Data Processing

A suite of programs for calculating x-ray absorption, reflection, and diffraction performance for a variety of materials at arbitrary wavelengths

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One of the most useful characteristics of synchrotron radiation is the wide spectral distribution of the source. For applications involving tuned monochromatic beams it is often helpful to predict the x-ray optical characteristics of a sample or the beam line optics at a particular wavelength. In contrast to this desire stands the fact that tabulated values for the optical parameters of interest are generally available only at wavelengths corresponding to typical x-ray tube sources. We have developed a suite of FORTRAN programs which calculate photoabsorption cross sections and atomic scattering factors for materials of arbitrary, uniform composition or for arbitrary layered materials. Further, the suite includes programs for calculation of x-ray diffraction or reflection from such materials. These programs are of use for experimental planning, data analysis, and predictions of performance of beam line optical elements.

I. INTRODUCTION

One of the advantages of doing experiments at a synchrotron radiation facility is that one can choose the x-ray wavelength for an experiment and the choice in many cases is critical to the success of the experiment. Much of the vast prior work on x-ray optics calculating such things as reflectivity and total integrated power are based on using characteristic lines from conventional radiation sources. In considering how to monochromatize the incident beam with the best combination of flux and resolution for an arbitrary wavelength the ability to calculate crystal performance at that wavelength is crucial. As a result we have combined a variety of existing data bases, a suite of subroutines for extracting parameters from them, and a set of programs for calculating such things as the crystal structure factors for an arbitrary crystal at an arbitrary wavelength. In this paper we describe the data bases, the subroutine calls which access these data, and a set of higher level routines and programs which we have developed.

The atomic scattering factor is wavelength and scattering angle dependent. For x rays scattered in the forward direction the corrections to the form factor are $f = f_0 + f' + if''$, where f' is the real and f'' is the imaginary term. These terms are known as the anomalous dispersion corrections or sometimes the Hönl terms. For calculating f' and f'' we use the data of Cromer and Liberman[1] which follows upon earlier work by the same authors.[2,3]

The imaginary anomalous dispersion term f'' is related to the total photoelectric absorption cross section via the optical theorem. To find the total attenuation cross section one must sum the photoelectric, Compton, and Rayleigh cross sections. The energy dependence of the latter two have been parametrized by McMasters et al.[4]

We have created three data bases for the VMS operating system. One data file stores both the Cromer and Liberman f' and f'' values, as well as the absorption edge energies and atomic weights and densities. The second contains the parametrized Compton and Rayleigh cross sections. The third data base is the parametrization of the atomic scattering factor vs scattering angle and wavelength. The data bases are accessible through a set of subroutine calls which follow the VMS calling standard. These calls have been packaged in a shareable image library. The advantage of a shareable image library is that one need not explicitly link to routines in such a library, just as one does not explicitly link to the FORTRAN intrinsic routine library. Table I shows the subroutines available. The atomic scat-

TABLE I. Basic subroutine calls for absorption package.

Name	Input parameters	Output parameters
ABS$CROMER	Z, Energy	f, f'' (in electron units)
ABS$ATOMIC__DATA	Z	Atomic weight, density absorption edge energies
ABS$RAYCOMP	Z, Energy	Rayleigh, Compton cross section (in electron units)
ABS$ELEMENT	Element symbol	Z
ABS$MASS__ABS	Z, Energy	MU/RHO, density
ABS$SFCOEF	Z, k [$4\pi \sin(\theta)/\lambda$]	$f(k)$, compton (k)

TABLE II. Programs based directly on absorption package.

Name	Description
TREF	Calculate Reflectivity, Penetration Depth, Field**2 as f (incidence angle) for arbitrary material.
EREF	Calculate reflectivity vs energy for a single element.
MLT	Calculate superlattice reflectivity vs angle.

TABLE III. Programs based on structure factor calculations.

Name	Description
STF	Calculate structure factors for an arbitrary crystal.
POWDER	Generate powder pattern delta functions for arbitrary crystal.
XDF	Calculate x-ray rocking curves for a dynamically scattering crystal.
XDSCAN	Calculate scattering vs energy for dynamically scattering crystal.

tering factor dependence on scattering angle and wavelength has been calculated by Cromer and Mann.[5] There is also a angle and wavelength dependence to the Compton cross section, which has been parametrized by Balyuzi.[6] There is a separate data base for those parameters and the subroutines which access them are also included in the shareable image library.

At its simplest, what these data bases achieve is the ability to calculate an absorption length for an arbitrary material at an arbitrary wavelength between roughly 100 eV and 1 MeV. If one is interested in the atomic scattering factors of crystals then the other components of the data bases become important. The entire package, built as an installation kit, consists of the data bases, the shareable image library, a program which is installed as a command similar to COPY or DIRECTORY for calculating absorption, and a help library which describes the program and how to access the data bases using the subroutines. The program, called ABSORPTION, calculates the transmitted intensity for a given thickness of material and lists the absorption length for the specified energy. It also lists the K and L edge energies (unless they do not exist). There is an extensive help library which is installed with the VMS command and which describes the specifics of the calling sequence. The driver program for the ABSORPTION package is also included in the installation kit as an example of how to use the routines. It should be made clear that close to an absorption edge the calculated and parametrized values will be inaccurate. Any variation in absorption due to crystal structure effects will not be present in the calculated values.

II. PROGRAMS BASED ON f' AND f''

A number of programs have been developed which take advantage of the subroutine calls described here. All the programs described below are written in FORTRAN. The simplest of these, TREF, involves calculating the reflectivity, penetration depth, and electric field intensity as a function of angle of incidence for an arbitrary material. This program is quite useful for research in grazing incidence x-ray scattering as it predicts the critical angle for total external reflection as well. The reflectivity as a function of energy, EREF, can also be calculated (Tables II–IV). This program is quite handy for determining synchrotron radiation mirror characteristics. The subroutines are also used for a program, MLT, to calculate multilayer reflectivities based on a paper by Bartels et al.[7]

III. CRYSTAL SCATTERING FACTORS

Perhaps the most significant capability these data bases enable is to calculate the crystal structure factor for an arbitrary set of miller indices at an arbitrary energy. The general formula for calculating crystal structure factors from the atomic positions for an arbitrary unit cell can be found in most standard texts on crystallography or x-ray diffraction.[8] We have developed a standardized data file for crystal lattices which defines the unit cell and the number, type, and position of the atoms within the unit cell. We have found it useful to include other parameters characteristic of a given crystal such as it elastic moduli and average coefficient of thermal expansion. The effect of thermal vibrations of the atoms in the lattice is treated by including a Debye temperature. Dynamical diffraction theory is germane to the case of perfect crystal monochromators, polarimeters, and spectrometers which are often the ideal choice of optical elements for synchrotron radiation beam lines. A main calling program, STF, has been written to permit interactive calculation of the complex structure factors and other parameters related to x-ray reflection and diffraction. The CHIs are the Fourier coefficients of the susceptibility[9] which are related to the structure factor. STF is for use with other programs requiring manual input of the crystal structure factors, etc.

IV. PROGRAMS BASED ON STF

Several main FORTRAN programs have been written to use the crystal structure factors directly from the subroutines. Three of these are discussed here, namely POWDER for calculating powder diffraction patterns, XDF which calculates perfect crystal Darwin–Prins curves and derives parameters descriptive of the diffraction performance of a particular crystal reflection, and XDSCAN which tabulated these same parameters as a function of photon energy. POWDER calculates the intensity of all the allowed reflections for a crystal given an incident photon energy. Multiplicities are summed (and noted in the data file). XDF calculates the single crystal Darwin–Prins curve using a modified set of equations taken from a paper by Bonse and Graeff.[10,11] The user has the option of performing the calculation for linearly polarized x rays, either s or p, or for unpolarized x rays. In addition to selecting the crystal and Miller planes of interest, the crystal temperature, asymmetry, and thickness are also variables. Both Bragg and Laue cases of diffraction are treatable, although XDF does not include multibeam cases nor cases involving grazing an-

TABLE IV. Crystal data file. This is a complicated crystal with a noncubic structure, with three bases but only one atom per basis. For each basis the atomic number of the atom and the fraction of that atom present is used, followed by the number of positions in that basis and the coordinates of each position.

LiNbO$_3$							
5.14739	5.14739	13.85614	90.00	90.00	120.00	293.00	5.
3	10.000	5.000	2.500		!imaginary isotropic matl.		
3	1	1	1				
Atoms in basis #1					!Ref: Abrahams Acta Cryst. B42 p. 61 ('86) (T-debye unk.)		
3	1.00						
6							
0.00000,	0.00000,	0.27872			!Li (6a3)	0,0,z = 0.27872	
0.00000,	0.00000,	0.77872			!	0,0,z + 1/2 = 0.77872	
0.66667,	0.33333,	0.61205			!	2/3,1/3,z + 1/3 = 0.61205	
0.66667,	0.33333,	1.11205			!	z + 5/6 = 1.11205	
0.33333,	0.66667,	0.94539			!	z + 2/3 = 0.94539	
0.33333,	0.66667,	1.44539			!	z + 7/6 = 1.44539	
Atoms in basis #2							
41	1.00						
6							
0.00000,	0.00000,	0.00000			!Nb (6a3)	z = 0.0	
0.00000,	0.00000,	0.50000			!	z + 1/2	
0.66667,	0.33333,	0.33333			!	z + 1/3	
0.66667,	0.33333,	0.83333			!	z + 5/6	
0.33333,	0.66667,	0.66667			!	z + 2/3	
0.33333,	0.66667,	1.16667			!	z + 7/6	
Atoms in basis #3							
8	1.00						
18							
0.04757,	0.34328,	0.06336			!O(18b1) x = 0.04757, y = 0.34328, z = 0.06336		
− 0.34328,	− 0.29571,	0.06336			!	$-y, x - y, z$	
0.29571,	− 0.04757,	0.06336			!	$-x + y, -x, z$	
− 0.34328,	− 0.04757,	0.56336			!	$-y, -x, z + 1/2$	
0.29571,	0.34328,	0.56336			!	$-x + y, y, z + 1/2$	
0.04757,	− 0.29571,	0.56336			!	$x, x - y, z + 1/2$	
0.71424,	0.67661,	0.39669			!	$x, y, z + 2/3, 1/3, 1/3$	
0.32339,	0.03762,	0.39669			!	$-y, x - y, z + 2/3, 1/3, 1/3$	
0.96238,	0.28576,	0.39669			!	$-x + y, -x, z + 2/3, 1/3, 1/3$	
0.32339,	0.28576,	0.89669			!	$-y, -x, z + 1/2 + 2/3, 1/3, 1/3$	
0.96238,	0.67661,	0.89669			!	$-x + y, y, z + 1/2 + 2/3, 1/3, 1/3$	
0.71424,	0.03762,	0.89669			!	$x, x - y, z + 1/2 + 2/3, 1/3, 1/3$	
0.38090,	1.00995,	0.73003			!	$x, y, z + 1/3, 2/3, 2/3$	
− 0.00995,	0.37096,	0.73003			!	$-y, x - y, z + 1/3, 2/3, 2/3$	
0.62904,	0.61910,	0.73003			!	$-x + y, -x, z + 1/3, 2/3, 2/3$	
− 0.00995,	0.61910,	1.23003			!	$-y, -x, z + 1/2 + 1/3, 2/3, 2/3$	
0.62904,	1.00995,	1.23003			!	$-x + y, y, z + 1/2 + 1/3, 2/3, 2/3$	
0.38090,	0.37096,	1.23003			!	$x, x - y, z + 1/2 + 1/3, 2/3, 2/3$	

gles. After calculating the Darwin–Prins curve for the reflection, the XDF program uses the curve to extract a number of useful diffraction parameters. These include the angle and energy integrated reflectivities,[12] Darwin–Prins angular width, energy resolving power, and the index of refraction shift of the Bragg angle. The polarization selectivity, defined as the ratios of the integrated reflectivity for the two linear polarizations can also be found. XDSCAN is based on the XDF program. Its purpose is to produce the photon energy variation of the diffraction parameters. This is particularly useful in testing the spectral performance of crystals for use as monochromators, spectrometers, or polarizers.[13]

V. CONCLUSIONS

The subroutines and programs described above have been in use at SSRL, NIST, ESRF, and other institutions for some time. The applications for which calculations have been performed ranges from studies of novel monochromator crystals to determining index of refraction corrections for high precision x-ray spectroscopy. The authors are willing to provide the ABSORPTION package as an installation package for VAX computers running the VMS operating system. Other codes based on that package are available upon request. Comments on possible improvements or bugs are invited. A number of related programs with other capabilities which have been adapted to the basic subroutines will be described elsewhere.[14]

[1] D. T. Cromer and D. A. Liberman, Acta Crystallogr. Sect. A **37**, 267 (1981). The original data and calling routine for calculating f' and f'' was obtained from D. T. Cromer by P. H. Fuoss as a stack of punch cards. In 1987, during a visit to Hasylab by the authors, the program was converted to F77 and direct access data base format. It has been upgraded extensively since then.

[2] D. T. Cromer, Acta Crystallogr. **18**, 17 (1965).

[3] D. T. Cromer and D. Liberman, J. Chem. Phys. **53**, 1891 (1970).

[4] W. H. McMaster, N. Kerr Del Grande, J. H. Mallett, and J. H. Hubbell, UCRL-50174 SEC II REV I, available from National Technical Information Service, U. S. Dept. of Commerce.

[5] D. T. Cromer and J. B. Mann, Acta Crystallogr. Sect. A **24**, 321 (1968).

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[7] W. J. Bartels, J. Hornstra, and D. J. W. Lobeek, Acta Crystallogr. Sect. A **42**, 539 (1986).

[8] B. E. Warren, *X-ray Diffraction* (Addison-Wesley, Reading, MA, 1969). (Now available in a Dover paperback edition.)

[9] Z. G. Pinsker, *Dynamical Scattering of X Rays in Crystals* (Springer, Berlin, 1978).

[10] U. Bonse and W. Graeff, in *X-ray Optics*, edited by H. J. Queisser (Springer, Berlin, 1977).

[11] O. Brümmer, H. R. Höche, and J. Nieber, Phys. Status Solidi A **37**, 529 (1976).

[12] A discussion of the definitions and applications of angle integrated reflectivity and energy integrated reflectivity will be published elsewhere.

[13] S. Brennan, P. L. Cowan, R. D. Deslattes, A. Henins, D. W. Lindle, and B. A. Karlin, Rev. Sci. Instrum. **60**, 2243 (1989).

[14] P. L. Cowan and S. Brennan (in preparation).