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ANALYSIS OF SPALLATION TIME-OF-FLIGHT (TOF) NEUTRON DATA FROM A 2-D POSITION-SENSITIVE DETECTOR

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Abstract - The single crystal diffractometer at the Argonne Intense Pulsed Neutron Source is based on the time-of-flight (TOF) Laue technique and employs a 30 x 30 cm. position-sensitive scintillation detector. The methods we use for producing reciprocal space intensity plots and for obtaining integrated structure factor amplitudes will be described.

I - INTRODUCTION

At the Intense Pulsed Neutron Source (IPNS), a target of depleted uranium is bombarded by high energy protons to produce intense bursts of neutrons by spallation./1,2/ Surrounding the target are four moderators, each with several beam lines which direct neutrons to the various instruments. At a distance \( z \) from the moderator, the wavelength \( \lambda \) of a neutron is determined by the de Broglie equation:

\[
\lambda = \frac{h}{m \cdot \frac{2}{t}}
\]

where \( h \) is Planck's constant, \( m \) is the neutron mass, and \( t \) is the TOF for flight path \( z \). The single crystal diffractometer (SCD) is based on the TOF Laue technique./3/ In this paper, we will describe some of the procedures we use to analyze data obtained with the SCD.

II - DATA COLLECTION

The SCD employs a position-sensitive \(^{6}\text{Li}\)-glass scintillation detector/4/ which is typically centered on the 90° scattering angle direction at a distance of 32 cm. from the crystal (Fig. 1). The scintillation glass is 30 x 30 cm., but due to edge effects, the active area is reduced to about 27 cm. in each dimension for actual data collection. Each neutron is characterized by three digitized coordinates representing position on the detector (X and Y) and TOF (T). For each stationary crystal orientation, data are accumulated for 2-12 hours (depending on sample size, unit cell size, etc.) in a histogram with typical channel dimensions of 85 x 85 x 120 corresponding to X, Y and T respectively. A full hemisphere of data requires up to 35 crystal settings, i.e., 35 histograms.

Conceptually, the histogram is constructed of a stack of 120 two-dimensional "time-slices." The time-slices are usually set up with variable widths (\( \Delta t \)) such that \( \Delta t / t = 0.015 \). With a sample-to-moderator distance of 660 cm., a sample-to-detector distance of 32 cm., and a nominal wavelength range of 0.7 to 4.2 \( \AA \), neutrons with TOF's between 1.2 and 7.3 millisecond. after \( t_0 \) (the start of the pulse) are histogrammed, and \( \Delta t \) varies from 18 to 110 \( \mu \text{sec.} \)

Associated with each time-slice is the fractional dead-time loss for that particular time-slice. These values are obtained during data collection by empirically determining the percentage of test signals which are rejected because
Fig. 1 - A schematic representation of the single crystal diffractometer at the IPNS. Typical distances for $a$ and $b$ are 660 and 32 cm., respectively. A low efficiency beam monitor (not shown) is located about 30 cm. from the crystal towards the source.

the detector and the digitizing electronics are busy. A recent upgrade of the detector has reduced the dead-time from 12 μsec. down to 3, such that the maximum dead-time losses are usually not more than 5%.

Each histogram is archived on high density magnetic tape, which can hold up to 50 histograms per reel. The data are analyzed offline on a VAX11/780 computer, as described below.

III - PEAK SEARCH AND AUTO-INDEXING

Our peak search program, PEKSER, is based on subroutine FRPEAK from the program FOURIER, written by R. J. Dellaca and W. T. Robinson at the University of Canterbury (New Zealand), which is derived from A. Zalkin's FORDAP. The program outputs the locations of up to 50 of the most intense peaks in a histogram. Typically, 5 to 10 low order reflections can then be input into the auto-indexing program BLIND,5 which was modified to accept Laue data. By including all the strong reflections from all the histograms in the least-squares program LSQRS, the accuracy of the unit cell parameter and the orientation matrix is improved.

IV - RECIPROCAL SPACE PLOTS

A highly useful feature of the TOF Laue data obtained with an area detector is that the histograms contain intensity information for a solid volume of reciprocal space including the regions between the fundamental Bragg reflections. This allows the user to examine the histogram offline for extra Bragg or diffuse scattering, which is especially useful for searching for structural phase transitions at low temperatures.6-8

The program RLPLN asks the user to define a plane in reciprocal space in terms of Miller indices, which may be simply setting $h$, $k$ or $l$ to a constant, or possibly requiring $h = k$ for an $hh\alpha$ plot. The user also defines the range and the increments along each axis. In the example in Figure 2, $h = 4.93$, $k = -4.5$ to $-1.5$ in steps
of 0.025 and \( \ell = -4.0 \) to 1.0 in steps of 0.04. The program sets up, in this case, a 121 x 126 array and evaluates the intensity at each point by using the previously determined orientation matrix to calculate its location in the histogram in terms of non-integer histogram coordinates \( x,y,z \). The intensity at \( xyz \) is then obtained by interpolating in three-dimensions using the formula:

\[
I(xyz) = \frac{\sum_i [C(XYT)_i/(D(XYT)_i)^2]}{\sum_i [(1/(D(XYT)_i)^2]}
\]  

(2)

where \( C \) is the neutron count at \( XYT \) normalized for its time-slice width, \( D \) is the distance in histogram coordinates between \( xyz \) and \( XYT \), and the summations are over the nearest integer location to \( xyz \), \((XYT)_i\), and the surrounding 26 nearest neighbor locations. Locations that fall outside the histogram limits are given intensity values of zero. The array is plotted as a 3D intensity plot with a 100 x 100 grid using DISSPLA graphics routines (Fig. 2).

This program strategy provides qualitative or semi-quantitative plots which in most cases are sufficient. If a more quantitative intensity plot is required, corrections for the incident spectrum, the detector efficiency, dead-time losses and the Lorentz factor can be applied.

V - PEAK INTEGRATION AND REDUCTION

For a complete structure analysis, integrated Bragg intensities are obtained by calculating (using the orientation matrix) the locations of all possible reflections in each histogram and integrating around the predicted histogram coordinates. Because the wavelength resolution from the source, \( \Delta \lambda/\lambda \), is nearly constant in the thermal range, and since the time-slice widths vary such that \( \Delta t/t \) is constant, the dimensions in histogram coordinates of all peaks are also fairly constant. The integration limits in terms of histogram coordinates are defined by the user after examination of several reflections. Background points are the histogram locations surrounding the peak integration rectangle on each time-slice.

For each reflection \( h\k\ell \), the program checks the calculated positions of its six nearest neighbors \( h\pm1, k\pm1, \pm\ell \) and tests for possible overlap of the integration envelopes. If the peak regions of two reflections overlap, both reflections are rejected. If the peak region of one reflection overlaps the background region of a second reflection, then only those background points which are affected are not included in the integration. Each count used in the integration is normalized for its time-slice width.

The conversion of integrated intensities to structure factor amplitudes is based on the Laue formula:

\[
I_{h\k\ell} = kT(\lambda)\psi(\lambda)\varepsilon(\lambda,r)A(\lambda)\gamma(\lambda)|F_{h\k\ell}|^2 \lambda^4/\sin^2\theta
\]  

(3)

where \( k \) is a scale factor, \( T \) is the normalized monitor count and \( F_{h\k\ell} \) is the structure factor. The wavelength-dependent factors are the dead-time loss correction \( \tau(\lambda) \), the incident flux \( \psi(\lambda) \), the detector efficiency \( \varepsilon(\lambda,r) \), the absorption correction \( A(\lambda) \), and the extinction correction \( \gamma(\lambda) \). Most of the factors in equation (3) are fairly straightforward to measure or calculate, or have been described previously. However, \( \psi(\lambda) \) and \( \varepsilon(\lambda,r) \) require some additional discussion.

Because the detector is flat, not curved, the pathlength through the \( ^6\text{Li} \)-glass of a neutron scattered from the crystal varies with position \( r \) on the detector, and \( \varepsilon \) is not only a function of wavelength \( \lambda \), but also of \( r \). Figure 3 shows the calculated detector efficiencies for a quadrant of the detector based on the formula:
Fig. 2 - Plot of the intensity distribution in the \( h = 4.93 \) reciprocal lattice plane of \( \beta-(ET)_{2}I_{3} \) at 20 K. Satellite peaks at \( (5,2,3)-q, (5,2,1)-q \) and \( (5,4,0)-q \) are clearly observable, where \( q = (0.07,0.27,0.21) \). The \( (hka)+q \) satellites are observed in a plot of the \( h = 5.07 \) plane (not shown).

\[
\epsilon(\lambda, r) = 1 - e^{-\sigma(\lambda) \xi(r)}
\]  

where \( \sigma(\lambda) \) is the linear absorption coefficient in the \( ^{6}\text{Li} \)-loaded glass at wavelength \( \lambda \) and \( \xi(r) \) is the slant path through the detector for neutrons scattered from the sample and entering the detector at position \( r \). At longer wavelengths, all efficiencies will approach 100\% such that the dependence on position will be negligible, but at the shorter wavelengths of the thermal spectrum the differences are significant. An analogous map to the one in Figure 3 is calculated for each time-slice of a histogram and the Bragg data are individually corrected for \( \epsilon(\lambda, r) \) based on the wavelength and detector position at which they were observed.

The incident spectrum term \( \phi(\lambda) \) is based on incoherent scattering data from a 3 mm sphere of vanadium alloyed with 7 at.\% niobium to give zero coherent scattering. The vanadium data are also corrected for \( \epsilon(\lambda, r) \), in addition to corrections for the vanadium sphere absorption and dead-time losses. The final values for the incident source spectrum are obtained after subtracting a background spectrum (no sample in the sample position) from the vanadium spectrum.

We have recently collected data at 20 K on a 2 mm\(^3\) crystal of \( \beta-(ET)_{2}I_{3} \cdot Br \) (ET is bis(ethylenedithio)tetrathiafulvalene, or BEDT-TTF) which has one formula weight of \( C_{26}S_{2}H_{1}I_{2}Br \) per unit cell. The crystals are triclinic, \( \pi \), with a unit cell volume of \( 810^{-3} \)\( \text{Å}^3 \) and with 28 independent atoms. Data in each of the 35 histograms was accumulated for about 10 hours. Of the 5849 integrated reflections, the 3285 with \( F^2 > 5\sigma(F^2) \) were used to refine 285 variables which yielded \( R(F) = 0.053 \) and \( R_w(F^2) = 0.098 \). The standard deviations are \( \pm 0.002 \) \( \text{Å} \) for C-C bonds and \( \pm 0.004 \) \( \text{Å} \) for C-H bonds.
**VI - CONCLUSIONS**

As described above, satisfactory results can be obtained with the SCD, although they indicate that higher flux and better resolution are desirable for full structural analyses. In addition, a technique for accurately abstracting the intensities of weak reflections from the data would be useful. At the IPNS, a factor of 3 in flux may be achieved in 1986 with a new enriched uranium target. At Los Alamos LANSCE and the Rutherford SNS spallation sources, fluxes orders of magnitude greater than that at IPNS may be realized within the coming years. Thus, the potential of the technique described in this paper may still lie mostly in the future.

For surveying reciprocal space and studying structural phase transitions, we believe the SCD has already proven to be highly successful. We are in the process of providing additional ancillary equipment for high pressure, and lower and higher temperature studies. We also plan to increase the source-to-sample flight path to provide better resolution when the new target is installed.

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