Dynamical x-ray diffraction from nonuniform crystalline films: Application to x-ray rocking curve analysis

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A dynamical model for the general case of Bragg x-ray diffraction from arbitrarily thick nonuniform crystalline films is presented. The model incorporates depth-dependent strain and a spherically symmetric Gaussian distribution of randomly displaced atoms and can be applied to the rocking curve analysis of ion-damaged single crystals and strained layer superlattices. The analysis of x-ray rocking curves using this model provides detailed strain and damage depth distributions for ion-implanted or MeV-ion-bombarded crystals and layer thickness, and lattice strain distributions for epitaxial layers and superlattices. The computation time using the dynamical model is comparable to that using a kinematical model. We also present detailed strain and damage depth distributions in MeV-ion-bombarded GaAs(100) crystals. The perpendicular strain at the sample surface, measured as a function of ion-beam dose (D), nuclear stopping power (S_n), and electronic stopping power (S_e) is shown to vary according to $(1 - kS_e)DS_n$ and saturate at high doses.

I. INTRODUCTION

The double-crystal x-ray diffraction (or x-ray rocking curve) technique is being widely used in the study of ion-implanted crystals,[1,6] superlattices.[2] MeV-ion-bombarded crystals,[3] epitaxial layers,[4] and thermally diffused crystals.[5] The x-ray rocking curve technique, which gives the sample reflecting power as a function of the angle between the sample surface and the incident x-ray beam, provides information on depth profiles of strain and damage for ion-implanted[1] or MeV-ion-bombarded[3] crystals, on layer thickness, composition, and strain for superlattices[2] and epitaxial layers,[4] and on diffusion profiles for thermally diffused crystals.[5]

To extract information, the x-ray rocking curve is analyzed using a kinematical[7] or a dynamical[6] diffraction theory. The kinematical theory, which is an approximation to the dynamical theory that neglects the extinction (i.e., multiple scattering), serves as an adequate model when the maximum reflecting power is less than about 6%.[7] When the reflecting power exceeds 6%, the dynamical theory should be used in the rocking curve analysis. The dynamical theory, however, has the shortcoming of taking too much computation time in the analysis.[6] In this paper, we present the dynamical theory in the layer approximation; this model requires computation time which is comparable to that by a kinematical model. This dynamical model is applied to the analysis of rocking curves taken from MeV-ion bombarded GaAs(100) crystals to get a detailed strain and damage depth distribution.

II. THEORY

The general theory of x-ray diffraction, which properly accounts for normal absorption (photoelectric process and Compton scattering) and extinction (coherent scattering or diffraction) of wave fields in a crystal medium, is called the dynamical x-ray diffraction theory. It is a first order theory in that it takes the deviation of the x-ray refractive index from unity up to first order, and the variation of the complex amplitudes over one x-ray wavelength to first order.[8] When the homogeneity of the crystal specimen varies in only one direction, i.e., in depth with no lateral variation, the change in the x-ray complex amplitudes in depth is well described by the Takagi–Taupin equation which was originally developed by Takagi[9] and independently by Taupin.[10] The Takagi–Taupin equation is:

$$\begin{aligned} i\frac{\lambda}{\pi}\beta_0\cdot\nabla\mathbf{D}_0(\mathbf{r}) &= \psi_0\mathbf{D}_0(\mathbf{r}) + \psi_H\mathbf{D}_H(\mathbf{r}), \\ i\frac{\lambda}{\pi}\beta_H\cdot\nabla\mathbf{D}_H(\mathbf{r}) &= \psi_0\mathbf{D}_H(\mathbf{r}) + \psi_H\mathbf{D}_0(\mathbf{r}) - \alpha_H\mathbf{D}_H(\mathbf{r}), \end{aligned} \tag{1}$$

where $\mathbf{D}_{0,H}(\mathbf{r})$ = complex amplitudes of the incident and diffracted waves, $\beta_{0,H}$ = wave vectors of the incident and diffracted waves, $\psi_{0,H} = -(e^2/mc^2)(\lambda^2/\pi)(F_{0,H}/V)$, $F_{0,H}$ = structure factors for the incident and diffracted waves, V = unit cell volume, and $\alpha_H \approx -2(\Theta - \Theta_B)\sin 2\Theta_B$.

The above equation expresses the spatial variation of the complex amplitudes of the incident and diffracted waves along the incident and the diffracted directions, respectively. The Takagi–Taupin equation has been used in the study of diffraction from curved crystals by Klar and Rustichelli.[11] They have presented the theory in a form useful for the present study, writing the depth dependent scattering amplitude in the following form:

$$i\frac{dX}{dA} = (1+ik)X^2 - 2(y+ig)X + (1+ik), \tag{2}$$

where $X = D_H(\mathbf{r})/\sqrt{b}\,D_0(\mathbf{r})$ = scattering amplitude, $b = |\gamma_0/\gamma_H|$, and $\gamma_{0,H}$ = direction cosines of the incident and diffracted waves with respect to the inward surface normal.

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$A = (\pi|\psi'_H|/\lambda\sqrt{|\lambda_0\lambda_H|})z,$

$\psi_{0,H} = \psi'_{0,H} + i\psi''_{0,H}, \quad z = \text{depth},$

$g = (1+b)\psi''_0/2|\psi'_H|\sqrt{b} = \text{absorption}, \quad k = \psi''_H/\psi'_H,$

$y = [(1+b)\psi'_0 - b\alpha_H]/2|\psi'_H|\sqrt{b}$

= deviation from the Bragg angle.

The damage reduces the structure factor to F_H.

$$F_H = F_H^0 \exp[-(8\pi^2 \sin^2\Theta_B/\lambda^2)u^2], \tag{3}$$

where F_H^0 = structure factor for the undamaged crystal and u = average atomic displacement.[7] The strain is taken into account through α_H.

$$\alpha_H = -2(\Theta - \Theta_B)\sin 2\Theta_B - (c_1\epsilon_1 + c_2\epsilon_2), \tag{4}$$

where

$c_1 = \cos^2\varphi \tan\Theta_B \pm \sin\varphi\cos\varphi,$

$c_2 = \sin^2\varphi \tan\Theta_B \mp \sin\varphi\cos\varphi,$

upper sign = for the incident angle of $\Theta_B - \varphi$ with respect to the sample surface, lower sign = for the incident angle of $\Theta_B + \varphi$ with respect to the sample surface, φ = angle between the surface and the reflecting lattice plane, ϵ_1 = strain perpendicular to the surface, and ϵ_2 = strain parallel to the surface.[2]

Larson and Barhorst have used Eq. (2) to obtain the strain depth distribution in an implanted and laser annealed silicon crystal.[6] They decomposed the complex differential equation (2) into two coupled real equations by putting $X = X_1 + iX_2$, and integrated numerically for each equation to obtain the rocking curve. We take, however, a different approach which takes less computation time.

Equation (2) can be integrated analytically[12]:

$$X(A) = \frac{sX_0 + i(B + CX_0)\tan[s(A - A_0)]}{s - i(C + BX_0)\tan[s(A - A_0)]}, \tag{5}$$

where

$X(A_0) = X_0, \quad B = -(1 + ik),$

$C = y + ig, \quad s = \sqrt{C^2 - B^2},$

and $|X(0)|^2$ = reflecting power at the sample surface.

In the analysis of rocking curves for ion-damaged crystals, the surface strained and/or damaged layer is divided into an arbitrary number of parallel laminae (layer approximation), and an average strain and/or damage is assumed in each layer [see Fig. 1(a)]. For a known scattering amplitude X at the interface of the bottom strained layer "1" and the undamaged and unstrained substrate crystal (i.e., the initial condition), one can calculate the scattering amplitude at the sample surface (i.e., at the top of the layer n) by repetitively applying the formula (5) from layer "1" to layer n. In the Bragg case, the initial condition of the scattering amplitude, which is given at the interface of the strained layer and the substrate, is an infinite crystal solution because the thickness of the substrate undamaged crystal is much larger than the x-ray absorption length in most cases. The initial condition is most easily obtained from Eq. (5) by the following consideration.

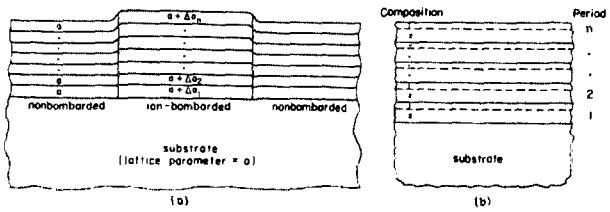

FIG. 1. (a) The surface strained/damaged region of an ion-irradiated crystal is divided into parallel laminae, in each of which the average strain and damage is assumed in the layer approximation. (b) Undamaged superlattice for which the formula (7) is used. The parameters P, Q, R, and T are the same in all periods.

Imagine a layer of thickness A_0 on an infinitely thick crystal. The layer and substrate crystals are assumed to be identical with no damage or strain. Then, "X_0" in Eq. (5) is the scattering amplitude of the substrate which is the infinite crystal solution, and "$X(0)$" at the layer surface will again be the infinite crystal solution because the layer crystal is identical with the substrate with no damage or strain. Thus, by putting $X(A) = X_0$ in Eq. (5) and solving for X_0 we get the infinite crystal solution,[14] which is

$$X_0 = \frac{-C \pm \sqrt{C^2 - B^2}}{B}. \tag{6a}$$

The lower sign in Eq. (6a) gives the correct infinite crystal solution. That is, the initial condition is

$$X_0 = -\frac{B}{C - \sqrt{C^2 - B^2}}. \tag{6b}$$

The rocking curve is calculated using Eqs. (5) and (6b) for given strain and damage in each layer, and the best fit of the calculated rocking curve to the experimental curve gives the strain and damage distribution in the layer approximation. Since the correct evaluation of s (which is a square root of complex variables) in Eq. (5) is important for the rocking curve calculation, we provide the formula in the Appendix. Equations (5) and (6b) can also be used for virgin or ion-damaged epitaxial layers of superlattices with suitable structure factors for each composition.

For virgin superlattices, the computation can be much simpler because the strain and structure factor is identical for all periods. The scattering amplitude X at the top of the layer which is the jth period from the substrate is given by

$$X_j = \frac{(P - iQ)X_{j-1} + (T - iR)}{(T + iR)X_{j-1} + (P + iQ)}, \tag{7}$$

where

$P = (B_1B_2 - C_1C_2)\tan(s_1A_1) + s_1s_2,$

$Q = C_1s_2\tan(s_1A_1) + C_2s_1\tan(s_2A_2),$

$R = s_1B_2\tan(s_2A_2) + s_2B_1\tan(s_1A_1),$

and

$T = (B_1C_2 - B_2C_1)\tan(s_1A_1)\tan(s_2A_2),$

X_0 is the infinite crystal solution, and A_1, A_2, B_1, etc., are for composition "1" and "2" [see Fig. 1(b)] and are defined in Eqs. (2) and (5). The parameters, P, Q, R, and T are the same for all periods, and thus Eq. (7) can be used for a very fast calculation of rocking curves for superlattices.

Finally, we present in Fig. 2 an experimental rocking curve (dashed) obtained from a virgin GaAs(100) crystal for Fe$K_{\alpha 1}$ (400) reflection, and a calculated rocking curve

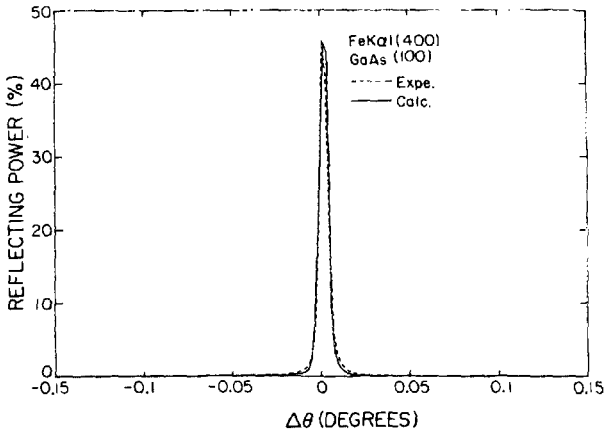

FIG. 2. Dynamical diffraction theory calculation of rocking curve. Experimental (dashed) and calculated (solid) curves are shown for an Fe$K_{\alpha 1}$ (400) reflection from a virgin GaAs(100) crystal. The formula (6b) was used for the calculation with 10-arcsec half-width Gaussian convolution.

(solid) from the formula (6b) with a 10-arc s half-width Gaussian convolution.

III. STRAIN/DAMAGE DISTRIBUTION IN MeV-ION-BOMBARDED GaAs(100) SINGLE CRYSTALS

Formulas (5) and (6b) were used for the analysis of rocking curves taken for GaAs single crystals bombarded with 15-MeV Cl ions to various doses at room temperature. The experimental rocking curve was taken by measuring the (400) reflection of Fe$K_{\alpha 1}$ radiation from ion bombarded (100)-cut GaAs at each angle which is varied in 0.0015-deg steps.

Figure 3 shows the experimental (dashed) and calculated (solid) rocking curves in the left column, and the depth distributions in the layer approximation of the perpendicular strain (solid) and average random atomic displacements (dashed) in the right column. The figure shows that the strain and damage in the surface layer saturate in a high-dose irradiation.

In the analysis of rocking curves measured for ion-damaged crystals, we choose our initial form of the strain and damage depth distributions based on the following two assumptions. The first assumption is that the strained/damaged layer thickness is the same as the ion range in the sample material. The second is that the strain/damage depth distribution follows the nuclear stopping power distribution; that is, the higher the nuclear stopping power, the higher the strain and damage.

From this starting point the initial distributions are varied so as to produce the best fit to the rocking curve data. In practice, the calculated rocking curve often needs to be convolved with a Gaussian function to fit the experimental curve. This is due to the mosaic structure of the crystal lattice due to the lattice disturbance from thermal vibrations and the lattice defects in ion-damaged crystals.

The strain at the sample surface, as obtained by the dynamical theory analysis, was measured as a function of beam dose (D), nuclear (S_n), and electronic (S_e) stopping powers by bombarding GaAs(100) crystals with various MeV ions (see Fig. 4). Figure 4 shows that the surface strain var-

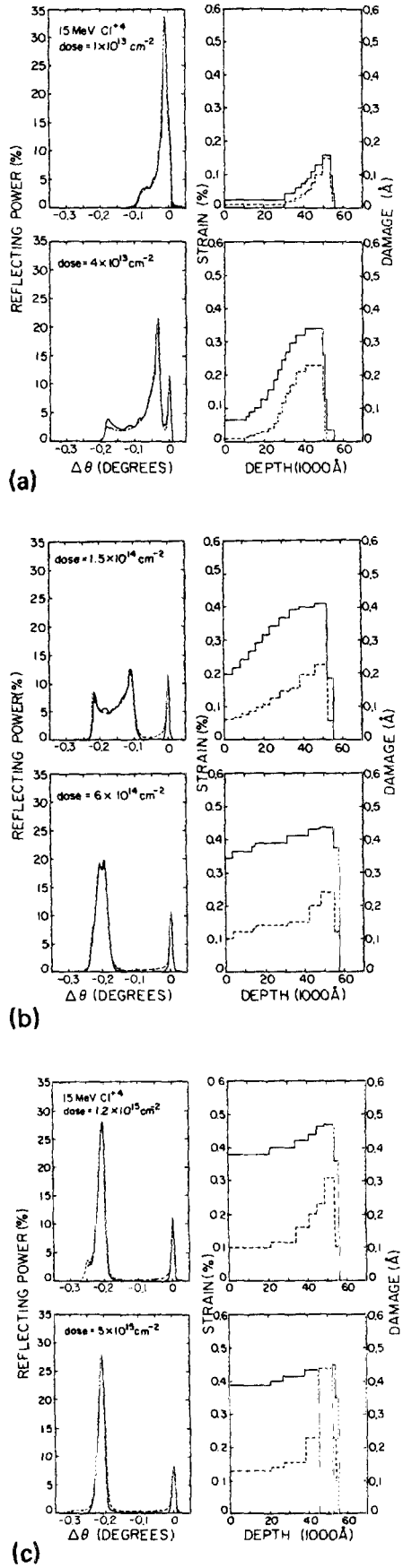

FIG. 3. Dynamical x-ray diffraction theory analyses of the GaAs rocking curves. Left-hand side: experimental (dashed) and calculated (solid) rocking curves. Right-hand side: strain (solid) and damaged (dashed) depth profile. Note that the layer at 5 μm depth has become amorphous at 5×10^{15} cm^{-2}.

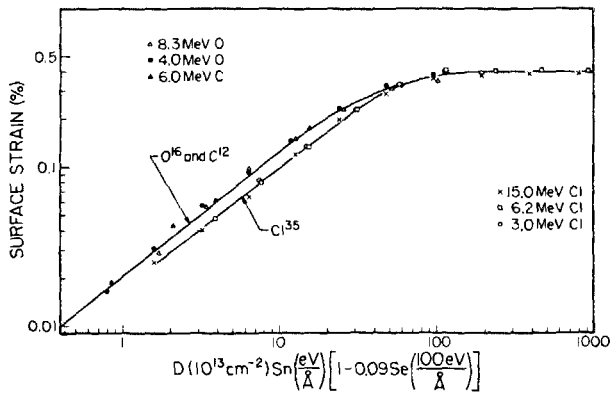

FIG. 4. The perpendicular strain at the surface of GaAs(100) crystals obtained by the dynamical theory analysis of the experimental rocking curves taken for (400) symmetric reflections of Fe$K_{\alpha 1}$ radiation from the GaAs(100) crystals bombarded with various MeV ions as shown in the figure.

ies with $(1 - kS_e)DS_n$ with $k = 0.09$ (appropriate units are given in the figure), and saturates to $\sim 0.4\%$ after a high-dose bombardment.

This result illustrates that the strain produced by nuclear collisions is partially annealed by the electronic collision processes. The saturation and stopping power dependence of strain has been explained in a separate paper by an ion-lattice single-collision model in which the point defect production and saturation is discussed for MeV-ion bombarded GaAs single crystals.[13]

ACKNOWLEDGMENT

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APPENDIX

Formula for evaluating $s = \sqrt{C^2 - B^2}$

$$s = s_1 + is_2 \quad \text{for } y < -q_1,$$
$$s = s_2 + is_1 \quad \text{for } -q_1 < y < q_2,$$
$$s = -s_2 + is_1 \quad \text{for } q_2 < y < q_1,$$
$$s = -s_1 + is_2 \quad \text{for } y > q_1,$$

where

$$s_2 = \sqrt{(r + r_1)/2},$$
$$s_2 = \sqrt{(r - r_1)/2},$$
$$q_1 = \sqrt{1 - k^2 + g^2},$$
$$q_2 = k/g,$$

and

$$r_1 = |y^2 - g^2 + k^2 - 1|,$$
$$r_2 = 2(yg - k),$$
$$r = \sqrt{r_1^2 + r_2^2}.$$

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