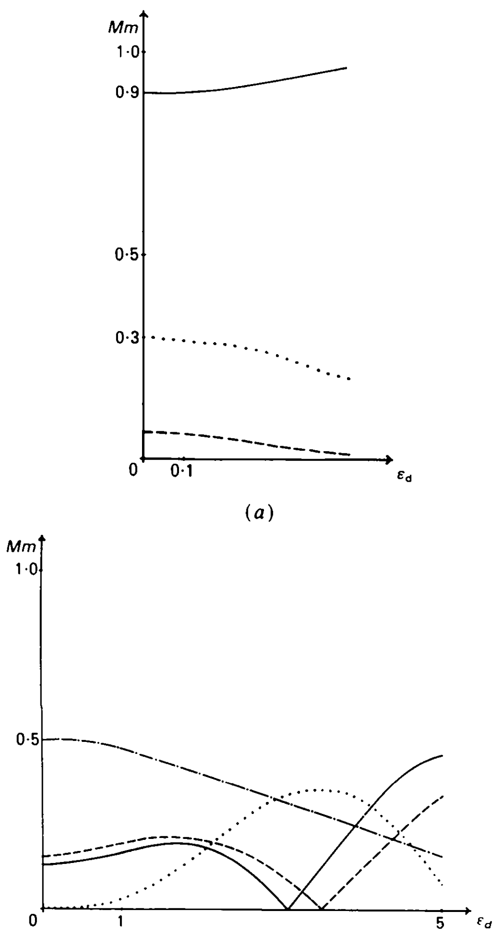

Fig. 3. ε_d dependence of M_m. (*a*) $\varepsilon_d + \varepsilon = 0{\cdot}5$ and (*b*) $\varepsilon_d + \varepsilon = 5$. Solid curve is for $m = 0$, dotted curve for $m = 1$, dashed curve for $m = 2$ and dot-and-dash curve for $m = 5$.

The fractional change of the structure factor is

$$\delta_m = \frac{M^t_m - M^r_m}{M^r_m} = -\frac{\pi^2}{6}\varepsilon_d^2 p_{1m} p_{2m}. \tag{37}$$

Since in the kinematical theory the intensity of the mth satelite is $R_m \sim M^2_m$, the fractional change of R_m is

$$\Delta_m = \frac{R^t_m - R^r_m}{R^r_m} = -\frac{\pi^2}{3}\varepsilon_d^2 p_{1m} p_{2m}. \tag{38}$$

Equations (28) and (29) show that M_m and R_m increase if s_{1m} and s_{2m} are of opposite sign, *i.e.* when the direction $\bar{s}_m$ lies within the directions $s_1 = 0$ and $s_2 = 0$, and decrease if s_{1m} and s_{2m} are of the same sign. This means that M_m increases when $-\varepsilon_{10} < m < \varepsilon_{20}$, and decreases otherwise. In the case when the initial model is of square-wave type, *i.e.* $\varepsilon_{10} = \varepsilon_{20} = \varepsilon_0/2$, then M_m increases if $|m| < \varepsilon_0/2$.

Thus, the structure factor of the principal maximum increases as a result of the interdiffusion, since the latter appears to reduce the superlattice to the crystal with averaged parameters. That is true if the thickness of the transition layer is small and, when the X-ray diffraction on the transition layer is significant, the structure factors will have an oscillatory behavior.

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