$(n_1n_2n_3n_4n_5n_6)$ can be written as

$$\begin{aligned} h &= n_1 + n_4 - n_5 \\ k &= n_2 + n_4 + n_5 \\ l_1 &= n_3 + n_6 \\ l_2 &= -2n_3 - n_6. \end{aligned} \tag{17}$$

Previously the diffraction spots of the σ' phase were indexed on the bases of the incommensurate composite structure (Cheng *et al.*, 1991). After carefully analyzing the simulated and experimental EDPs of the σ' phase, the sublattice parameter of substructure II along the c direction should be doubled to $c_2 = 7.42$ Å. Thus, the four indices hkl_1l_2 based on a phason-defected 1D quasicrystal are coincident with those based on the 1D incommensurate composite structure. This confirms once again that the description proposed in the present paper is equivalent to the regular description.

In principal, such a description is generally applicable for all 1D incommensurate composite crystals.

As a conclusion, the incommensurate composite σ' phase is described as the intermediate state between a fictitious 1D tetragonal quasicrystal and the commensurate σ phase. In other words, the incommensurate σ' phase and commensurate σ phase can be treated as a phason-defected 1D quasicrystal although the 1D quasicrystal is fictitious and has not been found in the Al–Cu–Fe alloy thus far. This implies that the incommensurate composite structure that shows two independent periodicities along the same direction may have some inherent relation with quasiperiodicity.

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Experimental Tests of the Statistical Dynamical Theory

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Abstract

A homogeneous distribution of SiO_2 precipitates in Czochralski-grown silicon containing different amounts of oxygen were produced by annealing the dislocation-free crystals at 1023 K. The resulting long-range strain field modifies the integrated reflecting power R of the Bragg reflections measured on an absolute scale with 316 keV γ-radiation. The thickness dependence of R has been modelled using the results of statistical dynamical theory. The assumption made in Kato's original theory, where the correlation length Γ for the wave-field amplitudes is proportional to the extinction length, has to be abandoned.

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Recent modifications to statistical dynamical theory by Becker & Al Haddad [*Acta Cryst.* (1990). **A46**, 123–139] lead to excellent agreement with the present experimental results. Furthermore, in the present case, the correlation length τ, describing short-range correlation in the fluctuations of the phase factor caused by the displacement field of the defects, turns out to be very small, so that the contribution of the mixed term to the integrated reflecting power could be neglected. Therefore, the defect scattering is characterized by the static Debye–Waller factor alone, which was determined accurately from the thickness dependence of the measured integrated reflecting power. From the ratio of the static Debye–Waller factors determined for different orders of reflection, the sizes of the SiO_2 precipitates have been calculated and the results are in very good agreement with the values determined directly from small-angle neutron scattering on the same samples.

Introduction

In X-ray crystal structure analysis, the electron charge density is, in general, deduced from the integrated intensities measured at a large number of Bragg reflections. This approach is straightforward in cases of weak scattering. After correction for normal absorption, one assumes that each atom in the crystal contributes equally to the scattered intensity and that multiple-scattering events are negligible, *i.e.* the diffraction data are interpreted within the kinematical theory. In most practical cases, however, these assumptions do not hold and the measured integrated intensity is smaller than the value expected from kinematical theory; the difference is called extinction. The search for the best possible correction of this effect has been an active field of research in crystallography since the early days of X-ray diffraction.

Any extinction correction needs a model to describe the defect structure of the actual sample in a statistical way. The mosaic model first suggested by Darwin (1922) has been the most successful so far. In this highly simplified model the imperfect crystal of thickness T_0 is assumed to be an aggregate of a large number of independently scattering plane-parallel perfect crystal blocks of thickness $t_0 \ll T_0$, tilted at small angles to each other. Their angular distribution is described by the mosaic distribution function, typically of Gaussian shape. There is a finite probability for the intensity scattered in one block to be scattered again in another block with identical lattice-plane orientation. This probability will increase with increasing sample thickness and decreasing mosaic spread. The rescattering of the Bragg-diffracted intensity leads to a reduction of the integrated reflecting power compared with the value expected from kinematical theory. The effect is called secondary extinction and is due to energy conservation during the scattering process. It is taken into account by solving Darwin's energy-coupling equations between incident and diffracted beams, which are the basis of most extinction correction schemes developed so far. A general solution of these equations is difficult because of the boundary conditions imposed by the sample and the mosaic blocks.

In a semi-infinite perfect crystal of plane-parallel-plate shape, the diffraction is rigorously described by dynamical theory, where the coherent interaction of all waves propagating in the crystal is taken into account; see, for example, Zachariasen (1945). In the limit of very thin crystals, the results obtained from dynamical theory are identical with those calculated from kinematical diffraction theory. In dynamical theory, a crystal is considered thin if its thickness is smaller than about half an extinction length Λ, which is inversely proportional to the wavelength and the structure factor. For 1 Å X-rays, a typical value of the extinction length is $\Lambda = 10$ μm. In perfect crystals with thicknesses of the order of an extinction length or larger, the integrated reflecting power is smaller than the value expected from kinematical theory. The difference is called primary extinction. In real crystals, perfect regions can easily reach dimensions of the order of 10 μm and therefore both contributions to extintion may become significant. It is the coexistence of primary and secondary extinction, *i.e.* of intensity and amplitude coupling in the diffraction process in real crystals, that complicated the development of extinction corrections in the past.

The conventional approach used to treat secondary extinction was developed by Hamilton (1957) and Zachariasen (1967*a*, *b*); empirical improvements have been suggested by Cooper & Rouse (1970). For plane-parallel crystal slabs Werner & Arrot (1965) and Werner, Arrot, King & Kendrick (1966) presented solutions of Darwin's energy-transfer equations for both Laue and Bragg geometry. Zigan (1970) discussed a formal solution for convex bound crystals. In crystallography, the most widely used extinction correction is due to Becker & Coppens (1974, 1975). Starting from the Tagaki–Taupin equations (Taupin, 1964; Tagaki, 1962, 1969) for a distorted crystal, Kato (1975, 1976) developed a dynamical approach to treat extinction that is not based on Darwin's energy-transfer equations and that covers mathematically the whole range from amplitude coupling in a perfect crystal to intensity coupling in a highly distorted crystal. The various theoretical schemes to correct for extinction developed up to 1977 have been reviewed by Becker (1977).

More recently, Sabine (1988) showed that the various extinction corrections derived on the basis of Darwin's mosaic-model and energy-transfer equations make closely similar predictions when identical functional forms for the coupling constant

in the transfer equations are used. Kulda (1987, 1988*a*) developed an extinction correction based on a random elastic-deformation model, where the imperfect crystal is supposed to consist of a stochastic ensemble of elastically deformed domains instead of the perfect mosaic blocks in Darwin's model. This model is more flexible and allows for a more realistic description of the coherent part of the wave interaction in imperfect crystals, *i.e.* a more realistic treatment of primary extinction. The approach has been tested and compared with the performance of the Becker–Coppens extinction correction (Becker & Coppers, 1974, 1975) using neutron diffraction data from steady-state reactors (Kulda, 1988*b*) and from pulsed neutron spallation sources (Jauch, Schultz & Schneider, 1989). The effect of the applied extinction correction on the calculated thermal parameters has been considered, in particular for variations introduced by reducing the size of the refined data set. In addition, the values of the mosaic-spread parameter obtained from the various refinements have been compared with the results from high-resolution γ-ray rocking-curve measurements performed on the sample studied with spallation-source neutrons.

Suortti (1982*a*) suggested a scheme to determine secondary extinction within Darwin's mosaic model from an analysis of the diffraction pattern measured on an absolute scale and the extrapolation to negligible primary extinction by varying wavelength and polarization – the factors that determine the amplitude coupling between coherent waves. The technique was tested on a parallel-sided crystal slab of beryllium using polarized Cu $K\alpha$ and unpolarized Mo $K\alpha$ radiations (Suortti, 1982*b*). All aspects of extinction corrections are much easier to handle if the diffraction experiments can be performed with γ-radiation with wavelengths of the order of 0.04 Å (Schneider, Hansen & Kretschmer, 1981; Hansen, Schneider & Larsen, 1984). If the dimensions of the sample are smaller than the extinction length of the strongest reflection for the wavelength used in the experiment, *i.e.* of the order of 1 or 10 μm, extinction is negligible (Bachmann, Kohler, Schulz & Weber, 1985; Höche, Schulz, Weber, Belzner, Wolf & Wulf, 1986). The potential of the extrapolation of data measured on a given sample to the limit of 'zero extinction' has been discussed in a series of papers (Mathieson, 1977*a*; Lawrence & Mathieson, 1977; Mathieson, 1977*b*, 1979).

In 1980, Kato published two fundamental papers on the statistical dynamical theory of crystal diffraction (Kato, 1980*a*, *b*), which are extensions of his earlier papers on extinction (Kato, 1976). Here, the integrated reflecting power is given as a sum of a coherent term, R_{coh}, which is closely related to the expression obtained from dynamical theory for perfect crystals, an incoherent term, R_{inc}, and a mixed term, R_{mix}:

$$R_{calc} = R_{coh} + R_{mix} + R_{inc}. \quad (1)$$

The degree of perfection of the crystal is described by a parameter E for long-range correlation and an effective correlation length τ_e for short-range correlation. The latter depends on the correlation length Γ for the wave-field amplitudes and the short-range correlation length τ for the phase factor

$$\varphi(\mathbf{r}) = \exp[2\pi i \mathbf{H}\cdot\mathbf{u}(\mathbf{r})], \quad (2a)$$

with $\mathbf{u}(\mathbf{r})$ representing the local displacement field and $\mathbf{H}$ the reciprocal-lattice vector. E is equal to the space average of the phase factor

$$E = \langle\varphi(\mathbf{r})\rangle_r, \quad (2b)$$

which is identical with the definition of the static Debye–Waller factor. Kato assumes that Γ is proportional to the ratio of the extinction length Λ, defined by (6*a*) later in this paper, and E. Γ is thus wavelength-dependent and is considered to be much larger than τ. A number of experiments have been performed to test Kato's statistical dynamical theory or to gain information on the defect structure in real crystals applying the theory as it stands.

Using thermal neutrons, the integrated reflecting power has been measured as a function of wavelength in Laue geometry on a high-quality $Y_3Fe_5O_{12}$ (YIG) crystal for both polarization states of the sample. *Pendellösung* oscillations have been observed for both polarization states that qualitatively agree with the predictions made using the dynamical theory for perfect crystals (Baruchel, Guigay, Mazuré-Espejo, Schlenker & Schweitzer, 1982; Guigay, Schlenker & Baruchel, 1982). Concerning the flipping ratio, there is a marked difference between experiment and dynamical theory, however, after fitting the parameters E and τ; Kato's statistical dynamical theory describes the experimental data well.

Pendellösung oscillations were first studied with X-rays by Kato & Lang (1959) on wedge-shaped perfect silicon crystals. Using thermal neutrons, Sippel, Kleinstück & Schulze (1965) measured in Laue geometry the integrated reflecting power as a function of the thickness of plane-parallel crystal slabs. With an initial thickness of 204 μm, the silicon wafer was successively etched down to a thickness of 6 μm and the theoretically predicted *Pendellösung* oscillations were observed. These oscillations can be measured more easily by tilting the sample around the reciprocal-lattice vector so that the effective sample thickness in the experiment varies. This technique was suggested by Lawrence & Mathieson (1977) and may be called the inclination method. It was first applied in neutron diffraction, where *Pendellösung* oscillations have been observed in this way on a sufficiently perfect Ge single crystal (Somenkov, Shilstein, Belova & Utemisov, 1978). Measurements

of the same kind were performed on silicon single-crystal wafers with dislocation densities up to $100\,mm^{-2}$ and on wafers with dislocation densities above $5000\,mm^{-2}$ (Olekhnovich, Karpei, Olekhnovich & Puzenkova, 1983). Olekhnovich *et al.* find the extinction corrections based on Darwin's energy-transfer equations unsuitable to describe their experimental data. The results from the low-dislocation-density wafers can be approached using Kato's statistical dynamical theory (Kato, 1980*a*, *b*). Since the incoherent and mixed terms in Kato's expression turn out to be small, Olekhnovich *et al.* assumed that only the coherent component can be affected significantly by anomalous scattering and absorption and they applied a correction scheme due to Kato (1968). However, there is still a marked difference between the experimental data and the best fit obtained with Kato's theory. The data taken at the high-dislocation-density wafers cannot be explained, even qualitatively, using Kato's statistical dynamical theory (Kato, 1980*a*, *b*).

Oscillations of the integrated intensity with respect to sample thickness and the anomalous transmission of plane-polarized Mo $K\alpha$ radiation have been studied on dislocation-free silicon crystals containing oxygen by Olekhnovich & Karpei (1984). They consider only coherent and incoherent components of the integrated reflectivity and, from measurements at different orders of reflection, they estimate the type and the size of the defects. Kato's theory has been applied to determine the concentration and the mean size of microdefects in silicon crystals from *Pendellösung* oscillations measured with σ-polarized Mo $K\alpha$ radiation (polarization factor $C = 1$) by using the inclination method on as-grown samples with oxygen concentrations in the range 10^{16}–10^{18} O atoms cm^{-3} (Voronkov, Chukhovskii & Piskunov, 1985; Voronkov, Piskunov, Chukhovskii & Maksimov, 1987). From their analysis, Voronkov *et al.* (1985, 1987) claim the existence of microdefects with an average radius of the order of 70 Å and a concentration of about $10^{13}\,cm^{-3}$ in their as-grown silicon crystal containing 10^{18} O atoms cm^{-3}. The observed variation of the correlation length τ with tilt angle is interpreted as an indication of some anisotropic nature of the displacement field of the defects.

Sugita, Sugiyama, Iida & Kawata (1987) took section topographs from 420 μm thick silicon wafers containing 1.15×10^{18} O atoms cm^{-3}. The intensity distribution measured on a sample that had been annealed for 12 h at 1223 K was fitted with Kato's statistical dynamical theory and static Debye–Waller factors were determined for seven different reflections. They clearly show the expected dependence on the square of the scattering vector. In a subsequent paper, Iida, Sugiyama, Sugita & Kawata (1988) report investigations of seven Si samples that had been annealed at 1073 K for times varying between 50 and 1000 h. In a second series of measurements, samples were annealed for 100 h at six different temperatures between 923 and 1173 K. Again, static Debye–Waller factors have been determined by fitting Kato's theory to the intensity distribution measured in section topographs. Referring to theoretical work by Dederichs (1973), Iida *et al.* (1988) determine the size and the number density of the oxygen precipitates.

As shown for the first time in a neutron diffraction study on highly perfect silicon crystals, *Pendellösung* oscillations can be measured with high accuracy by varying the wavelength of the diffracted radiation (Shull, 1968; Shull & Oberteuffer, 1972). Takama, Harima & Sato (1990) measured *Pendellösung* intensity beats with white X-radiation ($0.15 < \lambda < 0.8$ Å) on parallel-sided heat-treated Czochralski-grown silicon wafers using a solid-state detector. The technique has been described in detail by Takama, Iwasaki & Sato (1980) and Takama, Noto, Kobayashi & Sato (1983). Defects have been introduced by heating the 0.5 mm thick wafers in an argon atmosphere for times varying between 24 and 100 h at temperatures in the range from 1123 to 1273 K. The experimental data measured for different orders of reflections have been analysed with Kato's statistical dynamical theory and good agreement was found if the correlation length Γ for the wave-field amplitudes is treated as an additional independent fitting parameter. It turns out that the two correlation lengths τ and Γ are of the same order of magnitude, approximately 0.5 μm, which is in contrast to the original assumption made by Kato (1980*a*, *b*).

Stimulated by the experimental evidence of problems existing within the statistical dynamical theory, a number of authors have made improvements on Kato's original approach. Guigay (1989) derived the expressions for the integrated intensities from transfer equations requiring simpler mathematics and he found two errors in the original formulae for the mixed term of the integrated reflecting power. These findings are in agreement with the results obtained by Al Haddad & Becker (1988), who revised Kato's original treatment and compared their modified results with the experimental *Pendellösung* data from silicon single crystals containing dislocations (Olekhnovich *et al.*, 1983). The modified theory describes the experimental data much better than Kato's expressions; a variation of the correlation length Γ for the wave-field amplitudes improves the statistical agreement factor for the fit only slightly. In a subsequent paper, Becker & Al Haddad (1989*a*) showed that, in contrast to Kato's assumption, the correlation length Γ varies within the sample and is of the same order of magnitude as the correlation length τ for the phase factor.

To establish a fruitful interplay between experiment and further theoretical development of statistical

dynamical theory, experiments should probe the different aspects of the theory in a selective way and should be performed on samples with well characterized homogeneously distributed defects. The measurement of *Pendellösung* oscillations on annealed Czochralski-grown silicon single crystals by means of the inclination method and 316.5 keV γ-radiation is an experiment of that category. Because of its technological importance, the defect structure created by annealing of Czochralski-grown silicon single crystals containing about 20 O atoms in 10^6 at temperatures between 873 and 1573 K has been studied extensively by combining different techniques such as infrared spectroscopy and chemical etching coupled with optical microscopy (Livingston, Messoloras, Newman, Pike, Stewart, Binns, Brown & Wilkes, 1984), as well as by a variety of diffraction techniques such as electron microscopy (Bourret, 1987), small-angle neutron scattering (Bergholz, Binns, Booker, Hutchison, Kinder, Messoloras, Newman, Stewart & Wilkes, 1989), X-ray topography (Patel & Authier, 1975; Partanen & Tuomi, 1990), X-ray anomalous transmission (Patel & Batterman, 1963), γ-ray diffractometry (Kurbakov, Rubinova, Sobolev, Trunov & Shek, 1986), neutron backscattering (Magerl, Schneider & Zulehner, 1990), X-ray diffuse scattering (Patel, 1975; Dietrich & Zaumseil, 1985; Stojanoff, Pimentel, Bulla, Castro, Hahn & Ponce, 1986) and, recently, by diffraction of high-energy synchrotron radiation (Schneider, Nagasawa, Liss, Magerl & Zulehner, 1991). In the context of the present paper, it is emphasized that investigations with γ-rays, neutrons and high-energy synchrotron radiation can be performed on the same sample. *Pendellösung* measurements with 316.5 keV γ-radiation probe the basic concepts of statistical dynamical theory since absorption is very weak, anomalous dispersion does not occur and, owing to the small Bragg angles of typically 1°, the diffraction geometry is quasi-one-dimensional and polarization effects can be neglected. At present, theoretical efforts are being made to understand the role of the correlation length Γ for the wave-field amplitudes and to develop more appropriate expressions for it (Becker & Al Haddad, 1989*a*, *b*, 1990). As mentioned above, Kato assumes Γ to be proportional to Λ/E. If this assumption is wrong, a γ-ray experiment should reveal it clearly, because here the extinction length is larger by a factor of 40 than for the case of Cu $K\alpha$ radiation. Therefore, the γ-ray *Pendellösung* data should allow for a stringent test of the reliability of suggested new expressions for Γ.

The outline of this paper is as follows. Firstly, the technique of measuring *Pendellösung* intensity beats in large silicon single crystals using 316.5 keV γ-radiation is described, followed by a discussion of the origin of its sensitivity to weak diffuse scattering from point defects. Secondly, the mechanism of the formation of oxygen precipitates in dislocation-free silicon at temperatures around 1023 K and of dislocation loops and/or stacking faults at temperatures above 1173 K is described with emphasis on the expected homogeneity of the distribution of these defects in the crystal. Thirdly, the expressions for the integrated reflecting power as given by Kato (1980*a*, *b*) in his original papers, by Guigay (1989) and by Becker & Al Haddad (1989*b*, 1990) are presented. They are applied to fit the γ-ray *Pendellösung* data, which were taken in two groups of samples containing 7.5×10^{17} and 18×10^{17} O atoms cm^{-3}, respectively. The relative success of the various fits to these data represents the basis for the concluding discussion on the present state of the statistical dynamical theory of diffraction.

γ-ray *Pendellösung* intensity beat measurements

In the angular range in which Bragg diffraction occurs, a monochromatic X-ray beam excites in a perfect crystal two interfering wave fields with different wave vectors defined by the branches of the dispersion surface for each polarization state. This difference is much smaller than the length of the wave vectors. In Laue (transmission) geometry, it leads to oscillations of the intensity of the diffracted beam as a function of sample thickness. The period of these oscillations is called the *Pendellösung* length and is of the order of 1 mm for diffraction of 316.5 keV γ-radiation at low-order reflections of silicon. The measurement of such *Pendellösung* oscillations on an absolute scale is straightforward with γ-radiation because absorption is as weak as in neutron diffraction, so that samples of thickness of the order of 1 cm can be studied by means of the inclination technique. The thickness of such crystals can be measured very accurately. No anomalous dispersion occurs and, because of the small Bragg angles, the difference in the diffraction process for the two polarization states is extremely small and can be neglected in most cases. The half-life of the ^{192}Ir γ-ray source is 74.2 d, so the data can be corrected accurately for time decay of the source. γ-ray *Pendellösung* intensity beats have been measured in highly perfect float-zone-grown silicon crystals to check the reliability of the Thomson cross section for the interaction with matter of γ-radiation with energies in the range between 300 and 460 keV (Graf & Schneider, 1986). In this way, the 220 structure factor was measured with an accuracy of the order of ±0.05% and the agreement with the results from X-ray *Pendellösung* measurements is excellent. The experimental arrangement and the data analysis are described in detail by Graf & Schneider (1986).

The experimental data are corrected for absorption using the linear absorption coefficient $\mu_0 = 0.25(2)$ cm^{-1} measured with 0.0392 Å γ-radiation (Graf & Schneider, 1986). They can then be compared

with theoretical quantities derived for zero absorption. In the context of the present paper, the integrated reflecting power for a perfect plane-parallel single crystal in symmetrical Laue geometry is sufficiently well described by an expression given by De Marco & Weiss (1965) neglecting anomalous dispersion,

$$R_{dyn}(A) = QTW(2A) \quad \text{with} \quad Q = \lambda/[\Lambda^2 \sin(2\theta_B)], \tag{3}$$

where Q is the integrated reflecting power per unit volume in the kinematical limit. $W(x)$ represents the Waller integral,

$$W(x) = \int_0^1 J_0(x\rho)\,\mathrm{d}\rho = (1/x)\int_0^x J_0(y)\,\mathrm{d}y \quad \text{with} \quad y = x\rho, \tag{4}$$

where $J_0(x\rho)$ is the zeroth-order Bessel function of the first kind. $W(x)$ tends to 1 for large arguments x. Hence, in the present geometry, the kinematical integrated reflecting power of a crystal of thickness T_0 is equal to

$$R_{kin} = QT \quad \text{with} \quad T = T_0/\cos\theta_B, \tag{5}$$

$$A = T/\Lambda, \tag{6a}$$

$$\Lambda = V_{cell}/[C\lambda r_0 |F| \exp(-M)] = T_{ext}. \tag{6b}$$

The extinction length Λ is defined as the sample thickness corresponding to $A = 1$; the quantity A is thus a measure of the sample thickness in units of the extinction length. The polarization factor C may be put equal to unity since the Bragg angles are small in diffraction experiments with 0.0392 Å γ-radiation. r_0 is the classical electron radius, F the structure factor, $\exp(-M)$ the thermal Debye-Waller factor and V_{cell} the volume of the unit cell. λ represents the wavelength and θ_B is the Bragg angle.

If *Pendellösung* oscillations are determined by means of the inclination technique (Lawrence & Mathieson, 1977), the effective sample thickness, $T_{eff} = T/\cos\psi$, is varied by tilting the crystal plate around the scattering vector and the integrated reflecting power is measured for each tilt angle ψ.

Annealed Czochralski-grown silicon crystals

Dislocation-free silicon crystals used for the production of VLSI electronic devices are almost exclusively manufactured by means of the Czochralski (Cz) technique. During crystal growth, the silica crucible partly dissolves into the silicon melt and, consequently, oxygen is incorporated into the growing crystal. Oxygen is the most important impurity in Cz silicon crystals, which typically contain between 5×10^{17} and 1.5×10^{18} O atoms cm^{-3}, but only about 7.2×10^{12} C atoms cm^{-3}. In the dissolved state these O atoms are located at interstitial sites of the silicon matrix, occupying distorted bond-centred positions.

Upon heat treatment at temperatures above 823 K, O atoms diffuse through the lattice to produce small agglomerates that grow into precipitate particles of silica (SiO_2). The tendency to form SiO_2 precipitates depends strongly on the oxygen concentration, but also on the concentration of other impurities such as carbon. The precipitation of oxygen in dislocation-free silicon at 1023 K has been studied systematically by means of small-angle neutron scattering (SANS) (Messoloras, Schneider, Stewart & Zulehner, 1989; Gupta, Messoloras, Schneider, Stewart & Zulehner, 1990). For example, after annealing for 216 h a sample with an oxygen content of 7.44×10^{17} O atoms cm^{-3} contained 8.5×10^{11} cm^{-3} plate-shaped SiO_2 precipitates of dimensions $500\times500\times50$ Å.

The SiO_2 precipitates create strain in the dislocation-free silicon matrix, which gives rise to diffuse scattering. At temperatures above 1173 K, the silicon lattice becomes sufficiently soft that these strain fields can relax by emission of dislocation loops and/or self-interstitials, which subsequently agglomorate to stacking faults on {111} planes. These effects have been studied on large silicon crystals by high-resolution double-crystal γ-ray diffractometry (Schneider, Gonçalves, Rollason, Bonse, Lauer & Zulehner, 1988) and more recently by using a three-crystal diffractometer and 150 keV synchroton radiation (Schneider, Nagasawa, Liss, Magerl & Zulehner, 1991). Complementary information on the defect scattering has been obtained from neutron back-scattering experiments performed on the same samples (Magerl *et al.*, 1990; Liss, Magerl, Schneider & Zulehner, 1991). Because of the expected statistical nature of the defects, Cz silicon crystals, heat treated at temperatures above 1273 K, have been used to test extinction models based on Darwin's mosaic model (Schneider, Gonçalves & Graf, 1988).

In the present paper, [001] grown silicon crystals containing approximately 7.7 and 18×10^{17} O atoms cm^{-3} that have been annealed at 1043 and 1023 K, respectively, are studied. They should contain different amounts of SiO_2 precipitates of different shape and size but no dislocations. The sample characteristics are given in Table 1. Fig. 1 shows *Pendellösung* intensity beats measured with the inclination method and 0.0392 Å γ-radiation on sample II.2 annealed for 70 h at 1043 K. The observed *Pendellösung* oscillations show the period expected from dynamical theory for a perfect crystal, however, they are approximately 50% higher than the average value of $R_{dyn}(A)$. The experimental data approximate a straight line and can be well described by a superposition of $R_{dyn}(A)$ and a term proportional to the parameter A and thus to the effective sample thickness T_{eff}. The results shown in Fig. 1 suggest that the SiO_2 precipitates do not affect the coherent wave fields in the crystal; they only create some weak diffuse

Table 1. *Characterization of the* ⟨001⟩ *Czochralski-grown silicon crystal discs, of diameter* 10 cm, *which were annealed in an argon atmosphere*

All samples are doped with boron and their substitutional carbon content is less than 0.1×10^{17} C atoms cm^{-3}. The distribution of dissolved oxygen shows radial symmetry with respect to the growth direction, but the oxygen content in sample VII varies from the centre to the edges of the sample: z corresponds to the centre of the crystal plate, $r/2$ to a distance half-way between the centre and the edge and r-5 to a position 0.5 cm away from the edge of the crystal.

Sample	Annealing procedure	Resistivity (Ω cm)	Density of interstitial O atoms ($\times10^{17}$ cm^{-3}) z	$r/2$	r-5
II.2	70 h at 1043K	≥23	7.7	-	-
VII.1	24 h at 1023K	≥15	19.1	16.1	11.4
VII.2	72 h at 1023K	≥15	18.6	16.0	11.4
VII.3	216 h at 1023K	≥15	18.1	15.8	11.5

scattering, which adds to the integrated reflecting power calculated from dynamical theory.

It is interesting to note that the total thickness of the sample contributes to the observed defect scattering, whereas the integrated reflecting power related to the coherent part of the Bragg peak reaches its average value already at $A\simeq0.5$, which, in the present case, corresponds to a penetration depth of $T\simeq$ 0.01 cm, *i.e.* if the effect of the *Pendellösung* oscillations is neglected, after correction for absorption, a sample with $T\simeq0.01$ cm would show the same

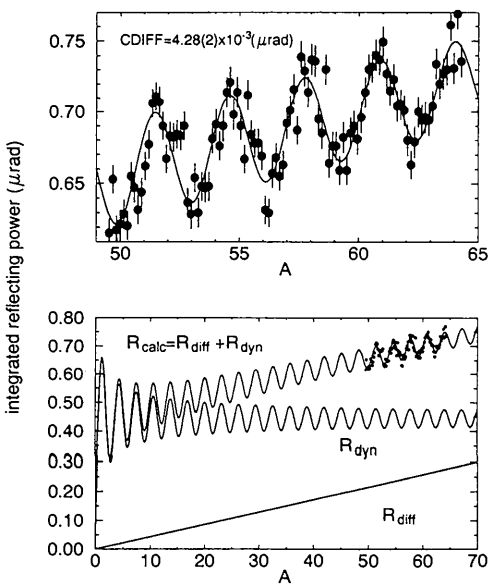

Fig. 1. *Pendellösung* intensity beats measured with 0.0392 Å γ-radiation for sample II.2 containing 7.7×10^{17} O atoms cm^{-3} after annealing for 70 h at 1043K. Comparison is made with the integrated reflecting power $R_{220}^{dyn}(A)$, calculated from dynamical theory, and its superposition with a scattering term R_{diff} proportional to the parameter A and thus the sample thickness. CDIFF is the proportionality constant.

integrated reflecting power as a perfect crystal with a thickness of $T\simeq1$ cm. Therefore, the diffuse scattering from nearly perfect crystals is very much enhanced with respect to R_{dyn}. The same argument holds for the weak thermal diffuse scattering (TDS), which is defined as a fraction of the kinematical integrated reflecting power, $R_{kin}\propto T$. However, model calculations showed that, for Si 220 at room temperature, the effect of TDS is not visible in the dependence of the integrated reflecting power as measured using 0.0392 Å γ-radiation.

Because the formation of SiO_2 precipitates depends strongly on the oxygen concentration, which shows some growth-induced fluctuations in standard Cz silicon crystals, a certain inhomogeneity in their spatial distribution is expected. However, the distribution of defects in the centre of the annealed crystals is sufficiently homogeneous to meet the basic assumption made in statistical dynamical theory.

Fig. 2 shows the absolute values of the integrated reflecting power measured with 0.0392 Å γ-radiation on samples VII.1–VII.3, which had been annealed at 1023 K for 24, 72 and 216 h, respectively. Data have been taken at reflections 220 and 440 and, after rotation of the crystal disc by 90° around its cylinder axis parallel to ⟨001⟩, at reflections $2\bar{2}0$ and $4\bar{4}0$. In general, good agreement between the pairs of {220} and {440} data sets is observed, which demonstrates the expected axial symmetry in the formation of SiO_2 precipitates with respect to the growth direction. For effective thicknesses up to 4 cm, the experimental data approximate a straight line and, at $T=0$, the fitted straight lines meet the values of the integrated reflecting power calculated from dynamical theory after averaging over the *Pendellösung* oscillations. Although the defect scattering is much stronger than in the case of sample II.2, the thickness dependence of the measured integrated reflecting powers can again be described by a superposition of $R_{dyn}(A)$ and a term $R_{diff}\propto T_{eff}$. The deviation from this linear thickness dependence of the data measured at effective sample thicknesses $T_{eff}>4$ cm may be caused by the observed lower oxygen concentration away from the centre of the disc-shaped samples, which affects the SiO_2 precipitation mechanism. In the following, only data for thicknesses $T_{eff}\leq3$ cm are considered.

The experimental data shown in Figs. 1 and 2 will be analysed quantitatively by applying the results from different approaches to statistical dynamical theory. The formulae used are presented in the next section.

Results of statistical dynamical theory

As mentioned in the *Introduction*, the foundations of statistical dynamical theory are due to Kato (1980*a*, *b*). His original expressions for the integrated

reflecting power,

$$R_{\rm calc}^{\rm Kato} = R_{\rm coh} + R_{\rm mix}^{\rm Kato} + R_{\rm inc}^{\rm Kato}, \quad (7)$$

will be used after incorporating the corrections made by Guigay (1989), which do not affect the physical principles of the theory. Because the experimental data are corrected for absorption, the integrated reflecting power is given for the case of zero absorption. Later, Becker & Al Haddad (1989*b*, 1990) discussed the statistical hypotheses of the theory in detail and proposed a self-consistent formulation of the problem. To distinguish their new results from the original ones, the following notation is introduced:

$$R_{\rm calc}^{\rm B\&A} = R_{\rm coh} + R_{\rm mix}^{\rm B\&A} + R_{\rm inc}^{\rm B\&A}. \quad (8)$$

The expressions for the coherent term $R_{\rm coh}$ are identical for the two approaches.

The imperfect nature of the crystal enters the theory *via* the phase factor $\varphi(r)$ defined in (2). If the distortion field $\mathbf{u}(\mathbf{r})$ is not known, a statistical hypothesis concerning the distribution of $\mathbf{u}(\mathbf{r})$ has to be made. It can be discussed in terms of its probability distribution function $\rho(u)$ (Becker & Al Haddad, 1989*a*). Let E be the quantity

$$E = \int \exp[2\pi i \mathbf{H}\cdot\mathbf{u}(\mathbf{r})] p(\mathbf{u})\, d\mathbf{u}. \quad (9)$$

Assuming that $p(\mathbf{u})$ is an even function so that its average is zero, one can write

$$E = (1/V)\int_v \varphi(\mathbf{r})\, d\mathbf{r}, \quad (10)$$

which corresponds to Kato's definition of the long-range-order parameter. If the displacements $\mathbf{u}$ are sufficiently random, it is reasonable to assume a Gaussian distribution for $p(\mathbf{u})$. If $p(\mathbf{u})$ is isotropic, one obtains the standard expression for the static Debye–Waller factor,

$$E = \exp[-(2\pi^2/3)H^2\langle u^2\rangle], \quad (11)$$

where $\langle u^2\rangle$ is the mean square displacement over the sample. The long-range-order parameter is thus equivalent to the static Debye–Waller factor and $E = 1$ corresponds to the case of a perfect crystal. For values of $\langle u^2\rangle^{1/2}$ much larger than the lattice-plane interspacing $d = 1/H$, E becomes very small and can often be neglected. The limit $E \to 0$ corresponds to the pure 'mosaic' crystal.

Kato introduces a short-range-order parameter *via* the pair-correlation function $f(\mathbf{t})$,

$$f(\mathbf{t}) = \langle \varphi^*(\mathbf{r}+\mathbf{t})\varphi(\mathbf{r})\rangle_{\mathbf{r}}, \quad (12)$$

which is defined by the ensemble average of the product of the phase factors at two positions in the sample separated by the distance $\mathbf{t}$. It is assumed to be real and symmetric. Owing to short-range-order correlations, the parameter E should be modified by some phase fluctuations $\delta\varphi$, *i.e.*

$$\varphi = E + \delta\varphi$$

and one obtains the expression for the correlation

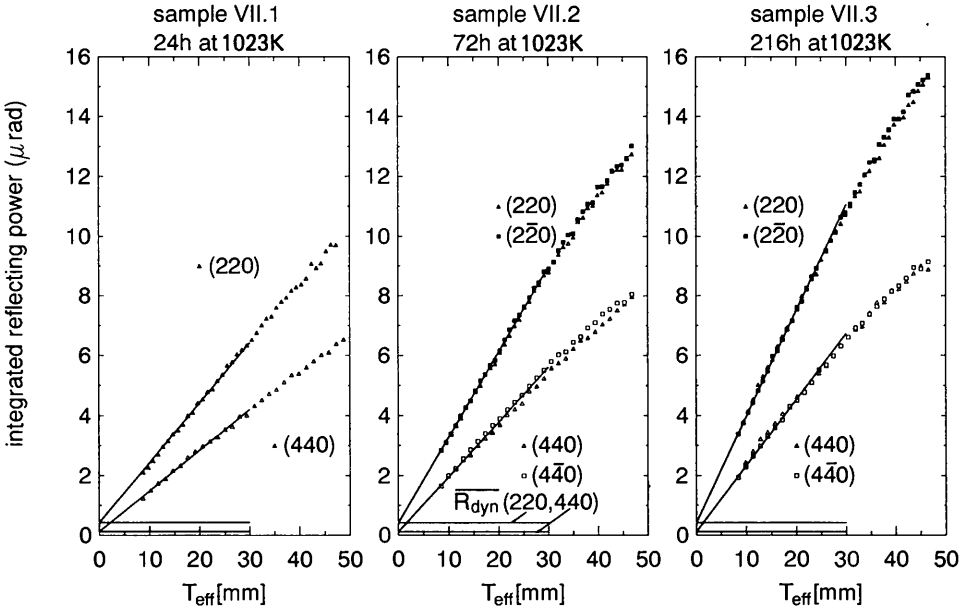

Fig. 2. Absolute values of the integrated reflecting power *versus* effective sample thickness $T_{\rm eff}$. The data are for Si samples VII.1–VII.3, which were annealed at 1023K for 24, 72 and 216 h, respectively. Data sets measured at reflections 220 and 2$\bar{2}$0 as well as at reflections 440 and 4$\bar{4}$0 are shown. The experimental data were corrected for absorption and time decay of the γ-ray source. $\overline{R_{\rm dyn}}(220)$ and $\overline{R_{\rm dyn}}(440)$ represent the values of the integrated reflecting power from dynamical theory for reflections 220 and 440 after averaging over the *Pendellösung* oscillations.

function

$$f(\mathbf{t}) = E^2 + \langle \delta\varphi^*(\mathbf{r}+\mathbf{t})\delta\varphi(\mathbf{r})\rangle_{\mathbf{r}}. \quad (13)$$

Finally, one can introduce a decreasing function $g(\mathbf{t})$ with $g(0)=1$ and $\lim_{|t|\to\infty} g(\mathbf{t}) = 0$, which describes the short-range phase correlation between two points separated by the vector $\mathbf{t}$, so that the full correlation function $f(\mathbf{t})$ is expressed as

$$f(\mathbf{t}) = E^2 + (1-E^2)g(\mathbf{t}), \quad (14)$$

with the limiting values $f(0)=1$ and $f(\infty)=E^2$. For simplicity, isotropy is assumed, so that $f(\mathbf{t})$ depends only on $|\mathbf{t}| = t$. The general shape of the correlation function $f(t)$ is shown in Fig. 3. The correlation length τ is defined in any direction as

$$\tau = \int_0^\infty g(t)\,\mathrm{d}t. \quad (15)$$

Analogously, one can define generalized correlation lengths τ_n,

$$\tau_n = \int_0^\infty g^n(t)\,\mathrm{d}t. \quad (16)$$

The formulae derived by Kato (1980*a*, *b*) for the coherent, R_{coh}, the mixed, $R_{\mathrm{mix}}^{\mathrm{Kato}}$, and the incoherent term, $R_{\mathrm{inc}}^{\mathrm{Kato}}$, of the integrated reflecting power, $R_{\mathrm{calc}}^{\mathrm{Kato}}$, given in (7) will now be presented.

The coherent term is

$$R_{\mathrm{coh}} = E^2 QTW(2EA)\exp[-2(1-E^2)\tau T/\Lambda^2], \quad (17)$$

where τ is the correlation length describing short-range order. This term will be determined together with the static Debye–Waller factor E by fitting $R_{\mathrm{calc}}^{\mathrm{Kato}}$ to the experimental data. Inspection of (17) reveals that R_{coh} is equal to R_{dyn} as given in (3) for $E=1$ and zero absorption. For $E<1$ and $\tau>0$, an exponential damping of the integrated reflecting power with sample thickness T becomes effective. Furthermore, the *Pendellösung* period and the extinction length are both increased by the factor $1/E$ owing to the modification of the upper bound of the Waller integral from $2A$ to $2AE$. In the case of negligible short-range correlation, *i.e.* $\tau \simeq 0$, the damping term disappears and only the increase of the *Pendellösung* period remains.

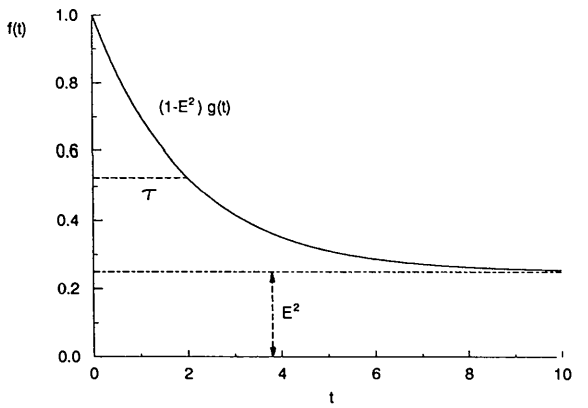

Fig. 3. Correlation function $f(t)$ for $E=0.5$ and $g(t)=\exp(-t/\tau)$ with $\tau=2$.

The incoherent term is

$$R_{\mathrm{inc}}^{\mathrm{Kato}} = [(1-E^2)/2\tau_e]Q\Lambda^2\tfrac{1}{2}[1-\exp(-4\tau_e T/\Lambda^2)] \quad \text{with } \tau_e = (1-E^2)\tau + E^2\Gamma, \quad (18)$$

where $\Gamma = \Lambda/E$ represents the correlation length for the wave-field amplitudes. It was assumed to be much larger than τ and constant within the sample. Furthermore, Kato assumed that Γ is proportional to the extinction length, *i.e.* it scales with the wavelength and the structure factor.

The mixed term is

$$R_{\mathrm{mix}}^{\mathrm{Kato}} = E(1-E^2)\tfrac{1}{2}Q\Lambda[\tfrac{1}{2}(m_1^{\mathrm{Kato}} + m_2^{\mathrm{Kato}}) - n_2^{\mathrm{Kato}}]. \quad (19)$$

The expressions for m_1^{Kato}, m_2^{Kato} and n_2^{Kato} are given in (32) and (33) of Kato (1980*b*).

Becker & Al Haddad (1989*b*, 1990) revised Kato's original approach to statistical dynamical theory and showed that the correlation length Γ for the wave-field amplitudes is variable within the sample and that it fluctuates around a value of the order of the short-range-correlation length τ, rather than being constant and of the order of the extinction length Λ, as proposed with some reservation by Kato (1980*a*). The expression $R_{\mathrm{calc}}^{\mathrm{B\&A}}$ given by Becker & Al Haddad (1989*b*, 1990) describes very well the thickness dependence of the integrated reflecting power measured with 0.0392 Å γ-radiation in Cz-grown Si crystals annealed at 1023 K and are therefore presented next.

The incoherent term is

$$R_{\mathrm{inc}}^{\mathrm{B\&A}} = (1/2\tau_2)Q\Lambda^2\tfrac{1}{2}\{1-\exp[-4(1-E^2)\tau_2 T/\Lambda^2]\} \quad \text{with } \tau_2 = \tau/2. \quad (20)$$

$g(t)$ is assumed to be of the exponential form $\exp(-t/\tau)$, with the correlation length τ, so that the correlation length of $g^2(t)$ is equal to $\tau_2 = \tau/2$.

The mixed term is

$$R_{\mathrm{mix}}^{\mathrm{B\&A}} = \tfrac{1}{2}\sigma(1-E^2)E^2Q[4\mu_2 a(Z_1 + \tfrac{1}{2}Z_2) + \gamma Z_3 - a(2\mu_2 n_1 + \alpha n_2) + (1+2\mu_2 a)n_3 + n_4], \quad (21a)$$

with

$$\sigma = 2\tau/\Lambda^2, \quad (21b)$$

$$\mu_2 = (1-E^2)2\tau_2/\Lambda^2, \quad (21c)$$

$$\alpha = 2^{1/2}E/\Lambda, \quad (21d)$$

$$a = 2\tau\alpha^2/(\alpha^2 + 4\mu_2^2), \quad (21e)$$

$$\gamma = [a(\alpha^2 - 4\mu_2^2) - 2\tau\alpha^2]/(2\mu_2) - 2, \quad (21f)$$

$$n_1=(\alpha^2+\mu_e^2)^{-2}[(\mu_e^2-\alpha^2)\cos(\alpha T)+2\mu_e\alpha\sin(\alpha T)-(\mu_e^2-\alpha^2)\exp(-\mu_e T)-\mu_e T(\mu_e^2+\alpha^2)\exp(-\mu_e T)], \quad (21g)$$

$$n_2=(\alpha^2+\mu_e^2)^{-2}[(\mu_e^2-\alpha^2)\sin(\alpha T)-2\mu_e\alpha\cos(\alpha T)+2\mu_e\alpha\exp(-\mu_e T)+\alpha T(\mu_e^2+\alpha^2)\exp(-\mu_e T)], \quad (21h)$$

$$n_3=\tfrac{1}{2}T^2\exp(-\mu_e T) \text{ provided } \tau_2=\tau/2, \quad (21i)$$

$$n_4=\mu_e^{-2}-(T/\mu_e+1/\mu_e^2)\exp(-\mu_e T), \quad (21j)$$

$$\mu_e=(1-E^2)2\tau/\Lambda^2, \quad (21k)$$

$$Z_1=(\alpha^2+\mu_e^2)^{-1}[\mu_e I_c\cos(\alpha T)+\mu_e I_s\sin(\alpha T)+\alpha I_c\sin(\alpha T)-\alpha I_s\cos(\alpha T)-\mu_e T\exp(-\mu_e T)W(2EA)], \quad (21l)$$

$$Z_2=(\alpha^2+\mu_e^2)^{-1}[\mu_e I_c\sin(\alpha T)-\mu_e I_s\cos(\alpha T)-\alpha I_c\cos(\alpha T)-\alpha I_s\sin(\alpha T)+\alpha T\exp(-\mu_e T)W(2EA)], \quad (21m)$$

$$I_c=\int_0^T \exp(-\mu_e t)J_0(2Et/\Lambda)\cos(\alpha t)\,dt, \quad (21n)$$

$$I_s=\int_0^T \exp(-\mu_e t)J_0(2Et/\Lambda)\sin(\alpha t)\,dt, \quad (21o)$$

$$Z_3=\int_0^T \exp(-\mu_e T)W(2Et/\Lambda)t\,dt. \quad (21p)$$

The calculation of the quantities Z_1, Z_2 and Z_3 involves the evaluation of integrals that are solved numerically.

In the limit of small values of the correlation length τ, *i.e.* $\tau/\Lambda\to 0$, one obtains from $R_{\rm calc}^{\rm B\&A}$ the much simpler asymptotic formula

$$R_{\rm calc}^{\rm asym}\simeq R_{\rm coh}+R_{\rm inc}^{\rm B\&A}\simeq QT[E^2W(2EA)+(1-E^2)]. \quad (22)$$

The same expression is obtained from Kato's original theory for $\tau=\Gamma=0$. In this limit the physics is evident. The value of the structure factor F_H is reduced by the static Debye-Waller factor E, which leads to an increase of the *Pendellösung* period. If the definition of $W(2EA)$ from (4) is taken into account, the average value of $R_{\rm dyn}(A)$ is proportional to F_H and is thus reduced by the factor E. As suggested in the qualitative discussion of the data presented in Fig. 1, the defect scattering is proportional to the thickness and has the weight $(1-E^2)$.

Experimental data *versus* theory

To compare the experimental data shown in Figs. 1 and 2 with theory, various theoretical expressions for the integrated reflecting power have been implemented as subroutines in the standard-fit program *MINUIT* (James & Roos, 1975). The function minimized in this program is

$$\chi^2=\sum_{i=1}^{N}[(R_{\rm obs}^i-R_{\rm calc}^i)/\sigma_{\rm obs}^i]^2. \quad (23)$$

N is the number of observations, *i.e.* the number of different tilt angles ψ_i at which the reflectivities $R_{\rm obs}^i$ were measured. $\sigma_{\rm obs}^i$ is the standard deviation of $R_{\rm obs}^i$, essentially derived from counting statistics. The quality of the fit is described as usual with the goodness of fit, GOF, and the reliability factor, R, defined as

$$\text{and} \quad \begin{aligned} &\text{GOF}=\chi^2/(N-p)\\ &R=100\left[\chi^2\Big/\sum_{i=1}^{N}(R_{\rm obs}^i/\sigma_{\rm obs}^i)^2\right]. \end{aligned} \quad (24)$$

p is the number of free parameters in the fit; GOF = 1 is expected for the best possible fit.

The performance of the various approaches to statistical dynamical theory has been studied using the data for sample II.2 and shown in Fig. 1 since in this case *Pendellösung* oscillations are clearly visible. Kato's original formula, (7), with (17)-(19), reproduces the experimental data rather well. However, a significant difference in the slopes of the lines corresponding to the experimental and theoretical data is observed. The main concern is that the slope for $R_{\rm calc}^{\rm Kato}$ is almost exclusively caused by the mixed term $R_{\rm mix}^{\rm Kato}$, whereas, from physical arguments, $R_{\rm inc}^{\rm Kato}$ should be dominant. Following the suggestion made by Takama *et al.* (1990), Kato's original formulae were changed by introducing the correlation length for the wave-field amplitudes, Γ, as an additional free fitting parameter. As shown in Fig. 4, a good fit to the experimental data was obtained. The resulting values, $\tau=1\,(1)$ μm and $\Gamma=0.5\,(8)$ μm do not differ significantly from zero. In any case, Γ is much smaller

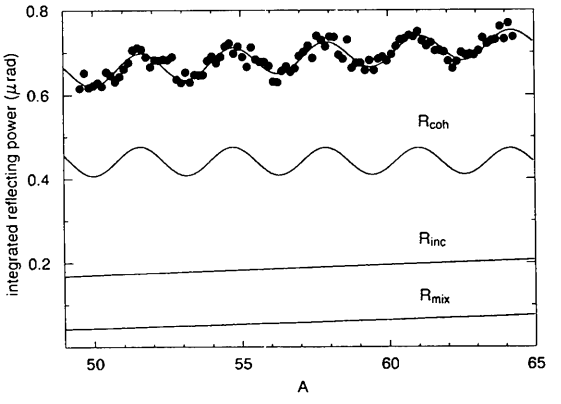

Fig. 4. A free fit of $R_{\rm calc}^{\rm Kato}$ to the experimental data for sample II.2 (the same as in Fig. 1), introducing the correlation length of the wave-field amplitudes, Γ, as an additional free parameter. The best fit was obtained for the parameter values: $E=0.99762\,(14)$, $\tau=1\,(1)$ μm and $\Gamma=0.5\,(8)$ μm. GOF = 2.1, $R=2.2\%$.

Table 2. *Results from fitting the theoretical integrated reflecting power due to Kato* (1980*a*, *b*), $R_{calc}^{Kato}(A)$ [(7) *and* (17)-(19)], *and to Becker & Al Haddad* (1989*b*), $R_{calc}^{R\&A}(A)$ [(8), (17) *and* (10)-(22)], *to the experimental data for sample* II.2

The quality of the fit is characterized by the GOF and the reliability factor R defined in (24). E is the static Debye-Waller factor, τ the correlation length describing short-range correlation [(15)], Λ is the extinction length [(5*c*)] and Γ the correlation length for the wave-field amplitudes [(18*b*)].

		Fitting parameters			Quality of fit	
	Fig.	E	τ/Λ	$\Gamma(\mu m)$	GOF	$R(\%)$
Kato (1980*a*, *b*)	4	0.9978 (4)	0.04 (1)	-	4.8	3.3
		0.99762 (14)	0.005 (6)	0.5 (8)	2.1	2.2
Becker & Al Haddad (1989*b*)	5	0.9977 (1)	0.0013 (11)	-	2.1	2.2
		0.99758 (1)	Fixed at 0.0	-	2.1	2.2

than the value of the extinction length for Si 220 and 0.0392 Å γ-radiation of $\Lambda = 216$ μm. It is also interesting to note that $R_{inc}^{Kato} > R_{mix}^{Kato}$ now, as expected from physical arguments. The results of the two fits based on Kato's approach to statistical dynamical theory are summarized in Table 2.

The same experimental data have been fitted with the theoretical expression $R_{calc}^{B\&A}$ derived by Becker & Al Haddad (1989*b*, 1990), which contains only the two parameters E and τ/Λ to be determined by the fitting procedure. As demonstrated in Fig. 5, this theory closely describes the experimental data now $R_{mix}^{B\&A} \ll R_{inc}^{B\&A}$. The numerical values obtained for the two parameters that determine the correlation function $f(t)$ are $E = 0.9977\,(1)$ and $\tau/\Lambda = 0.0013\,(11)$, *i.e.* $\tau = 0.3$ μm. The value for τ/Λ is close to zero because of the large value of the extinction length due to the short wavelength of the γ-radiation used. Therefore, it is justified to apply (22), the asymptotic form of $R_{calc}^{B\&A}$ for $\tau/\Lambda = 0$, to the present experimental data set. The quality of the fit, which contains only one free parameter, the static Debye-Waller factor E, is as good as that obtained for the full formula $R_{calc}^{B\&A}$; the numerical value of $E = 0.99758\,(1)$ agrees within statistical accuracy with that obtained from fitting the full formula.

Determination of static Debye–Waller factors

As shown in Fig. 2, the defect scattering from the Si crystals VII.1-VII.3 containing about 18×10^{17} O atoms cm^{-3} and annealed at 1023 K is much stronger than in the case of sample II.2 discussed earlier. Since the amplitude of the *Pendellösung* oscillations is of the order of 10% of the average value of the integrated reflecting power $R_{dyn}(A)$ of a perfect crystal, it is only of the order of 1% of the integrated reflecting power measured for samples VII and is thus not easily observed. Therefore, the effective crystal thickness was varied in the range from 1 to 5 cm in steps of 1 mm, instead of looking for *Pendellösung* oscillations in a rather small thickness range.

At first, the full formula for $R_{calc}^{B\&A}$ from Becker & Al Haddad (1989*b*, 1990) was fitted to the (220) data for sample VII.1. Subsequently, since the resulting value of τ/Λ was equal to zero within statistical error, the asymptotic expression R_{calc}^{asym} was fitted to these data. The result is presented in Fig. 6. The fit is

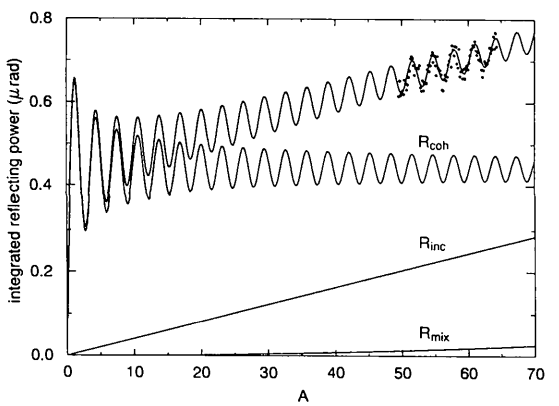

Fig. 5. A free fit of $R_{calc}^{B\&A}$ to the experimental data for sample II.2 (the same as in Fig. 1). The two parameters defining the correlation function $f(t)$ are determined to be $E = 0.9977(1)$ and $\tau/\Lambda = 0.0013\,(11)$. GOF = 2.1, $R = 2.2\%$.

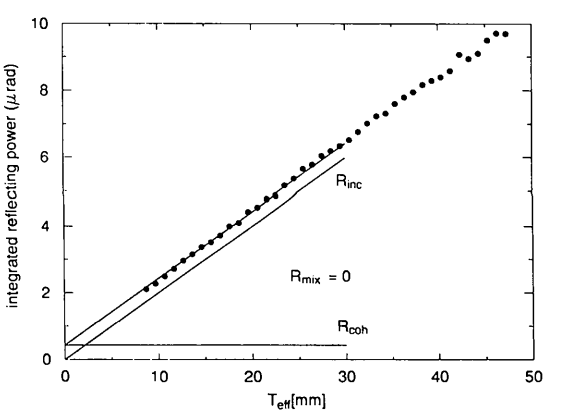

Fig. 6. A free fit of the asymptotic formula R_{calc}^{asym} of the Becker & Al Haddad theory for $\tau/\Lambda = 0$ to the experimental data for sample VII.1 (the same as in Fig. 2). The static Debye-Waller factor was determined to be $E = 0.9755\,(1)$. GOF = 0.6, $R = 1.8\%$.

Table 3. *Results from fitting the asymptotic formula of Becker & Al Haddad* (1989*b*), $R_{calc}^{asym}(A)$ [(22)], *to the thickness dependence of the integrated reflecting power measured for reflections* 220 *and* 440 *of samples* II.2 *and* VII.1-VII.3

The quality of the fit is characterized by the GOF and the reliability factor R defined in (24). E is the static Debye-Waller factor.

	220			440		
Sample	E	GOF	R(%)	E	GOF	R(%)
II.2	0.99758 (1)	2.1	2.2	-	-	-
VII.1	0.9755 (1)	0.6	1.8	0.9146 (2)	7.2	2.6
VII.2	0.9648 (1)	0.2	1.2	0.8832 (2)	7.0	2.6
VII.3	0.9559 (1)	0.4	1.5	0.8569 (3)	10.3	3.2

extremely good and, as expected, the obtained value of the static Debye-Waller factor, $E = 0.9755$ (1), is smaller than in the case of sample II.2. To determine static Debye-Waller factors for samples VII.1-VII.3, the asymptotic formula R_{calc}^{asym} of Becker & Al Haddad (1989*b*, 1990) [(22)] had therefore been fitted to all the data sets shown in Fig. 2. The results of these fits are summarized in Table 3. It is known from small-angle neutron scattering (Messoloras *et al.*, 1989) that, at 1023 K, the size of the SiO_2 precipitates increases with annealing time, which leads to an increase of the diffuse scattering and a decrease of the values of the static Debye-Waller factors E as observed in the experimental data presented in Table 3 (Bouchard, Schneider, Gupta, Messoloras, Stewart, Nagasawa & Zulehner, 1992).

In the framework of their X-ray *Pendellösung* study of structure factors of silicon, Aldred & Hart (1973) also deduce a thermal parameter $B = 0.4613$ Å^2 at 293.2 K from the thermal Debye-Waller factor

$$\text{TDWF} = \exp(-M) = \exp[-B(\sin\theta_B/\lambda)^2], \quad (25)$$

For reflections 220 and 440, one obtains TDWF = 0.9692 and 0.8824, respectively. These values are of the same order of magnitude as those for the static Debye-Waller factors E obtained *via* statistical dynamical theory for samples VII.1-VII.3, which means that the total amount of diffuse scattering should be similar in the two cases. On the other hand, it has been shown by Graf & Schneider (1986) that the contribution of thermal diffuse scattering to the integrated reflecting power is negligible in *Pendellösung* beat measurements with 316 keV γ-radiation at low-order reflections. The fact that a significant amount of diffuse scattering is found in the annealed silicon crystals must therefore be due to a localization of the diffuse scattering in **k** space around the reciprocal-lattice point, caused by the formation of defect clusters. Fig. 7 shows iso-intensity contours as measured at room temperature for sample VII.2 with a high-resolution three-crystal Laue diffractometer (Siddons, Hastings, Schneider & Berman, 1989) using synchrotron radiation with energies around 150 keV from the DORIS storage ring at HASYLAB. The diffuse scattering is concentrated within a distance of the order of 10^{-3} Å^{-1} from the 220 reciprocal-lattice point. A quantitative analysis of double-crystal γ-ray rocking curves reveals that this corresponds to a cluster radius of the order of 2000 Å (Bouchard *et al.*, 1992). The fine structure in the iso-intensity contours in the narrow stripe approximately 2×10^{-5} Å^{-1} wide and parallel to $\mathbf{H}_{220}$ is due to the resolution function of a three-crystal diffractometer in Laue geometry (Neumann, Schneider & Nagasawa, 1991).

Such large clusters cannot be brought into line with the results from small-angle neutron scattering with regard to the size of SiO_2 precipitates in silicon, which contains approximately 18×10^{17} O atoms cm^{-3} and which was subject to annealing for several hours at 1023 K. The following working hypothesis is thus adopted for the interpretation of the static Debye-Waller factors determined for samples VII.1-VII.3: firstly, compact precipitates of amorphous SiO_2 are formed and reach dimensions of the order of 40 Å; these precipitates then become grouped in loosely packed clusters with dimensions of the order of 4000 Å. This hypothesis can be verified using the theory of diffuse X-ray scattering and its application to the study of point defects and their clusters as described by Dederichs (1973).

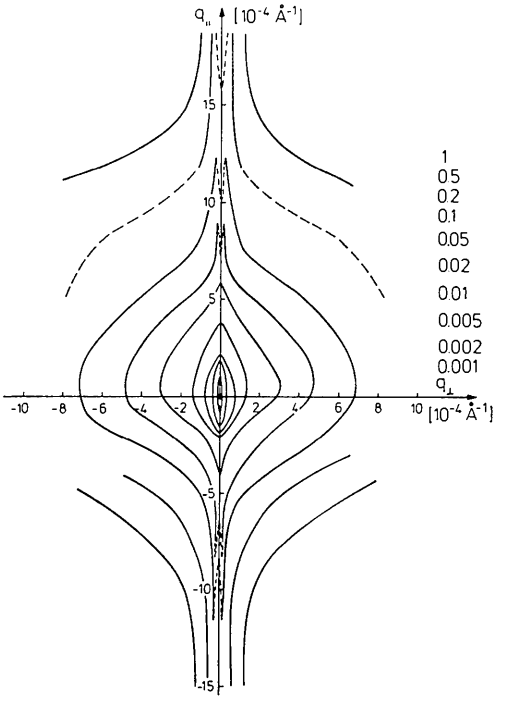

Fig. 7. Iso-intensity contour plot measured around reflection 220 for sample VII.2 using a high-resolution three-crystal diffractometer operated with 150 keV synchrotron radiation.

Table 4. *Radii, in* Å, *of spherical* SiO_2 *precipitates in samples* VII.1-VII.3, *measured directly by means of small-angle neutron scattering (SANS) and deduced from the static Debye-Waller factors listed in Table* 3

The deduced values are different depending on the assumption concerning the nature of the SiO_2 precipitate. The precipitates are significantly larger if they are assumed to be in the form of amorphous quartz instead of coesite, a high-pressure form of quartz.

		γ-ray diffractometry		
Sample	SANS	Coesite $V_{SiO_2}=34.13$ Å^3* $\varepsilon=-0.0518$	Cristobalite $V_{SiO_2}=43.48$ Å^3 $\varepsilon=0.0279$	Amorphous quartz $V_{SiO_2}=43.02$ Å^3† $\varepsilon=0.0242$
VII.1	17 (3)	9.9 (8)	18.3 (14)	21.1 (16)
VII.2	18 (3)	11.7 (9)	21.7 (17)	25.0 (19)
VII.3	22 (3)	12.3 (6)	22.9 (11)	26.3 (13)

* Liebau (1985).
† Livingston *et al.* (1984).

Determination of the size of SiO_2 clusters from static Debye–Waller factors

The number density of the SiO_2 precipitates is assumed to be sufficiently small that the single-defect approximation can be used in the calculation of the static Debye-Waller factor due to their displacement field. Since the SiO_2 molecules are densely packed inside the cluster, the static Debye-Waller factor is calculated according to (Dederichs, 1973)

$$\text{SDWF} = E = \exp(-L_H^{\text{prec}})$$

with (26)

$$L_H^{\text{prec}} = (8c_{\text{cl}}/V_{\text{cell}}) \int \{1-\cos[\mathbf{H}\cdot\mathbf{u}(\mathbf{r})]\}\,\mathrm{d}\mathbf{r}.$$

c_{cl} is the ratio of the number density of the precipitates to the number density of the matrix atoms. Owing to the large displacements $\mathbf{u}(\mathbf{r})$ in the vicinity of the clusters, the cosine cannot be expanded and, unlike the thermal Debye-Waller factor defined in (25), L_H is not proportional to $[(\sin\theta_B)/\lambda]^2$. According to Iida *et al.* (1988), the displacement field $\mathbf{u}(\mathbf{r})$ around a spherical precipitate in an isotropic medium can be written as

$$\mathbf{u}(\mathbf{r}) = \begin{cases} \varepsilon\mathbf{r} & |\mathbf{r}| \le R_0, \\ (\varepsilon R_0^3/r^3)\mathbf{r} & |\mathbf{r}| \ge R_0, \end{cases} \tag{27}$$

where R_0 is the radius of the precipitate and ε represents the strain parameter. Since one SiO_2 molecule replaces two Si atoms in the crystal, the strain parameter is fixed by the following relation between the volume of the precipitated SiO_2 molecule, V_{SiO_2}, and the volume of one Si atom, *i.e.* one eighth of the unit-cell volume, V_{cell},

$$(1+\varepsilon)^3 = 4V_{SiO_2}/V_{\text{cell}}. \tag{28}$$

V_{SiO_2} is subject to the nature of the precipitates. If SiO_2 precipitates as coesite, one obtains $V_{SiO_2}=34.13$ Å^3 (Liebau, 1985). If it precipitates in the form of amorphous quartz, one has to use $V_{SiO_2}=43.02$ Å^3 (Livingston *et al.*, 1984). After substitution of (27) into (26), Iida *et al.* (1988) obtain

$$L_H^{\text{prec}}(A) = (8c_{\text{cl}}V/V_{\text{cell}})\left[1+(3/A^3)(A\cos A-\sin A) + 3\int_1^\infty \{1-[\sin(A/x^2)]/(A/x^2)\}x^2\,\mathrm{d}x\right] \tag{29}$$

where

$$A = H\varepsilon R_0, \quad x = r/R_0, \quad V = (4\pi/3)R_0^3.$$

Equation (29) describes the situation where the SiO_2 precipitates are uniformly distributed in the crystal. If they form groups of precipitates, for example, N_{cl} precipitates are confined in a sphere of radius R_{cl}, then $L_H^{\text{prec}}(A)$ has to be multiplied by a large factor, which is independent of the scattering vector $\mathbf{H}$. As will be shown in a future paper (Bouchard *et al.*, 1992) the clusters of radius R_{cl} are formed by weakly interacting precipitates of radius $R_0 \ll R_{\text{cl}}$ so that their displacement fields superimpose. In this case, one obtains for the static Debye-Waller factor of the clustered system (Dederichs, 1973)

$$L_H^{\text{cl}} = [1+N_{\text{cl}}(R_0/R_{\text{cl}})]L_H^{\text{prec}}. \tag{30}$$

Since the static Debye-Waller factor has been determined for samples VII.1-VII.3 and reflections 220 and 440, the radius of the SiO_2 precipitates can be determined from the ratio $L^{\text{cl}}_{\{440\}}(\varepsilon R_0)/L^{\text{cl}}_{\{220\}}(\varepsilon R_0)$ for a given value of ε, as all factors involving number densities or the radius of the large clusters cancel. The results presented in Table 4 show very good agreement between the radii of the SiO_2 precipitates deduced from the static Debye-Waller factors and the values directly determined by means of small-angle neutron scattering. As expected, the agreement

is better for the assumption that SiO_2 precipitates in the form of amorphous quartz or cristobalite.

Concluding remarks

The contribution of thermal diffuse scattering to the integrated reflecting power is very weak in measurements using 316 keV γ-radiation. This is due to the wide distribution of TDS in **k** space and the special resolution element in γ-ray diffractometry, which is a thin disc of large dimensions perpendicular to the scan direction. The values of the thermal and static Debye–Waller factors are of the same order and therefore the total amount of defect scattering should be comparable to that of TDS. The fact that the defect scattering could be measured by means of γ-ray diffractometry suggests that the defect scattering is rather localized in **k** space around the reciprocal-lattice vector, which has been verified by mapping the diffuse scattering around $\mathbf{H}_{220}$ with a high-resolution Laue-case three-crystal diffractometer operated with 150 keV synchrotron radiation. As a consequence, only clustered SiO_2 molecules contribute to the integrated reflecting power measured by means of γ-ray diffractometry and the contribution of small defects will most probably disappear in the background of the measured rocking curve. In the present case, two models of clustering must be considered. In sample II.2 the SiO_2 molecules form large plate-shaped precipitates of approximate dimensions $280\times 280\times 22$ Å. These precipitates cause a sufficiently strong long-range strain field to concentrate a substantial amount of diffuse scattering around the reciprocal-lattice point. In samples VII.1–VII.3 the SiO_2 precipitates are spherical with diameters of the order of only 40 Å. This should result in a weaker diffuse scattering extended further into **k** space, the contribution of diffuse scattering to the measured integrated reflecting power should be smaller than in the case of sample II.2. However, the contrary was observed. The defect scattering from samples VII.1–VII.3 is much stronger and this cannot be explained by their higher oxygen content. Instead, it is suggested that the relatively small SiO_2 precipitates are loosely bound within large clusters that create the long-range strain field leading to the contraction of a large amount of defect scattering around the reciprocal-lattice points.

A comparison of the results of statistical dynamical theory with the experimental data for sample II.2 reveals that the incoherent term of the integrated reflecting power, calculated from Kato's original formula, is very small. The defect scattering is almost completely described *via* the mixed term, which is against physical intuition. If the correlation length Γ for the wave-field amplitudes is introduced as an additional free fitting parameter, the experimental data are well described. The incoherent term of the calculated integrated reflecting power is now larger than the mixed term. In contrast to Kato's original suggestion, Γ is of the same order of magnitude as τ and much smaller than the extinction length Λ for Si 220 and 316 keV γ-radiation.

The expression for the integrated reflecting power calculated by Becker & Al Haddad (1989*b*, 1990) within the framework of statistical dynamical theory describes the experimental data very well. A fit of the full formula shows that the parameter τ is very small. This means that the crystal imperfections can be characterized by the static Debye–Waller factor alone. The asymptotic formula for $\tau = 0$ describes the experimental data equally well and allows a unique determination of the static DWF from the measured thickness dependence of the integrated reflecting power. The ratio of the static DWF determined for reflections 220 and 440 provides the product of the strain parameter ε and the radius R_0 of the SiO_2 precipitates, which have been assumed to be spherical in agreement with the results from small-angle neutron scattering. The R_0 values determined directly from SANS are within the limits of the precipitate size deduced from the static DWF using the values of the strain parameter for the possibilities that SiO_2 precipitates in the form of cristobalite or amorphous quartz. This excellent agreement is in strong support of statistical dynamical theory in the version presented by Becker & Al Haddad (1989*b*, 1990).

In spite of the success of the statistical dynamical theory in the interpretation of the integrated reflecting power measured with 316 keV γ-radiation for Czochralski-grown dislocation-free silicon crystals, one has to admit that the distribution of precipitates in an otherwise perfect crystal is probably the simplest type of defect structure to be treated by such a theory. At these short wavelengths, the Bragg peak related to the coherent scattering is highly localized in **k** space and the overlap with the diffuse scattering, despite it being confined to an area of dimensions only 10^{-3} Å^{-1}, is very small. This is also the probable cause of the mixed term of the calculated integrated reflecting power being negligible compared with the coherent and incoherent terms. Investigations of silicon crystals annealed at temperatures above 1173 K represent a more stringent test of statistical dynamical theory, because here dislocations and/or stacking faults are formed in addition to large SiO_2 precipitates and short-range correlation should be crucial for the diffraction process. A recent paper by Kato (1991) gives a more rigorous foundation of the statistical dynamical theory of diffraction, which is intended to provide the basis of more practical theories for specific topics to be developed in the future.

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Reformulation of the Dynamical Theory of Coherent Wave Propagation by Randomly Distorted Crystals

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Abstract

The coherent terms in the spherical-wave approach of the statistical dynamical theory are reformulated using rigorous boundary conditions in contrast to the intuitive boundary conditions of the original formulation by Kato [*Acta Cryst.* (1980), A**36**, 763-769, 770-778]. These boundary conditions are explained physically by a general interference effect between the forward-diffracted wave and the incident undiffracted wave (using the optical theorem) and their consequences on the total (Bragg and forward-diffracted) incoherent intensity are also discussed.

1. Introduction

The statistical dynamical theory (SDT) of Bragg diffraction by randomly distorted crystals, first formulated by Kato (1980*a*, *b*), can be divided into two parts. Only the first part, consisting in the calculation of the so-called coherent waves diffracted into the Bragg and forward directions, will be considered explicitly in the present paper. Even in the case of a nonabsorbing crystal, these coherent waves lose intensity, which is transferred to the incoherent beams calculated in the second part of the theory. We shall consider the case of a nonabsorbing crystal with a centrosymmetrical structure, in Laue symmetrical conditions. Our purpose is to reveal the differences between our approach and the previous one by Kato (1980*a*, *b*) and by Al Haddad & Becker (1988), Becker & Al-Haddad (1989, 1990, 1992). This is particularly clear when the static Debye-Waller factor is equal to zero; in the previous approach there are then no coherent diffracted intensities at all but in our approach there is indeed some coherent intensity in the forward-diffracted beam.

The present paper deals with the point-source functions (PSF) that represent the coherent waves generated by a point source on the entrance surface of the crystal (this is known as a 'spherical wave'). A slightly different form of the differential equations satisfied by the coherent PSF is proposed and special attention is paid to the boundary conditions which must be in agreement with the usual dynamical theory of diffraction by perfect and by nonrandomly deformed crystals. We show that the transmitted wave is not affected by the random distortion of the crystal in a narrow region close to the direction of the undiffracted wave. This has a physically meaningful consequence: the transmitted intensity is reduced by interference between the undiffracted wave and the forward-diffracted wave, this reduction being compensated for in the total diffracted intensity (this is simply a statement of the 'optical theorem' described in most textbooks on quantum mechanics).

Our results for the coherent PSF are in agreement with the paper by Polyakov, Chukhovskii & Piskunov (1991) and it is proposed to use a similar approach for the calculation of incoherent beams. This is discussed here and will be the topic of forthcoming papers.

2. The PSF in diffraction by a distorted crystal

Let O be a point source on the entrance surface of the crystal; we shall use the nonorthogonal coordinate system (OS_o, OS_h) shown in Fig. 1. Let $G_h(s_o, s_h)$ and $G_{od}(s_o, s_h)$ be the amplitudes of the Bragg-diffracted and forward-diffracted waves respectively.

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