The Fourier transform is then given as

$$f(q_z) = S(q_z) * \sum_n \exp(2\pi i q_z nc)$$
$$= [\sin(\pi q_z Nc)/\pi q_z] * c^{-1} \sum_{g_z} \delta(q_z - g_z)$$
$$= \sum_{g_z} \sin[\pi(q_z - g_z)Nc]/\pi(q_z - g_z)c. \quad (21)$$

Equation (21) is identical with (19) since (18) equals (20) and hence

$$\sum_{g_z} \frac{\sin[\pi(q_z - g_z)Nc]}{\pi(q_z - g_z)c} = \frac{\sin(\pi q_z Nc)}{\sin(\pi q_z c)}. \quad (22)$$

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Investigation of Mosaicity of Epitaxic Multilayers by the Statistical Theory of X-ray Dynamical Diffraction

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Abstract

Based on the statistical theory of X-ray dynamical diffraction for thin films, the mosaicity of three types of semiconductor epitaxic layers has been investigated by analyzing their rocking curves by the X-ray double-crystal diffraction method. It is shown that the statistical theory can provide quantitative information on the mosaicity of the epitaxic layers such as the mean size and the mean disorientation of mosaic blocks in the layers. Some misunderstandings in interpreting experimental data are cleared up by taking into account the effect of diffuse scattering. It is emphasized that attempts to obtain structural parameters of specimens from their rocking curves by means of the Takagi–Taupin equations for coherent fields only are not strictly correct since diffuse scattering causes additional changes in the tails of the rocking curves.

1. Introduction

As is well known, X-ray double-crystal diffraction combined with computer simulations has been accepted as a nondestructive and sensitive method in the investigation of the structure of semiconducting epitaxic multilayers. Structural parameters, such as the period, thickness, composition, lattice mismatch and perfection of the epilayers, can be obtained by analyzing rocking curves (RCs). With the recent development of semiconductor techniques, device structures are becoming smaller and smaller. Therefore, it is important to characterize microdefects inside epitaxic layers and the state of interfaces because they influence the physical properties of materials and the quality of devices. The parameters describing the microdefects can be obtained quantitatively from X-ray diffuse scattering which has a contribution on the tails of the RCs. The key point is to

distinguish the diffuse scattering caused by the microdefects in the epilayer from the radiation coherently diffracted by the average crystal lattice.

Kato developed a statistical theory of dynamical diffraction to characterize the perfection of bulk crystals (Kato, 1980). Although the theory is applicable to general geometry, he dealt with the Laue case in detail. Bushuev (1989*a*,*b*) and Punegov (1991) modified the theory into a form suitable for the study of the Bragg case of X-ray double-crystal diffraction, in particular, for the study of multiple epilayers.

In this paper, the statistical theory of dynamical diffraction is applied to investigate the mosaicity of the epitaxic layers. Three types of device structures of III–V compound semiconducting materials were investigated by simulating their RCs using a curve-fitting method. The degree of perfection of these device structures as well as the mean size and the mean disorientation of mosaic blocks in them were determined. It is shown that the statistical theory is necessary to characterize the epilayers correctly.

2. Theoretical background

Real epitaxic layers often contain randomly distributed microdefects which cause local fluctuation in the lattice. The total displacement of atoms from their perfect lattice is given by (Bushuev, 1989*a*,*b*)

$$\mathbf{u} = \langle \mathbf{u} \rangle + \delta\mathbf{u}, \tag{1}$$

where $\langle \mathbf{u} \rangle$ is a statistically averaged lattice displacement relative to the substrate, representing the modulation of the lattice constant along the growth direction, which is assumed to be z, the normal of the crystal surface, and $\delta\mathbf{u}$ is the fluctuational displacement caused by microdefects.

The presence of the microdefects leads to X-ray incoherent (diffuse) scattering as well as coherent scattering. These two scatterings are recorded in double-crystal diffraction RCs. The amplitudes of the transmitted and diffracted waves $D_{0,g}$ in Takagi's equations (Takagi, 1969) are modified by

$$D_{0,g} = D^c_{0,g} + \delta D_{0,g}, \tag{2}$$

where $D^c_{0,g} = \langle D_{0,g} \rangle$ are coherent component fields, $\delta D_{0,g}$ are the fluctuations so that $\langle \delta D_{0,g} \rangle = 0$. Taking a statistical average of Takagi's equations, we obtain the following equations for coherent amplitudes:

$$\begin{aligned} \mathrm{d}D^c_0/\mathrm{d}z &= i a_{0_g} E \exp(i\mathbf{g}\cdot\langle\mathbf{u}\rangle)\Phi D^c_g - \mu D^c_0, \\ \mathrm{d}D^c_g/\mathrm{d}z &= i a_{g0} E \exp(-i\mathbf{g}\cdot\langle\mathbf{u}\rangle)\Phi^* D^c_0 - \mu^* D^c_g, \end{aligned} \tag{3}$$

where $\Phi = \exp(-i\psi z)$, $\psi = a_{00} - (a_{gg} - \eta)$, $\mu = a_{0g}a_{g0}(1 - E^2)\tau$, $\eta = -\pi\beta K/|\gamma_g|$, $\beta = -2\Delta\theta \sin 2\theta_B$, $a_{00} = \pi\chi_0 K/\gamma_0$, $a_{0_g} = -(\pi\chi_{-g}K/\gamma_0)C$, $a_{gg} = -\pi\chi_0 K/|\gamma_g|$ and $a_{g0} = (\pi\chi_g K/|\gamma_g|)C$.

The static Debye–Waller factor, $E = \langle \exp i\mathbf{g}\cdot\delta\mathbf{u} \rangle$, is introduced to characterize the degree of perfection of the layers. The complex quantity

$$\tau = \int_0^\infty \mathrm{d}\zeta \exp(-i\psi\zeta) G(\zeta) \tag{4}$$

is the correlation length of the lattice fluctuation. The statistical distribution of microdefects over the multilayers is determined by the correlation function

$$G(\zeta) = (1 - E^2)^{-1}(\langle \exp\{-i\mathbf{g}[\delta\mathbf{u}(\zeta) - \delta\mathbf{u}(0)]\} \rangle - E^2). \tag{5}$$

In numerical calculations of the intensity of the coherent scattering, (3) is reduced to a one-dimensional differential equation of type (Taupin, 1964)

$$\mathrm{d}Q/\mathrm{d}z = -iAQ^2 + BQ + iD, \tag{6}$$

where $Q = (D^c_g/D^c_0)\exp(i\mathbf{g}\langle\mathbf{u}\rangle)$, $A = (|\gamma_g|/\gamma_0)^{1/2} a_{0_g} E\Phi$, $B = (|\gamma_g|/\gamma_0)^{1/2}(\mu - \mu^*)$ and $D = (|\gamma_g|/\gamma_0)^{1/2} a_{0_g} E\Phi^*$. Equation (6) can easily be solved by a recurrent formula (Takagi, 1969; Mai, Cui & He, 1990). Therefore, starting from the interface between the substrate and the epitaxic layers, we can obtain the reflecting coefficient at the surface of specimens whose complex square will be the intensity of the coherent reflectivity. The substrate can be treated as an infinitely thick crystal, so the reflecting coefficient at the interface can be calculated by the dynamical diffraction theory.

In the Bragg geometry of X-ray double-crystal diffraction, both the diffuse absorption of the incoherently scattered waves and the coherent scattering of the diffuse waves can be neglected (Punegov, 1991). As a result, the intensity for the incoherently scattered wave $I^i_g = I_g - I^c_g$, where $I_g = \langle D_g D^*_d \rangle$ and $I^c_g = \langle D_g \rangle \langle D^*_g \rangle$, is obtained from the kinematic approach,

$$I^i_g = -2|a_{g0}|^2(1 - E^2) \int_0^l \tau_r \exp(-\mu_c z) I^c_0 \mathrm{d}z, \tag{7}$$

where μ_c is determined by photoelectric and diffuse absorption, l is the thickness of the layers and $\tau_r = \mathrm{Re}(\tau)$.

As in Kato's (1980) theory, the condition of application of the equations derived above has the form $\tau_r \ll \Lambda$, where Λ is the extinction distance.

We notice that when the thickness of a highly strained epilayer is larger than the critical thickness, misfit dislocations will be introduced. The presence of misfit dislocations will result in a local tilting of lattice planes. Thus, it can be assumed that a real epilayer consists of small mosaic blocks with dimension $l_0 \ll \Lambda$, disoriented with the angular distribution $W(\alpha)$ (something like a mosaic crystal with dislocations). Moreover, these blocks are randomly displaced relative to the average lattice. This argument was confirmed by our first-principle computer simulations and cross-section TEM observa-

tions (Cui, Mai, Wu, Wang & Dai, 1991). Assuming $W(\alpha)$ has a Gaussian distribution with an angular width Δ_m, we have (Bushuev, 1989*b*)

$$E = E_b/[1 + (\Delta_m/\Delta_0)^2]^{1/2} \quad (8)$$

and

$$\tau_r = (l_0\Delta_0/2\Delta)\exp[-\pi(\Delta\theta/\Delta)^2], \quad (9)$$

where E_b is determined by the root-mean-square displacement of the blocks, Δ_0 is the angular width of the diffraction spectrum of the individual block and $\Delta = [(\Delta_m)^2 + (\Delta_0)^2]^{1/2}$. Equation (9) is for a single layer. For multilayers, the value τ_r is a convolution of τ_r in (9) with the envelope function of the kinematic diffraction spectrum of the multilayers. In this case, τ_r may be larger than the thickness of each layer but smaller than the total thickness of the multilayers.

By fitting the measured rocking curves using a curve-fitting method, the structural parameters can be obtained with the aid of the above formulae. In analyzing the diffraction peak profile of our samples, we will neglect the diffuse scattering from the interface. This implies that the diffuse scattering from the defects in the epilayers is much stronger than the diffuse scattering from the interface roughness. When superlattices are relaxed, layers in the superlattice would be distorted to form mosaic blocks of some hundreds of nanometers (Cui, Mai, Wu, Wang & Dai, 1991). The size of the blocks may be larger than the thickness of one single lamella in the superlattices. For the superlattice sample studied, the average size of the blocks is about 400 nm, *i.e.* each block is composed of 5–6 lamellar segments. In this case, the diffuse scattering from the interface roughness is negligible when compared to that from the defects in the layers. For samples with a small number of interfaces, the diffuse scattering from the interface roughness is much weaker. Thus, it is justified for us to neglect the diffuse scattering from the interface roughness in this work.

3. Experimental results and discussions

Three types of device structures were investigated in this work. The experiments were carried out on X-ray double-crystal diffractometer with Cu $K\alpha_1$ radiation. The first crystal was Si(111) and the 004 reflection. All samples studied in this work were grown by molecular-beam epitaxy (MBE).

Fig. 1 shows a (004) diffraction rocking curve for $In_{0.47}Ga_{0.53}As$(300 nm)/InP single heterostructure. One can see a considerable increase of intensity in the tails of the layer peak in Fig. 1(*a*), which is due to the diffuse scattering. Comparing Figs. 1(*b*) and (*c*), one can see that attempts to obtain structural parameters of the specimens from rocking curves by means of the Takagi–Taupin equations for the coherent fields only are not strictly correct, since diffuse scattering causes additional changes in the tails of the curves. If the diffuse scattering is not taken into account, this effect may be incorrectly regarded as a modification of positive and/or negative strains in the layer. On the contrary, if it is assumed that the layer consists of mosaic blocks with a mean thickness $l_0 = 170$ nm and a mean disoriented angle $\varphi_0 = 115''$, the parameters $E = 0.62$ and $\tau = 355$ nm, which characterize the degree of perfection of the layer, are obtained. The theoretical rocking curve (Fig. 1*b*) fits the experimental one (Fig. 1*a*) very well.

Laser structures often consist of two layers with the same composition *A* above and below the active layer which has a different composition *B*. Recent experimental and theoretical studies showed that the wave fields in such an *ABA* structure exhibit an interference effect (Tanner & Hill, 1986). This effect may result in a splitting of the peak related to the two layers *A*. It is very sensitive to the composition and the thickness of layer *B*. This effect, however, may be strongly reduced by the presence of microdefects because the incoherent waves weaken the contrast of the interference fringes. Therefore, the structural parameters would be incorrectly estimated if the diffuse scattering was neglected. Fig. 2 shows an experimental rocking curve and the theoretical simulations of an $Al_xGa_{1-x}As$ *ABA* laser structure. In calculating Fig. 2(*b*), we assumed the structural parameters $Al_{0.467}Ga_{0.533}As$(900 nm)/$Al_{0.155}Ga_{0.845}As$ (95 nm)/$Al_{0.467}Ga_{0.533}As$(900 nm) with $E = 0.8$ and $\tau = 201$ nm, while in calculating Fig. 2(*c*) by means of the Takagi–Taupin equations, we assumed the parameters $Al_{0.47}Ga_{0.53}As$(900 nm)/$Al_{0.146}Ga_{0.854}As$(260 nm)/$Al_{0.47}Ga_{0.53}As$(750 nm). It is clear from the inset of Fig. 2 that the total diffracted intensity is the sum of the coherent and incoherent components simulated by statistical theory while the Takagi–Taupin equations only consider the coherent component. This is the reason why Fig. 2(*b*) fits Fig. 2(*a*) well. Therefore, the statistical theory gives us a correct estimate of the structural parameters.

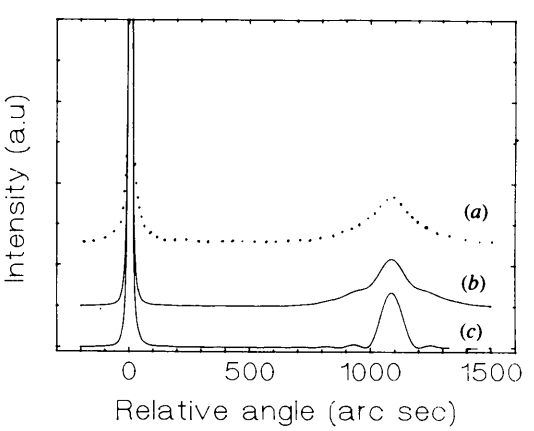

Fig. 1. (004) diffraction rocking curves of $In_{0.47}Ga_{0.53}As$/InP single heterostructure. (*a*) Experimental; (*b*) statistical theoretical simulation; (*c*) Takagi–Taupin-equation simulation.

The structures of superlattices are more complicated. Their diffraction patterns are more strongly influenced by diffuse scattering. An abrupt variation in the layer thickness and/or the composition leads to the superlattice subpeaks occurring in pairs or being accompanied by strong oscillation fringes (Li, Mai & Cui, 1993). However, the superlattice subpeaks will be broadened when the layers are imperfect. These broadened subpeaks would be incorrectly interpreted if one did not consider the presence of structural defects in the superlattices.

Fig. 3(*a*) is the experimental (004) diffraction rocking curve of a nominal $In_{0.15}Ga_{0.85}As(70\,Å)/GaAs(100\,Å)$ superlattice with 15 periods. It looks like a diffraction pattern from a superlattice with uniform structural parameters. This leads to the result, simulated by means of the Takagi–Taupin equations, $In_{0.12}Ga_{0.88}As(68\,Å)/GaAs(85\,Å)$ [see curve (*d*)]. But it is evident that the ±1st-order peaks in curve (*a*) are not the same. If we assume abrupt changes of thickness and composition in the superlattice, we obtain curves (*b*) and (*c*) by the statistical theory and by the Takagi–Taupin equations, respectively. Although the asymmetry of the ±1st-order satellite peak is predicted in Fig. 3(*c*), the difference between (*a*) and (*c*) is still great. However, Fig. 3(*b*) is in good agreement with the experimental data. The optimum parameters used in the simulation are: periods 1–7, $In_{0.12}Ga_{0.88}As(68\,Å)/GaAs(85\,Å)$; periods 8–15, $In_{0.14}Ga_{0.86}As(72\,Å)/GaAs(84\,Å)$; we obtain $E = 0.66$ and $\tau = 350\,nm$ for the superlattice as a whole. It is believed that the structural parameters derived from the statistical theory are closer to the real structure of the samples.

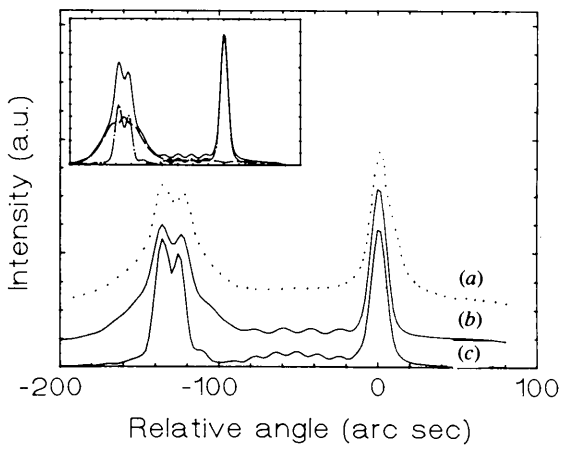

Fig. 2. (004) diffraction rocking curves of $Al_xGa_{1-x}As$ *ABA* laser structure. (*a*) Experimental; (*b*) statistical theoretical simulation; (*c*) Takagi–Taupin-equation simulation. Inset: the total diffraction intensity (solid line) is the sum of the coherent (dot-dashed line) and incoherent (dotted line) components.

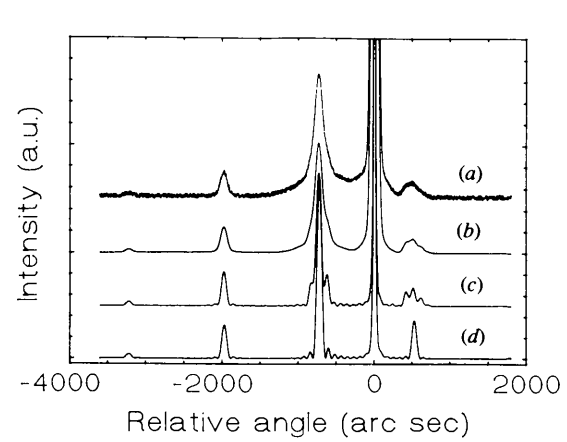

Fig. 3. (004) diffraction rocking curves of $In_xGa_{1-x}As/GaAs$ superlattice. (*a*) Experimental; (*b*) statistical theoretical simulation assuming an abrupt variation in structural parameters; (*c*) Takagi–Taupin-equation simulation with the same abrupt variation as (*b*); (*d*) Takagi–Taupin-equation simulation without abrupt variation.

4. Summary

The statistical theory of X-ray dynamical diffraction for thin films has been applied to characterize the mosaicity of epitaxic layers. The theory enables us to characterize quantitatively the perfection of the structures as well as thickness, composition and strain relaxation of the layers from the X-ray double-crystal rocking curves. The validity of the present analyzing method was confirmed by studying three types of semiconducting device structures. Some otherwise wrongly interpreted experimental data were corrected by taking into account the effect of diffuse scattering. Moreover, it was emphasized that attempts to obtain structural parameters of the device structures from RCs by means of the Takagi or Taupin equations for coherent fields only are not strictly correct since diffuse scattering causes additional changes in the tails of the rocking curves.

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