X-ray refractive-index measurement in silicon and lithium fluoride

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The refractive indices n of silicon and lithium fluoride were measured noninterferometrically with Mo $K\bar{\alpha}$ and Ag $K\bar{\alpha}$ x rays to a sub-part-per-billion accuracy. This high accuracy allows experimental determination of the real dispersion correction f' to ± 2 millielectron accuracy. The f' values obtained are in excellent agreement with the best interferometric measurements, part of which are less accurate than the present results. The predictions of both the Cromer-Liberman and the modified Hönl theories are found to deviate significantly from the measured f' values, thus indicating the need for modification of the wave functions or, more likely, the exchange potential used.

I. INTRODUCTION

The refractive index n of matter for x rays is given by[1]

$$n=1-\delta-i\beta$$
$$=1-(1/2\pi)r_e\lambda^2\sum_j N_j(Z_j+f_j'+if_j'') , \quad (1)$$

where r_e is the classical electron radius and λ is the wavelength of the radiation. The sum j is carried over all atomic constituents of the sample and N_j, Z_j, and f_j', and f_j'' are the number of atoms per unit volume, the atomic number, and the real and imaginary dispersion terms, respectively, of atom j. δ is of order 10^{-6} while f' is only a few percent of Z at most outside the immediate vicinity of an absorption edge. Thus a measurement of f' to a few percent implies a measurement of n to sub-part-per-billion accuracy, a level achievable, until recently, only by x-ray interferometric[2-6] methods.

Recently, we have reported[7] noninterferometric measurements of n for Be at several wavelengths with a level of accuracy equal to that of the best interferometric[3,4] methods. The method used in that, as well as the present study, is that of measuring the angular deviation of x rays passing through a sample wedge by a monolithic thin wafer Laue-Laue diffractometer of a novel design.[7,8]

We report here measurements of the real decrement δ of LiF and Si for Mo $K\bar{\alpha}$ and Ag $K\bar{\alpha}$ energies. From the measured values of δ the dispersion correction f' was calculated to an accuracy of ~ 2 millielectrons. The results obtained are in excellent agreement with previous interferometric[4,9,10] and, in part,[9,10] less accurate measurements, but significantly deviate from the best theoretical estimates.[11-15]

II. THE METHOD

The experimental arrangement is depicted in Fig. 1. The Laue-Laue diffractometer is a monolith whose "crystals" are two thin wafers cut out of a single block of perfect silicon crystal oriented with its (hhh) Bragg planes perpendicular to the wafers' faces. A rotation of one wafer relative to the other is done by bending a leaf spring cut into the monolith in between the wafers, using a linear force generator which consists of a small electromagnet and a small permanent magnet attached to the silicon blocks carrying the wafers. In the absence of a sample the maximum intensity is transmitted for an angle of $\Delta=0$ between the wafers. A sample wedge positioned between the wafers will, however, cause the beam impinging on the second wafer to deviate by an angle Δ proportional to n, from its original direction so that the second wafer will have to be rotated by Δ to obtain maximum transmission. Thus a measurement of the required rotation yields the refractive index n of the sample. Measurements of two deviations Δ and Δ' for two different Bragg planes are required to obtain n, which is the solution of the equation[7]

$$A-B+(\tan\gamma)(1+BA)=0 , \quad (2)$$

where

$$A=-a+(a^2-1+2a/\tan\alpha)^{1/2} ,$$
$$B=-b+(b^2-1+2b/\tan\alpha)^{1/2} ,$$

and

$$a=\Delta/[2(1/R-1)] ,$$
$$b=\Delta'/[2(1/R-1)] .$$

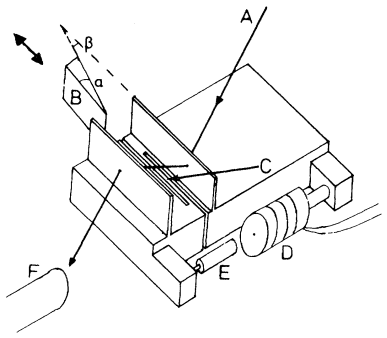

FIG. 1. Experimental arrangement. The entire diffractometer is a monolithic perfect silicon crystal. *A*, x-ray beam; *B*, sample wedge; *C*, leaf spring; *D*, electromagnet; *E*, permanent magnet; *F*, detector.

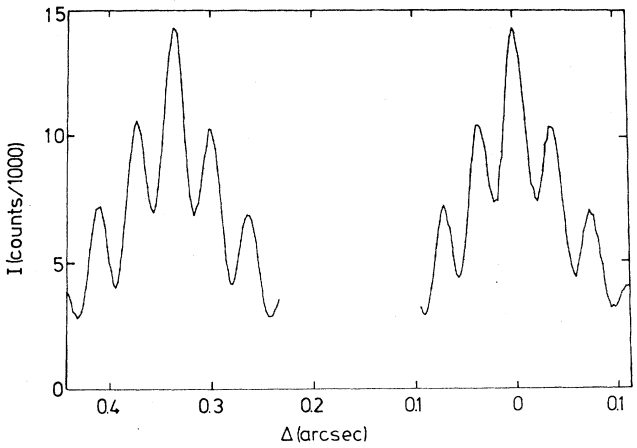

FIG. 2. 777 rocking curves measured with Ag $K\bar{\alpha}$ radiation without (right) and with (left) a Si sample wedge in the beam path. Note the large separation and narrow central peak, which allow for the high accuracy in determining the angular deviation in the wedge.

α and γ are the known wedge angle and the difference between the Bragg angles of the two planes, respectively. $R=n/n_a$ where

$$n_a=1-1.59\times10^{7}\lambda^{2}$$

(λ is in cm) is the refractive index of air.[4]

The monolith used in this experiment is the one used in the previous study[7] with both wafers nominally 275 μm thick. Collimation to 10 min of arc in the diffraction plane and 2° perpendicular to it were used on the primary beam side and a collimator made to two steel blocks was employed in front of the detector to select the R beam of the second Laue reflection. A solid-state Ge detector followed by a single-channel pulse height analyzer was used to obtain very high signal-to-noise ratios and the experiment was carried out under microcomputer control. The angular deviation Δ was determined by fitting Gaussians to the central peaks of the rocking curves measured with and without a wedge sample in the beam path. Further details of the method, experimental technique, and data analysis are given in Ref. 7.

The wedges were cut, lapped, and polished from single crystals of LiF and Si, the impurity contents of which were negligible as far as possible influence on the results presented here is concerned. The wedge angles were

$$\alpha_{\mathrm{LiF}}=19.9681°\pm0.0026°,$$

$$\alpha_{\mathrm{Si}}=30.0500°\pm0.0020°.$$

A typical set of rocking curves obtained with and without a sample wedge is given in Fig. 2.

III. DISCUSSION OF RESULTS

The Bragg planes and wavelengths employed and the deviation Δ measured for each sample are listed in Table I. The table also lists the real decrement δ of the refractive index calculated from these deviations using Eq. (2), and the f' values computed from them using Eq. (1). Several theoretical[11–15] as well as experimental f' values measured interferometrically[4,9,10] are also listed.

The agreement between the different measured f' is very good. Although for Si our values seem to be slightly lower at both wavelengths than those of Cusatis and Hart[4] the difference is well within the combined experimental error. Good agreement is achieved with the differential measurements of Takeda and Kato[16] where $f'_{\mathrm{Mo}}-f'_{\mathrm{Ag}}=0.0248\pm0.0024$. Our LiF results agree as well with the less accurate interferometric measurements of Creagh,[10] and Creagh and Hart.[9] The agreement between theory and experiment is, however, much less satisfactory.

Two theoretical approaches which achieved a measure of success in interpreting anomalous dispersion data will be discussed here. The first of these, the original Hönl[11,17,18] approach, uses hydrogenic wave functions and takes into account the contribution of K electrons only. Consequently it is expected to be a good approximation for light atoms and energies far above the K absorption edge only, as is the case here. Unfortunately, the excellent numerical agreement of the measured values and f'_B does not reflect a real success of this theory, since inclusion of

TABLE I. Anomalous dispersion correction f' in Si and LiF. Δ is the deviation in the peak position caused by the wedge, and δ is the real decrement of the refractive index. f'_{expt} is the dispersion correction measured in this experiment. The other values of f' are marked by the initials of the authors: f'_{CH}, Cusatis and Hart (Ref. 4) for silicon only; f'_C, Creagh (Ref. 10) for lithium fluoride only; f'_B, Barnea (Ref. 11); f'_W, Wagenfeld and co-workers (Refs. 12 and 13); f'_{CL}, Cromer and Liberman (Refs. 14 and 15).

Sample	λ (Å)	Bragg planes	Δ (msec)	δ_{expt} ($\times10^{6}$)	f'_{expt} (electrons)	f'_{CH}, f'_C	f'_B	f'_W	f'_{CL}
Si	0.5608	(555)	210.986±0.032	0.989 95±0.000 18	0.0537±0.0025	0.0568±0.0026	0.06	0.0706	0.042
		(777)	372.358±0.030						
	0.7107	(444)	353.674±0.040	1.593 40±0.000 21	0.0847±0.0018	0.0863±0.0018	0.09	0.101	0.071
		(555)	492.698±0.032						
LiF	0.5608	(777)	187.729±0.030	1.037 62±0.000 19	0.0133±0.0022	0.014±0.006	0.014	0.015	0.006
		(888)	267.164±0.025						
	0.7107	(555)	272.881±0.032	1.667 62±0.000 21	0.0217±0.0015	0.020±0.005	0.021	0.020	0.014
		(777)	808.785±0.030						

contributions from higher shells as calculated by Wagenfeld and co-workers[12,13] and denoted by f'_W in Table I, destroys rather than improves the agreement between theory and experiment. For a somewhat lower energy, e.g., Cu $K\bar{\alpha}$, the difference between this theory and experiment is quite large, even for the relatively light LiF: $f'_{\text{expt}}=0.099$ (Ref. 9) vs $f'_B=0.071$.[11]

The second and more recent approach, that of Cromer and Liberman,[14,15] employs the relativistic Dirac-Slater wave functions introduced by Brysk and Zerby[19] to calculate the required photoelectric absorption cross sections. In addition, measured[20] energy levels rather than computed eigenvalues are used in the calculation. This approach is generally considered[10,13] superior both conceptually and quantitatively to the previous one and has, therefore, been repeatedly[2,4–6,9,10,13] compared with experimental f' values. Nevertheless the agreement between f'_{CL} and the experimental results in Table I is not good both for LiF and Si. While the possibility of a computational error in f'_{CL}, as suggested by Wagenfeld[13] and Creagh,[10] cannot be ruled out completely, it is highly improbable as two independently written computer codes[14,21] gave the same values for f'_{CL} for a number of elements and wavelengths. It seems, therefore, that the flaw in the Cromer and Liberman theory is of a more fundamental nature. The wave functions or, more likely, the Kohn-Sham exchange potential[22] employed in the calculations, will have to be modified to achieve better agreement between theory and experiment. A much larger body of high precision $f'(\lambda,Z)$ data than that available at present would, however, be required to gain a detailed understanding of the dispersion of x rays in matter.

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