should be pointed out. First, a broad second-neighbor peak is clearly resolved in the DDF's and higher broad coordination shells are quite evident. Information on such broad shells cannot be obtained from EXAFS analysis because of the lack of low-k information. Another advantage of DAS is that electron phase shifts and mean-free-path terms are not required. On the other hand, DAS is not as sensitive to the coordination of dilute species since it is obtained by looking at the difference between two large signals. Also, the DAS cannot be Fourier transformed to yield DDF's unless a fairly high energy-absorption edge is used (due to the limited k range available).

The current accuracy of DAS is mainly limited by inadequate knowledge of f'. The composition range is limited to, perhaps, $>5\%$, except for heavy elements in a light matrix, and the elemental range to above Cr if k information beyond 6 Å^{-1} is desired. This elemental limit applies mainly to Fourier transformation. Small- and low-angle scattering information useful for model building can be obtained on much lower-Z materials. Thus we are confident that DAS can be successfully applied to a wide range of problems in structural analysis of amorphous materials, poorly crystallized materials, and materials on low-Z substrates (e.g., catalysts).

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Absolute Measurement of the (220) Lattice Plane Spacing in a Silicon Crystal

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The (220) lattice plane spacing of an almost perfect crystal of silicon was measured by means of a combined scanning (*LLL*) x-ray interferometer and a two-beam optical interferometer. From 170 measurements, a value $d_{220}=(192\,015.560\pm0.012)$ fm results in vacuum at 22.50 °C. This value is smaller by $1.8\times10^{-6}d_{220}$ than that reported by Deslattes *et al.* for another crystal. Generic variabilities of the two crystals account only for a part of this difference.

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Bonse and Hart[1] proposed to measure the lattice plane spacing of crystals by means of a scanning x-ray interferometer and Bonse and te Kaat[2] observed x-ray fringes in a two-crystal (*LLL*) interferometer. Deslattes and Henins,[3] using this method, succeeded first in measuring the (220) lattice plane spacing of silicon with a stated uncertainty of 3×10^{-7}. The reported value has become the basis of the latest evaluation of characteristic x-ray wavelengths (Cu $K\alpha_1$ and Mo $K\alpha_1$). An extended series of measurements with a reduced uncertainty led to a new value for the Avogadro constant.[4] The d_{220} value was corrected for systematic errors,[5] and its uncertainty finally reduced to 1×10^{-7}.[6-8]

In this paper, a new and significantly different value for d_{220} of silicon is reported. It was measured by the same method as before, but with a

measuring device rather different from that employed by Deslattes. The silicon material investigated was grown and purified by multiple-float-zone melting by Wacker-Chemitronic.[9] Its impurity concentrations were found to be less than 1×10^{13} cm^{-3} for boron atoms and less than 1×10^{16} cm^{-3} for carbon and oxygen atoms.[10]

From this material, the two crystals of the (*LLL*) x-ray interferometer were cut. The analyzer was mounted on a translation stage, similar to that described earlier.[11] The translation was measured by two-beam interferometry with use of an optical polarization interferometer according to Curtis *et al.*[12] The principle of the arrangement is shown in Fig. 1.

During the displacement of the analyzer crystal, in the outgoing x-ray and laser beams intensity variations are simultaneously observed, which are periodic with the lattice plane spacing d of the crystal and half the wavelength λ of the laser radiation, respectively.

The displacement of an integer number m of $\lambda/2$ corresponds to a number n of x-ray fringes. Thus the lattice plane spacing results from

$$d=(n/m)^{-1}\lambda/2, \tag{1}$$

with $n=N(m)+f(m)$. Here $N(m)$ is the integer part of n, while $f(m)$ is the residual fraction. In a series of measurements n/m was determined over pathlengths $m\lambda/2$, starting with $1\lambda/2$ and proceeding over $2\lambda/2$, $4\lambda/2$, and so on successively until $N(m)$ was ascertained for $m=120$. Then $f(m)$ alone was measured as a function of the displacement $m\lambda/2$. For each of the 170 measurements of d mentioned below typical values of m were 0, 30, 60, 90, 120, 90, 60, 30, and 0. Details with regard to the determination of the relative phase of optical and x-ray signal have been reported previously.[13]

The measuring device possesses the following special features[13,14]: Two entirely separated interferometer crystals, each with three steel balls attached to its base, rest on supports S_a and S_b, as shown in Fig. 2, without being fixed to them. By this provision the crystals may be exchanged easily. The direct comparison of crystal material from different sources will be facilitated. For the optical measurement of the displacement, the front faces of the crystals are polished in order to form optical mirrors which are a part of the lattice itself. Stability problems which may arise from having glue or resin somewhere in the link between the optical mirrors and the interferometer crystals are thus avoided. The point of impact of the laser beam is optimized in order to get a negligible Abbe correction. By making the waist of the laser beam not too narrow only a

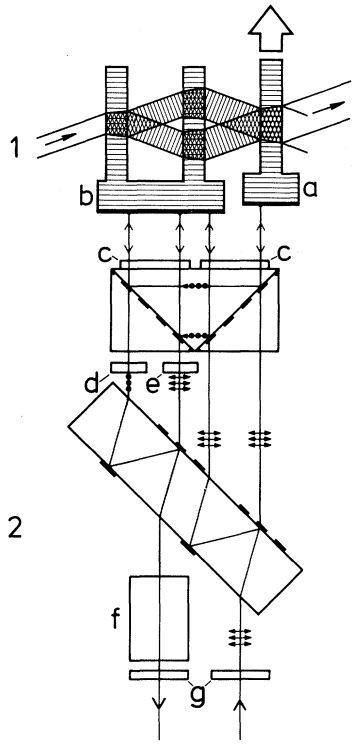

FIG. 1. Principle of the measuring device x-ray interferometer 1 with the movable analyzer crystal a and fixed crystal b. Optical polarization interferometer 2 with $\lambda/4$ plates, c; $\lambda/2$ plate, d; compensation plate, e; phase modulator (Pockels cell), f; and polarizers, g.

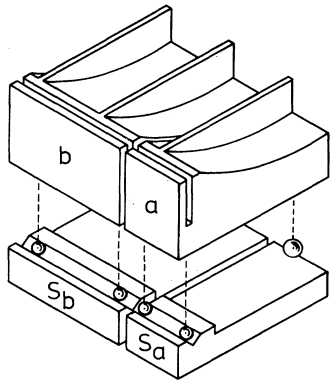

FIG. 2. View of the x-ray interferometer with optical mirrors a and b. Each crystal stands on three steel balls, two in a groove, avoiding undesired rotations and translations.

very small correction due to the nonplanarity of the wave fronts[15] (Fresnel phase shift) is necessary (see correction k_1 below). We used a Lamb-dip stabilized He-Ne laser (Spectra Physics model 119), the frequency of which was controlled repeatedly during measurements by a $^{127}I_2$ stabilized He-Ne laser. Optical feedback was avoided by a light trap (> 30 dB attenuation[14]). The double parallel spring used as a translation stage has very small guiding errors, so that the visibility of the x-ray fringes did not noticeably decrease for a translation of 40 μm, achieved with a piezoelectric drive. With an improved drive the translation stage is expected to be capable of translations up to 500 μm. With computer-aided on-line evaluation, fractions $f(m)$ of one thousandth can be detected. Detailed off-line evaluations with the fitting of a sine function to the x-ray signal with use of the least-squares methods yielded identical results.

With this apparatus 170 measurements of the ratio n/m were carried out in vacuum at temperatures between 22.42 and 22.50 °C. Each value was reduced to 22.50 °C with use of the thermal expansion coefficient[16] $\alpha = (2.56 \pm 0.03) \times 10^{-6}$ K^{-1}. From these reduced values, a histogram of which is displayed in Fig. 3, a mean value $\langle n/m \rangle = 1648.281\,626$ with a standard deviation of the single measurement, $\sigma = 0.000\,095$, and a standard deviation of the mean, $\sigma_m = 0.000\,007$, were calculated.

The sequence of mean values each calculated from ten successive measured values—shown in Fig. 4 together with the related σ_i values—does not exhibit any significant change of the measured value with time. Fourteen of the seventeen values (more than 67%) fall into the interval $\langle n/m \rangle \pm \sigma/\sqrt{10}$. Large values of σ_i indicate ground vibrations which could be correlated to stormy weather.

Before d was calculated according to Eq. (1), the value $\langle n/m \rangle$ had to be corrected for several effects:

(a) The laser beam has a vacuum wavelength $\lambda_0 = 632\,991\,415 \pm 5$ fm and a Gaussian beam profile. The beam radius in the waist within the interferometer is $w_0 = 0.52 \pm 0.02$ mm. To take into account the Fresnel phase shift in the laser beam, a correction $k_1 = (-3.8 \pm 0.2) \times 10^{-8} \langle n/m \rangle$ had to be made.[17]

(b) The translation of the analyzer crystal is correctly measured only if the normal of the mirror, the direction of the laser beam, and the direction of movement are parallel to the normal of the lattice planes. In this experiment, however, residual misalignments are present, leading to a cosine correction $k_2 = (-0.1 \pm 0.2) \times 10^{-8} \langle n/m \rangle$.

(c) The laser beam, impinging on the mirror of the movable crystal, does not point exactly to the center of the x-ray beam at the entrance surface of the analyzer crystal. The lateral offset, together with the tilting of the crystal during movement, produces the well-known Abbe error, given by the product of offset and tilting angle. Since the tilting angle was determined to be zero, no Abbe correction was necessary. From the uncertainty in determining this angle of $\pm 5 \times 10^{-9}$ rad combined with a horizontal offset of 40 μm and a vertical offset of 240 μm an uncertainty of $\pm 3.0 \times 10^{-8} \langle n/m \rangle$ follows.

(d) An uncertainty of $\pm 5.1 \times 10^{-8} \langle n/m \rangle$ results from the uncertainties of the crystal temperature (±0.02 K) and of the thermal expansion coefficient ($\pm 3 \times 10^{-8}$ K^{-1}).

Taking into account the corrections (a) to (d) and summing up all uncertainties by quadrature, the following value for the lattice plane spacing d_{220} of our silicon crystal at 22.50 °C in vacuum was

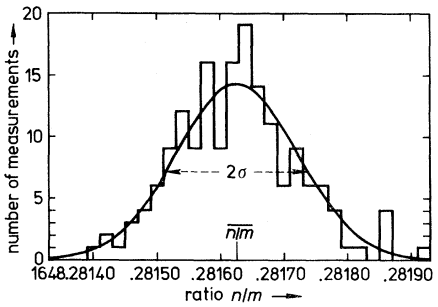

FIG. 3. Histogram of 170 measurements of the ratio n/m; the drawn line represents a least-squares fit of a Gaussian curve. All values reduced to 22.5 °C and vacuum; no systematic corrections were performed.

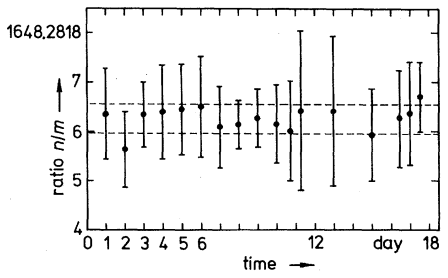

FIG. 4. Temporal fluctuations of the mean of ten successive measurements.

determined:

$$d_{220} = 192\,015.560 \pm 0.012 \text{ fm}.$$

This value (relative uncertainty 6×10^{-8}) is by $1.8\times10^{-6}d_{220}$ smaller than that reported by Deslattes for his PERFX crystal under the same conditions. According to Ando, Bailey, and Hart,[18] the lattice spacings of dislocation-free float zone crystals of Wacker and crystals of comparable generic characteristics differ from carbon- and oxygen-free PERFX by $(-0.6\pm0.2)\times10^{-6}d_{220}$. The difference can well be assumed to be due to carbon (dominant) and oxygen impurities ($\Delta d/d = -6.5\times10^{-24}n_C$ and $\Delta d/d = +3.8\times10^{-24}n_O$ with n_C, n_O number of C and O atoms per cm^3). With use of the above-stated impurity concentrations for our sample crystal, a maximum impurity-induced lattice-spacing deviation of $-6.5\times10^{-8}d_{220}$ to $+3.8\times10^{-8}d_{220}$ is possible. From this we argue that the material we used is closer to PERFX than to any other float zone material investigated by Ando, Bailey, and Hart and that the difference in d_{220} cannot be explained by impurities of our crystal material.

The unit cell volume of a PERFX crystal was used in the latest determination of the Avogradro constant.[4,19] If crystal to crystal comparison should show that the lattice parameters of that PERFX crystal and our crystal are very close to each other, as we assume, our result would imply an increase in the Avogadro number by $5.4\times10^{-6}N_A$. Should such comparison show a considerable difference in lattice parameters, the reasons for that difference (and its influence on the Avogadro number) must be explored in detail.

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