New Phase-Sensitive Method of Single-Crystal Characterization under X-Ray Diffraction Conditions

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A new phase-sensitive method for crystal lattice strain determination is proposed. The phase of the diffracted x-ray wave can be obtained from direct measurements of backscattered intensity. For this purpose we design conditions for creating a standing x-ray wave in a vacuum between two separated crystals. The measurement of the intensity of this wave as a function of the angular position of the crystal makes it possible to uniquely determine the relative phase of the wave scattered by a crystal with a deformed lattice. The experimental setup and some preliminary results are discussed. [S0031-9007(96)00220-7]

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X-ray Bragg diffraction is a very widely used nondestructive technique for single-crystal structure determination. Detailed analysis of the angular dependence of the scattered intensity distribution under the Bragg-diffraction condition provides information about the crystal lattice strain distribution with high precision and sensitivity. A recently developed model-independent method for determining the one- and two-dimensional crystal lattice strain [1–3] uses the angular dependence of the intensity in the vicinity of the Bragg reflection. The method is based on the solution of a one-dimensional inverse problem for single crystals [4]. The method has been successfully applied to silicon single crystals with ions implanted through a periodic oxide mask pattern and to SiGe/Si superlattices. There exists another model-independent method for the reconstruction of a density profile in thin films using anomalous x-ray reflectivity [5]. However, the method [5] does not retrieve the phase distribution of the diffracted x-ray wave along the scattering vector and, therefore, it does not allow one to determine the strain distribution in different crystallographic directions.

Most of the existing model-independent methods do not give a unique solution. Since the measured intensity does not carry any information about the phase of the scattered x-ray wave, we cannot *a priori* predict how many shifts of 2π occur in the phase of the diffracted wave [3]. Thus, the task for the inverse problem solution is to obtain unique information about the phase of the reflected wave. Actually, we need to know only the relative phase of the diffracted wave in comparison to the incident beam.

The most direct way to observe the phase relation between the incident and the diffracted beams is to arrange interference between them. The method of x-ray interferometry [6] allows one to arrange an experimental setup in such a way as to observe an interference pattern of the direct and double-reflected beam. This arrangement requires the use of a bulk single-crystal interferometer. Therefore, it is very difficult to apply this method to the problem of crystal lattice strain determination in real microelectronics structures. Another way to observe an interference pattern is to arrange the interaction of two beams having the same direction of propagation in a single crystal [7]. If we have an incident (reference) and scattered beam propagating in the same or exactly the opposite direction, it is possible to observe interference fringes due to the relative phase shift of the beams.

The present paper aims to describe a system where interference may be observed using separated crystals for the case of interference between the incident and the diffracted beams.

It can be shown that the conditions for having both Bragg and backward diffraction are the following:

$$2d_{hkl}\sin\theta = n\lambda\,, \tag{1a}$$

$$2d_{h_\perp k_\perp l_\perp}\cos\theta = m\lambda\,. \tag{1b}$$

Here, Eq. (1a) is the standard Bragg condition for constructive interference between different atomic planes with Miller indices (hkl); θ is the angle of incidence, and λ is the x-ray wavelength. Equation (1b) is the Laue condition for constructive interference of waves scattered by atomic planes which are orthogonal to the (hkl) planes. If both these conditions are valid simultaneously, there will be reflection in the backward direction.

Thus, we can describe the configuration where for a single incident x-ray beam there arise simultaneously three diffracted beams in three different directions: the standard Bragg case diffraction with intensity I_B; the exactly backward scattered diffraction with intensity I_b; and the standard Laue case diffraction with intensity I_L. Assuming a cubic lattice, $d_{hkl} = a_0/\sqrt{h^2 + k^2 + l^2}$ (interplanar distance of the Bragg planes) and $d_{h_\perp k_\perp l_\perp} = a_0/\sqrt{h_\perp^2 + k_\perp^2 + l_\perp^2}$ (interplanar distance of the corresponding Laue planes), where a_0 is the lattice constant and $n = m = 1$, we obtain an expression for the wavelength which satisfies both conditions (1a) and (1b), namely,

$$\lambda = \frac{2a_0}{\sqrt{(h + h_\perp)^2 + (k + k_\perp)^2 + (l + l_\perp)^2}}. \quad (2)$$

The condition (2) is written for the cubic lattice but can easily be extended to crystals with noncubic symmetry. The backreflection can be observed only from a sample with the proper azimuthal orientation and for a given radiation wavelength. Table I shows the radiation wavelengths for the observation of forward and backscattered reflections. They are given for the two main azimuthal orientations of the crystal surface.

To obtain the phase of the reflected wave we will consider a nonabsorbing crystal for simplicity. In the kinematical approximation, for the case of one-dimensional lattice distortions (we will be concerned with the case when the displacement vector has one component along the z axis, i.e., in depth), the amplitude reflection coefficient of diffracted x rays can be written in the form [4]

$$R(q) = \int_0^\infty \psi(z) \exp(iqz)\, dz\,, \quad (3)$$

where $\psi(z)$ is the structure factor of the distorted crystal and $q = (4\pi/\lambda)\Delta\theta \sin\theta$ is the length of the scattering vector; $\Delta\theta$ is the angular deviation from the exact Bragg position. Let us define $\psi_B(z)$ and $\psi_b(z)$ as the structure factors corresponding to the standard Bragg and backward reflections, respectively. Then,

$$R_B(q) = \int_0^\infty \psi_B(z) \exp(iqz)\, dz,$$
$$R_b(q) = \int_0^\infty \psi_b(z) \exp(iqz)\, dz\,. \quad (4)$$

For slowly changing atomic displacements $\mathbf{u}(z)$,

$$\psi_H(z) = \psi_H^{\text{perf}} \exp\{2\pi i \mathbf{H} \cdot \mathbf{u}(z)\}, \quad (5)$$

where $\psi_H^{\text{perf}} = \chi_H$ is the Fourier coefficient of the dielectric susceptibility of the perfect crystal for the given reflection $\mathbf{H} = (h, k, l)$. Therefore, using $\mathbf{H}_b \cdot \mathbf{u}(z) = \mathbf{H}_B \cdot \mathbf{u}(z) = |\mathbf{H}_B| \cdot u_z(z)$ we can rewrite the second equation of Eq. (4) as

$$R_b(q) = \alpha \int_0^\infty \psi_B(z) \exp(iqz)\, dz = \alpha R_B(q)\,, \quad (6)$$

where $\alpha = \chi_b/\chi_B$. Thus, the phase of the backscattered wave is *the same* as the phase of the standard Bragg-reflected wave. The measured intensity, $I_{I+b}(q)$, corresponds to the sum of the incident and backscattered beams,

$$I_{I+b}(q) = |1 + R_b(q)|^2 = 1 + |R_b|^2 + 2|R_b| \cos\phi_b\,, \quad (7)$$

where ϕ_b is the phase of the backscattered wave. Then, assuming the incident beam to be a plane wave with the amplitude equal to unity and the initial phase equal to zero, we obtain

$$\phi_B = \phi_b = \arccos\left\{\frac{I_{I+b} - \alpha^2 I_B - 1}{2\alpha\sqrt{I_B}}\right\}, \quad (8)$$

where ϕ_B is the phase of R_B and $I_B = |R_B|^2$. Thus, we have full information about the Bragg reflected wave, $R_B(q) = \sqrt{I_B} \exp i\phi_B$, and we can obtain the structure factor $\psi(z)$ via the Fourier transform of $R_B(q)$. Then we extract the phase of the complex function $\psi(z)$ which is proportional to the displacement distribution $u_z(z)$ [see Eq. (5)] and determine the strain profile $\epsilon(z) = du_z(z)/dz$. A more precise theory can be constructed by taking into account the absorption. Nevertheless, we can uniquely determine the phase of the Bragg-reflected wave from backscattered intensity via Eq. (8) (cf. Ref. [8]).

We observed interference between backreflected and incident beams in the experimental setup described in [1] (Fig. 1). There were three samples studied in that experiment: a Si(111) wafer with a periodic oxide mask pattern on the surface; a Si(111) wafer with a periodic oxide mask pattern on the surface and which had been implanted through the mask with 300 keV boron ions; and a Si(111) wafer which had been implanted through the mask with 300 keV boron ions and which had its oxide layer removed in HF acid (for more details see [1]). The experiment was carried out using a triple-crystal

TABLE I. Radiation wavelengths for the observation of Bragg, Laue, and back diffraction.

Bragg reflection	Laue reflection	Backreflection	Wavelength (Å)
Si(111)	Si($4\bar{2}\bar{2}$)	Si($5\bar{1}\bar{1}$)	2.0904
	Si($2\bar{2}0$)	Si($3\bar{1}1$)	3.2750
Si(022)	Si(400)	Si(422)	2.2172
	Si($0\bar{2}\bar{2}$)	Si(040)	2.7155
Si(400)	Si(040)	Si(440)	1.9201
	Si(022)	Si(422)	2.2172

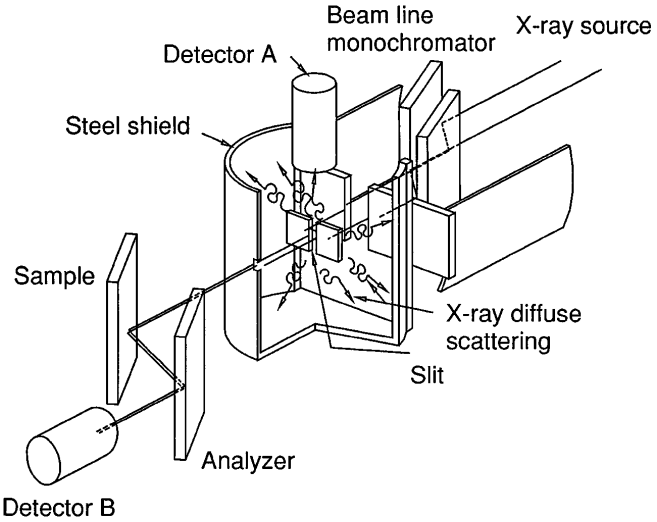

FIG. 1. Experimental setup for the observation of the incident-reflected beam interference described in [1].

diffractometer at beam line BL-14B (5 T superconducting vertical wiggler x-ray source) of the Photon Factory, a synchrotron facility at KEK in Tsukuba, Japan. The radiation wavelength was 0.138 nm. A symmetric Si(111) reflection was used in both the monochromator and analyzer crystals arranged in a nondispersive position with respect to the sample (111) planes in the Bragg-case geometry [1]. We used a scintillator NaI detector to measure the diffuse scattering from the slit which was employed to limit the size of the incident beam (detector A in Fig. 1). Assuming that the intensity of diffuse scattering on the slit is proportional to the primary beam intensity, we used this detector to monitor the primary beam intensity. As seen from Fig. 1 all possible effects of the fluorescence and thermal diffuse scattering from the sample were excluded, since the detector A was surrounded by a steel shield.

Figure 2 represents the experimental observations in the detectors A (thick line) and B (thin line). The detector A was expected to measure only the diffuse scattering intensity from the slit edges. However, it showed the dispersive shape of the monitor intensity while the sample angular position was close to the exact Bragg position. Similar curves were observed for all three samples. Assuming that there existed reflection back from the planes with the indices 651 for 0.138 nm radiation, we can conjecture that we have observed the interference fringes I_{I+b}. The sum of squared indices for the reflection close to 180° for the 0.138 nm radiation in silicon is 62, e.g., $h, k, l = 6, 5, 1$ closest to $\langle 110 \rangle$ or $h, k, l = 7, 3, 2$ closest to $\langle 211 \rangle$. There is no allowed reflection with this squared indices sum in a diamond lattice. The specimen azimuthal orientation was such that 651 reflection was more plausible than 732 reflection. Although Si(651) is a forbidden reflection in the ideal diamond-type lattice, in the case of a deformed crystal any reflection can be excited [9] since the lattice cell is not cubic and centrosymmetric anymore.

Unfortunately, the dispersive curves observed in the above experiment are not well suited for a quantitative analysis. First, it is difficult to determine the exact shape of the curves because of the inherent noise. It is much more reliable to use a transparent (e.g., gas proportional) detector instead of measuring the diffuse scattering from the slit edges. In the case of the use of an ionization chamber [2,3] the accuracy of the incident beam flux measurement is about 0.01%. In addition, the samples had a periodic surface modulation and the fundamental order was accompanied by a number of satellite reflections (see thin line on Fig. 2). Therefore, the dispersive curves, which correspond to the phase distribution in the fundamental diffraction order, were contaminated by contributions from satellite reflections. Below we will discuss the enhanced setup for the two-dimensional phase reconstruction.

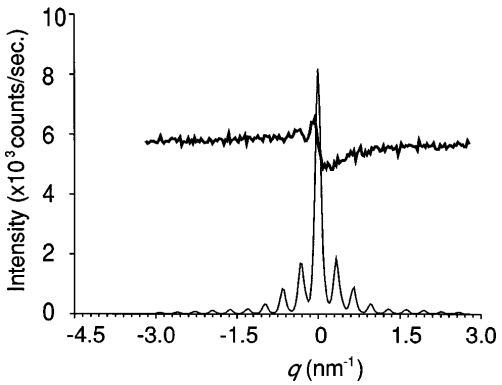

FIG. 2. Experimental intensity profiles for the experimental setup presented in Fig. 1 observed from the sample with the periodic oxide mask pattern in the detector A (thick solid line) and in the detector B (thin solid line). The intensity profile observed in detector B is divided by 10 for the better comparison.

The crucial point for the observation of interference fringes is the alignment of the incident and backscattered beams. In fact, the angular divergence of the incident beam can be taken into account by an additional phase shift of the reflected wave, $\phi = \beta x k$, where β is the angular coordinate, x is the coordinate across the wave propagation vector, and k is the wave number ($k = 2\pi/\lambda$). If the transparent detector has width S and the angular divergence of the incident beam is $\delta\theta$, then the intensity $I_{I+b}(q)$ averaged across the whole detector width and angular divergence of the incident wave is [see Eq. (7)]

$$I_{I+b}(q) = |1 + R_{\mathrm{av}}(q)|^2 = \left| \frac{1}{S\delta\theta} \int_0^S \int_{-\delta\theta/2}^{+\delta\theta/2} [1 + R_{\mathrm{al}}(q) \exp(i\beta x k)]\, d\beta\, dx \right|^2 = |1 + R_{\mathrm{al}}(q) A|^2, \qquad (9)$$

where $R_{\mathrm{al}}(q)$ is the reflection coefficient for precisely aligned and $R_{\mathrm{av}}(q)$ is the averaged reflection coefficient for the case of beam divergence. It is obvious that

$$A = \frac{1}{S\delta\theta} \int_0^S \int_{-\delta\theta/2}^{+\delta\theta/2} \exp(i\beta x k)\, d\beta\, dx \qquad (10)$$

does not depend on q and, therefore, $I_{I+b}(q)$ has the same shape as for the perfectly aligned direct and back-scattered beams. However, the contrast will be worse because A is less than unity. For the evaluation of the quantity A in Eqs. (9) and (10) the divergence of the backreflected beam is not important since at the fixed angular position q a crystal reflects radiation in an angular range not exceeding the incident beam divergence. Simple calculation for the realistic values of $S = 1$ mm and $\delta\theta = 7$ arcsec [Si (111) reflection, Cu $K\alpha$ radiation) gives the maximal and minimal values of fringe intensity fringes of +20% and

-10%, respectively. This calculation is quite consistent with the results presented in Fig. 2.

We have also provisionally calculated different profiles of I_{I+b} for three different phase distributions reconstructed from the experimental data that have been collected from the SiGe/Si superlattice [3] (Fig. 3). These three curves have very different shapes, while the Bragg rocking curve is absolutely the same for all three complex amplitude profiles [3]. This means that we can have the same function $|R_b(q)|$ while ϕ_b is different in Eq. (7).

Another plausible way to observe the interference between the direct and backscattered beams with high resolution in reciprocal space is to employ a crystal analyzer instead of a transparent detector. To avoid double reflection from both sides of the analyzer it is necessary to cut it in the form of a wedge. Because of the refraction effect the exact angles of the strong Bragg reflections will be different for the different sides of the crystal analyzer. If the following condition is valid,

$$\sqrt{\frac{\gamma_0}{|\gamma_h|}}\left(\frac{||\gamma_0| - |\gamma_h||}{|\gamma_0| \times |\gamma_h|}\right) > \frac{4\chi_h}{\chi_0}, \qquad (11)$$

where γ_0 and γ_h are cosines and χ_0 and χ_h are the Fourier coefficients of the dielectric susceptibility of the crystal for the incident and diffracted beams, respectively, then we have a semitransparent crystal mirror, which will reflect the coming of an x-ray beam incident from the front side and at the same time will not reflect a parallel beam incident from the exit side. We now arrange the crystal analyzer such that the scattered beam from the sample undergoes a Bragg reflection at the exit side, and such that the incident beam undergoes a coincident Laue reflection also at the exit side. By this means the resolution of I_{I+b} in reciprocal space may be significantly improved over measurements observed using either the diffuse scattering arrangement or the gas-proportional counter arrangement described here.

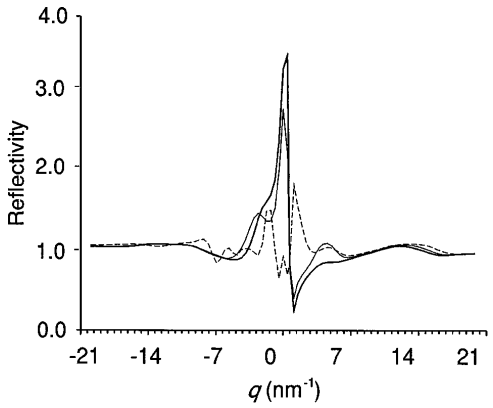

FIG. 3. Calculated profiles of I_{I+b} for three different phase distributions reconstructed from the experimental data collected from the SiGe/Si superlattice [3] for the perfectly aligned setup. Intensity profiles are calculated for the reflection amplitude without 2π shifts (thick solid line), with one 2π shift (thin solid line), and with three 2π shifts (dashed line) in the phase distribution [3].

Moreover, the suggested principle of obtaining information about the phase shift between the incident and diffracted beams can solve the problem of the relative phases of the satellite reflections in a two-dimensional inverse problem [1,2]. In the general case, each satellite reflection has unknown *a priori* phase relative to the fundamental order. Hence, without knowing signs and values of the relative phases of individual satellite reflections we have 2^{N-1} solutions, where N is the number of measured satellites. The satellite reflections measured through the crystal analyzer will carry the relative phase information. Thus, the problem of satellite relative phases can be solved uniquely.

It should be noted that the conditions presented could be applied for both Bragg or Laue geometry of diffraction from the sample. The authors are grateful to K. Nugent, S. Wilkins, C. Chantler, and T. Gureyev for fruitful discussions. We also acknowledge ARC grant support.

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