

WRDC-TR-90-4069

ADA 231802



**A USER'S MANUAL FOR FIBER DIFFRACTION:
THE AUTOMATED PICKER AND HUBER DIFFRACTOMETERS**

**P. Galen Lenhert
W. Wade Adams
Polymer Branch
Nonmetallic Materials Division**

July 1990

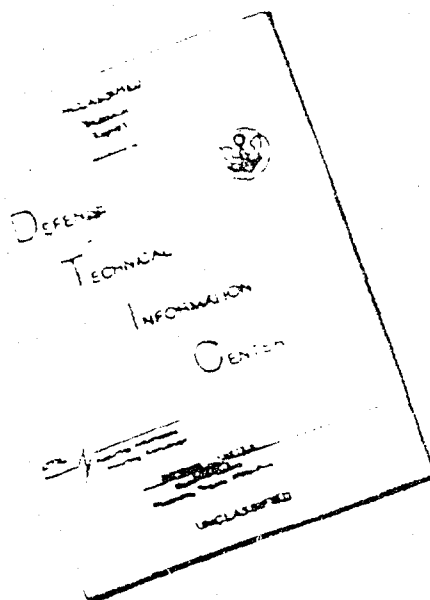
Final Report for the Period July 1986 - June 1990

Approved for public release; distribution unlimited

**MATERIALS LABORATORY
WRIGHT RESEARCH AND DEVELOPMENT CENTER
AIR FORCE SYSTEMS COMMAND
WRIGHT-PATTERSON AIR FORCE BASE, OHIO 45433-6533**

20040219291

DISCLAIMER NOTICE



THIS DOCUMENT IS BEST
QUALITY AVAILABLE. THE COPY
FURNISHED TO DTIC CONTAINED
A SIGNIFICANT NUMBER OF
PAGES WHICH DO NOT
REPRODUCE LEGIBLY.

REPRODUCED FROM
BEST AVAILABLE COPY

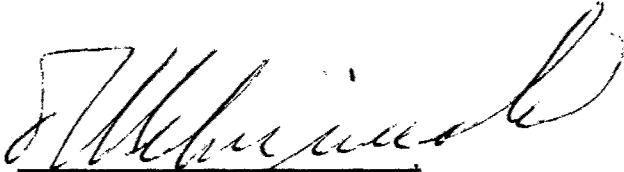
NOTICE

WHEN GOVERNMENT DRAWINGS, SPECIFICATIONS, OR OTHER DATA ARE USED FOR ANY PURPOSE OTHER THAN IN CONNECTION WITH A DEFINITELY GOVERNMENT-RELATED PROCUREMENT, THE UNITED STATES GOVERNMENT INCURS NO RESPONSIBILITY OR ANY OBLIGATION WHATSOEVER. THE FACT THAT THE GOVERNMENT MAY HAVE FORMULATED OR IN ANY WAY SUPPLIED THE SAID DRAWINGS, SPECIFICATIONS, OR OTHER DATA, IS NOT TO BE REGARDED BY IMPLICATION, OR OTHERWISE IN ANY MANNER CONSTRUED, AS LICENSING THE HOLDER, OR ANY OTHER PERSON OR CORPORATION; OR AS CONVEYING ANY RIGHTS OR PERMISSION TO MANUFACTURE, USE, OR SELL ANY PATENTED INVENTION THAT MAY IN ANY WAY BE RELATED THERETO.

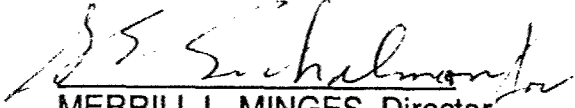
THIS REPORT HAS BEEN REVIEWED BY THE OFFICE OF PUBLIC AFFAIRS (ASD/PA) AND IS RELEASABLE TO THE NATIONAL TECHNICAL INFORMATION SERVICE (NTIS). AT NTIS, IT WILL BE AVAILABLE TO THE GENERAL PUBLIC INCLUDING FOREIGN NATIONALS.

THIS TECHNICAL REPORT HAS BEEN REVIEWED AND IS APPROVED FOR PUBLICATION.


R. C. EVERS
Polymer Branch
Nonmetallic Materials Division


T. E. HELMINIAK, Chief
Polymer Branch
Nonmetallic Materials Division

FOR THE COMMANDER


MERRILL L. MINGES, Director
Nonmetallic Materials Division

If your address has changed, if you wish to be removed from our mailing list, or if the addressee is no longer employed by your organization, please notify WRDC/MLBP, Wright-Patterson AFB, OH 45433-6533 to help us maintain a current mailing list.

Copies of this report should not be returned unless return is required by security considerations, contractual obligations, or notice on a specific document.

UNCLASSIFIED

SECURITY CLASSIFICATION OF THIS PAGE

REPORT DOCUMENTATION PAGE				Form Approved OMB No. 0704-0188	
1a. REPORT SECURITY CLASSIFICATION Unclassified			1b. RESTRICTIVE MARKINGS		
2a. SECURITY CLASSIFICATION AUTHORITY			3. DISTRIBUTION/AVAILABILITY OF REPORT Approved for public release; Distribution Unlimited		
2b. DECLASSIFICATION/DOWNGRADING SCHEDULE					
4. PERFORMING ORGANIZATION REPORT NUMBER(S) WRDC-TR-90-4069			5. MONITORING ORGANIZATION REPORT NUMBER(S)		
6a. NAME OF PERFORMING ORGANIZATION Wright Research & Development Center, Materials Laboratory		6b. OFFICE SYMBOL (If applicable) WRDC/MLBP	7a. NAME OF MONITORING ORGANIZATION Same as 6a		
6c. ADDRESS (City, State, and ZIP Code) Wright-Patterson Air Force Base Ohio 45433-6533			7b. ADDRESS (City, State, and ZIP Code)		
8a. NAME OF FUNDING/SPONSORING ORGANIZATION Same as 6a		8b. OFFICE SYMBOL (If applicable)	9. PROCUREMENT INSTRUMENT IDENTIFICATION NUMBER		
8c. ADDRESS (City, State, and ZIP Code)			10. SOURCE OF FUNDING NUMBERS		
PROGRAM ELEMENT NO. 61102F		PROJECT NO. 2303	TASK NO. Q3	WORK UNIT ACCESSION NO. 07	
11. TITLE (Include Security Classification) A User's Manual for Fiber Diffraction: The Automated Picker and Huber Diffractometers					
12. PERSONAL AUTHOR(S) P. Galen Lenhert* and W. W. Adams					
13a. TYPE OF REPORT Final		13b. TIME COVERED FROM Jul 86 TO Jun 90		14. DATE OF REPORT (Year, Month, Day) July 1990	
15. PAGE COUNT 45					
16. SUPPLEMENTARY NOTATION *On leave from Dept. of Physics, Vanderbilt Univ., as an AFOSR Univ. Resident Research Program Fellow, July 1988 - June 1990					
17. COSATI CODES			18. SUBJECT TERMS (Continue on reverse if necessary and identify by block number)		
FIELD	GROUP	SUB-GROUP			
11	05		Polymer Morphology Modulus Fourier Analysis		
07	04		Fiber Diffraction Diffractometer Orientation		
			X-Ray Diffraction Automated		
19. ABSTRACT (Continue on reverse if necessary and identify by block number) This report describes the automated Picker and Huber X-ray diffractometer systems in use at the Materials Laboratory Polymer Branch. The diffractometer hardware is described from the user's point of view and a complete description is given of the use of the operational software for both the Picker and Huber systems. The control computer is a VAX 11/730. The use of the system to collect polymer fiber diffraction data, especially 2θ and χ scans and layer line scans. A device for applying tension to the fiber sample allows studies of the effect of tension on fiber morphology as seen by diffraction methods. Analysis programs developed to process the diffraction data from the Picker and Huber systems are explained and examples are given of program use, and in some cases, the results obtained are presented. An Appendix is included which contains a discussion of some of the hardware adjustments that may be required. The organization of the FORTRAN programs used by the VAX computer to control the diffractometer functions is explained. Utility programs for making various operational tests are also covered.					
20. DISTRIBUTION/AVAILABILITY OF ABSTRACT ☒ UNCLASSIFIED/UNLIMITED ☐ SAME AS RPT. ☐ DTIC USERS			21. ABSTRACT SECURITY CLASSIFICATION Unclassified		
22a. NAME OF RESPONSIBLE INDIVIDUAL W. W. Adams			22b. TELEPHONE (Include Area Code) (513)255-9148		22c. OFFICE SYMBOL WRDC/MLBP

FOREWORD

This report was prepared by the Polymer Branch, Nonmetallic Materials Division. The work was initiated under Project No. 2303, "Nonmetallic and Composite Materials," Task No. 2303Q3, Work Unit Directive (WUD) 2303Q307, "Structural Resins." It was administered under the direction of the Materials Laboratory, Wright Research and Development Center, Air Force Systems Command, Wright-Patterson Air Force Base, Ohio with Dr R. C. Evers as the Materials Laboratory Project Scientist (WUD Leader). Coauthors were Dr P. Galen Lenhert, Department of Physics, Vanderbilt University, Nashville, TN 37235, on leave as an AFOSR University Resident Research Program (URRP) Fellow, July 1988-June 1990, and Dr W. Wade Adams, Materials Laboratory (WRDC/MLBP). This report covers research conducted from July 1986 to June 1990.

The authors thank Mr Gary Price, University of Dayton Research Institute, for technical assistance, and Mr Joseph O'Brien, Polymer Branch, for assistance with initial computer system analysis. Thanks are also due to Dr Chuck Strause of Crystal Logic, Inc., Los Angeles, CA for many helpful discussions concerning the diffractometer automation.

TABLE OF CONTENTS

SECTION.....	PAGE
1. INTRODUCTION.....	1
2. HARDWARE NOTES	2
Diffractometer Angles	2
Settings for the Picker and Huber Detector Circuits	2
Collimator, Beta Filter and Monochromator	3
The FDD	3
3. CONTROL PROGRAMS for the PICKER, HUBER & FDD.....	4
The Manual Programs, PIKMAN, HUBMAN and MOTOR.....	4
Running PIKMAN or HUBMAN	4
Running MOTOR	5
4. DATA COLLECTION with PICKER or HUBER.....	6
General Comments on PICKER and HUBER	6
Preparation of Fiber Samples.....	6
The Menu	7
5. THE X-RAY MODULUS PROGRAM.....	12
6. DATA ANALYSIS PROGRAMS.....	14
Utility Plotting Program, PLOT1	14
2 θ Scans - Data Reduction and Curve Fitting, FIT	14
Layer Line Scans and Plots, LLPLOT.....	17
χ Scans, an Instrumental Broadening Correction	19
The Programs, FITSM and UNFOLD	19
An Example, KEVLAR 149 χ Scans.....	20
7. REFERENCES	24
8. APPENDICES	25
A. THE CRYSTAL LOGIC HARDWARE SYSTEM.....	25
Picker and Huber Adjustments	25
FDD Adjustments and Hardware.....	26
B. COMPUTER PROGRAM ORGANIZATION.....	26

TABLE OF CONTENTS (Continued)

C. UTILITY PROGRAMS	28
Stability Test of the Picker Mount, STABLE.....	28
Angle Driver Test Program, ANGTST.....	29
Alignment Software for the Picker and Huber, PALIGN & HALIGN	29
D. ADJUSTING the AMPLIFIER and PHA SETTINGS	30
E. DEAD TIME DETERMINATION and CORRECTION	31
Method of Chuck Strouse.....	31
Rates for Strong and Weak "Reflections" on the Picker System.....	32
Correction for Coincidence Loss.....	33
F. RU 200 X-RAY STABILITY TEST.....	33
G. HUBER ALIGNMENT NOTES.....	34
H. BUILDING EXE FILES.....	37
I. VAX PORT PARAMETERS.....	38

LIST OF TABLES

TABLE	PAGE
1. SCAN.IN file for PICKER.....	6
2. TTSDAT.TMP file showing the diffractometer set up and sample information.....	11
3. Selected portions of the output file, FOR007.DAT of FIT	16
4. KEVLAR 149 χ SCANS at 20.55° 2 θ	23
5. The four lines in this table show the format of the files HKL.DAT, PEAKS., CPEAKS. and LEAST., respectively.	30

LIST OF ILLUSTRATIONS

FIGURE	PAGE
1. Diagram showing a simple sample support.....	7
2. Plot generated by FIT.....	17
3. Layer line scan of degummed silk (<i>Bombyx mori</i>).....	18
4. χ scan, KEVLAR 149 equatorial reflection at 20.55° 2 θ	21
5. χ scan of the KAP reflection 19.93° 2 θ	22
6. χ intensity distribution, KEVLAR 149 equatorial peak at 20.55° 2 θ with instrumental and sample size factors removed.....	22
7. Count rate at 400 second intervals.....	34
8. Count rate at 1000 second intervals.	34

1. INTRODUCTION

The Materials Laboratory Polymer Branch facilities include two systems for X-ray diffraction measurements on polymers, the Picker and Huber Diffractometers. Both are used with the same rotating anode X-ray generator and are driven by the VAX 11/730 computer through similar interfaces supplied by *Crystal Logic, Inc.*⁺ The software for these systems is similar because most features of the computer-diffractometer interfaces are identical. The Picker is equipped with an automated fiber deformation device (FDD). The purpose of the FDD is to allow measurement of fiber diffraction as a function of tension.

The user communicates with both the Huber and Picker systems (and the FDD) *via* a VAX terminal used in the interactive mode. The control programs reside in the VAX and information and commands pass to and from the Picker through the VAX port TXA0:. TXA1: is used for the Huber and TXA2: for the FDD. Since the connecting port for the diffractometer must be assigned to FORTRAN unit FOR001, only one diffractometer may be accessed from any account at one time. Accounts named PICKER and HUBER are used for the respective systems.

For routine use, the Picker diffractometer is accessed through the PICKER account on the VAX computer. On login the required VAX port TXA0: is automatically allocated and assigned to FOR001 by means of commands in a LOGIN.COM file. The Picker can then be controlled *via* the programs PIKMAN and PICKER and for X-ray modulus measurements, XRYMOD (see below). Use of the Huber diffractometer is through the VAX account HUBER, which makes, on login, the required allocation and assignment of TXA1: and provides access through the programs HUBMAN and HUBER (also described below).

The present Picker diffractometer with VAX-*Crystal Logic* automation replaced earlier automation with a PDP-8/I computer using a Picker Nuclear FACS-I interface. The use of the earlier system was reported previously (Lenhart, O'Brien and Adams, 1986).

**BE SURE THE SHUTTER IS CLOSED WHEN YOU REMOVE THE
COLLIMATOR.**

**TO PREVENT AN IMMINENT COLLISION, TURN THE INTERFACE
OFF.**

TO PREVENT A COLLISION, PRESS ONE OF THE LIMIT SWITCHES.

⁺ *Crystal Logic Inc.*, 10573 West Pico Blvd., Los Angeles, CA 90064.

2. HARDWARE NOTES

Diffraction Angles

The diffractometer circles are driven by stepping motors controlled by the VAX. The computer has no way to tell what the initial angle values are; therefore, these must be provided when you enter the program. This can be done in any of several ways as explained below. As soon as the interface is turned on, the stepping motors are powered up and therefore locked in position. At this point the motors can be driven only by commands from the computer. If you want to change the angles manually, you must do so before turning the interface on. The Picker angles can be easily changed if the interface is off. 2θ and ω can be changed with the cranks, χ with the small knob on the far side of the χ circle and ϕ with the large gear on the χ circle. One can also change χ and ϕ on the Picker by decoupling the appropriate gear. To change the angles on the Huber, one would normally decouple the gears from the circles.

The main control programs and the hardware drivers are in FORTRAN. The main control programs were written in the Materials Laboratory, the hardware drivers, supplied by *Crystal Logic*, have been modified slightly. The control programs for the Picker and Huber use the same code, only the hardware-related subroutines differ for the two systems.

The principal difference between the Picker and the Huber instruments (from the programming standpoint) is the θ - 2θ coupling. (On both instruments 2θ measures the motion of the detector arm with respect to the X-ray beam direction.) On the Picker, the Bragg condition is maintained mechanically when 2θ is driven. This means that the χ circle stays in the bisecting position when 2θ is changed, *i.e.*, ω remains at zero. (On the Picker ω measures the angle between the bisecting position for the χ circle and its actual position.) On the Huber, 2θ is driven independently of ω . That is, when 2θ is driven, the χ circle is stationary, *i.e.*, ω remains at zero. (On the Huber, ω measures the angle between the normal to the χ circle and the X-ray beam.) In order to maintain the Bragg condition on the Huber, one must drive ω at half the rate of 2θ . Normally the subroutine BRAG2T is used to drive 2θ for both the Picker and the Huber. It handles the ω setting so as to maintain the Bragg condition for both instruments.

Settings for the Picker and Huber Detector Circuits

The High Voltage Power Supply settings that will give satisfactory results with $\text{CuK}\alpha$ are: coarse HV, 500; mid range HV, 100; fine HV, 2.00.

The Log/Linear Ratemeter settings are: zero suppression, 0; range and time constant, as needed; audio threshold, as needed to give sound at the count rate you are working with or expect to obtain; audio volume, at maximum if you want to hear your counts in the X-ray laboratory.

The Amplifier and Pulse Height Analyzer settings depend on the radiation in use. For $\text{CuK}\alpha$, set the coarse gain at 50 and the fine gain at the third mark above 0.5. Set the switches to POS and UNI and WINDOW. A window of 4.5 and a Lower Level of 3.0 are about right. They should be adjusted as needed.

The above settings are approximate. The detailed procedure for setting the detector circuits using an oscilloscope is given in the Appendix D.

Collimator, Beta Filter and Monochromator

There is no monochromator on the Picker diffractometer so a Ni filter must be used if you want the Cu K β line eliminated. The filter provided contains two foils of Ni with a total thickness of 0.001 in. It is inserted into the incident beam collimator. Be careful of your sample when you remove the collimator. Remember also that you may want to use the 1.0 or 1.5 mm collimator. The collimators that are not in use are kept in the PICKER drawer. Be sure to use incident beam collimators in the incident beam and diffracted beam collimators in the diffracted beam! You can tell the difference because the diffracted beam collimators have two grooves cut in the center (non tapered) section. When you put the incident beam collimator back on the Picker diffractometer, be careful to clamp it in the correct position. The redesigned collimator clamp should insure this, but all counts can be lost if the collimator points a little too high or too low.

The Huber diffractometer is set up with a highly oriented graphite monochromator. An incident beam collimator with a 1.2 mm diameter is normally used. The diameter of the diffracted beam collimator is 2.0 mm.

In both instruments, the size of the collimators should relate to the sample size but the actual diffracted beam aperture is adjusted independently of the collimator. In the Picker system, a screwdriver is used to adjust the symmetrically variable aperture (SVA). The size of the opening is set by closing the aperture on drill rods to obtain the opening desired. The drill rods are labeled and kept in a holder mounted on the diffractometer base. In the Huber system, a pair of calibrated vertical and horizontal slits is used to set the diffracted beam aperture.

The FDD

A power supply and a signal amplifier made by Omega Engineering are used in the strain gauge bridge circuit of the FDD for measuring tension. The input voltage to the strain gauge bridge is adjustable and is continuously displayed on the Keithley 175 Multimeter which should be set to the 2 V scale. The Keithley 177 Microvolt DMM is set to the 20 mv scale to display the bridge output. The NRC micrometer can be driven by the FORTRAN subroutine MOTOR.FOR (see below) which is also used to read the strain gauge output. The components are packaged together in a chassis box. Details of the construction and use of the FDD are given in a separate report (Lenhert and Adams, 1990).

3. CONTROL PROGRAMS for the PICKER, HUBER & FDD

The Manual Programs, PIKMAN, HUBMAN and MOTOR

PIKMAN and HUBMAN allow the operator to test all the diffractometer functions for the Picker and Huber diffractometers from the computer terminal. The available commands, displayed in a menu, allow all angles to be set at zero (with the zero positions determined automatically by the zero sensors), any angle to be driven to any (permitted) position, the X-ray shutter to be opened or closed and counts to be registered and displayed on the terminal.

MOTOR is the manual program used to test and operate the automated FDD. It drives the micrometer motor in either direction, tests the motor status and reads the strain gauge bridge output.

The diffractometer angles at the end of any run are stored in a file, DIFF.;1. This allows a start up option, *i.e.*, "use the angle values stored in DIFF. as the initial angle settings". In order to avoid cluttering the computer disk, only the latest set of angles is saved. That is, any time the current angles are saved in DIFF., the old copy of DIFF.;1 is replaced by an up-to-date copy. If an abnormal program exit occurs, the previous angles stored in DIFF. will still be there but they will be incorrect. The user must then redetermine the zero settings by use of the zero angles option in PIKMAN or HUBMAN or, if the last angles are accurately known, they can be entered manually.

Running PIKMAN or HUBMAN

Log on to the VAX (account PICKER or HUBER, password DIFFRACTOMETER--for either). Assignment of the VAX port TXA0: or TXA1: is done automatically by commands in the LOGIN.COM file. Turn the interface on and RUN PIKMAN or HUBMAN. You will first be asked if the correct diffractometer angles are stored in DIFF.;1. If the last use of the diffractometer was from this account and there was a normal program exit, you should answer "Y". If the answer is "N" you will get the "are all angles zero?" choice. If they are not, the program will ask for the correct angles. Read them from the odometers or scales but unless you are very sure that the scales are correctly set, use the zero angle command "Z" as explained below. (Caution: on the Picker, ϕ is very difficult to read accurately---but ϕ may not matter for your particular experiment.) After the angles are initialized, the diffractometer functions available in the program will be displayed.

In using the zero angles command (Z), remember that all angles must be set at a small positive value before the "Z" command is given. Drive each angle to a positive setting of about 1° to 2° by using the "T", "O", "P" or "C" commands. (Be sure the wire to the ϕ coarse sensor--on the Picker--is connected and the χ circle is engaged near zero, not near 180° if these angles are to be set.) The "Z" command can now be given. You will observe that each angle drives in the negative direction, overshoots zero slightly, then backs up slowly to exactly zero. If true zero is different from the sensor position, a final change in the angle will be observed as the true zero setting is made. Each angle is zeroed automatically if you select option 5 after typing "Z". Caution: Be sure 2θ is zeroed before using "Z" to zero ω .

PIKMAN and HUBMAN can also be used to open and close the shutter ("S" command) and count repeatedly for as many one second intervals as you wish ("M" command). The "L" command allows you to count for a specified number of seconds. These commands are useful in setting up your sample.

When you exit from PIKMAN or HUBMAN you have the option of saving all angles in a file named DIFF.;1. If you reject that option, all angles will be driven to zero and DIFF.;1 will be deleted.

There is one important difference between PIKMAN and HUBMAN. In using HUBMAN, if you wish to maintain the Bragg condition when 2θ is changed you must use the "B" command rather than the "T" command. "B" drives 2θ and θ at speeds which approximate a 2:1 ratio and the total change in θ will be exactly half the change in 2θ .

Running MOTOR

Log on to the VAX *via* account PICKER. The assignment of the VAX port TXA2: which connects the FDD to the VAX is done automatically. RUN MOTOR to display the menu. "S" will turn off the Newport motor on the FDD. "F" and "R" will drive the motor continuously forward or reverse until "S" is given or the forward or reverse limit is encountered. "Fx" or "Rx" will drive the motor forward or reverse by x steps where x is any positive integer. "C" causes the status of the FDD interface to be printed. It should indicate normal, forward limit or reverse limit. "G" gives the strain gauge reading in μ volts.

MOTOR is useful for manipulating the FDD tension arm and for verifying that all the FDD functions are working normally. The input and output shown on the monitor screen are recorded in the file MOTOR.DAT.

4. DATA COLLECTION with PICKER or HUBER

General Comments on PICKER and HUBER

Enter these programs by typing RUN PICKER or RUN HUBER after logging into the appropriate VAX account. The first input requested will be the choice of diffractometer angle values: a) values stored in the file DIFF., b) all angles zero, or c) initialize to the values you enter. PICKER and HUBER store the current values of the angles when you type the EX command to exit, so DIFF.;1 should contain the correct angles if the previous run has been with PICKER, PIKMAN, HUBER or HUBMAN. DIFF.;1 is updated only for a normal program exit. If the run was aborted because of an abnormal condition, the angles in DIFF. will, in general, be incorrect. The program will respond with a menu which includes several utility functions in addition to the main data collection items. When you choose an item, the program will respond by requesting all the information needed to make the 2θ , ω , χ or layer line scan you need.

Table 1. *SCAN.IN* file for PICKER

Specific input requests for each of the menu items are discussed below. All of the data collection options offer to initialize the output file if you request it. If this option is chosen, the diffractometer setup information and experiment title stored in SCAN.IN will be displayed. You may choose to accept this as is; if you don't, each item will be presented individually to be accepted or changed. Table 1 shows an example of a SCAN.IN file.

```
PGL POLYESTER
SAMPLE FROM SLUTZKER
  1.0   1.5
NI  0.0010
  3.0   6.0
  45   70
  0.000
```

This file contains the experiment title, sample ID, collimator diameters in mm, filter material and thickness in inches, SVA opening, KV and ma settings, and, on the last line, the angle between the fiber sample axis and the true χ zero.

A title change is requested even if the output file has just been initialized. The title will be written into the output file as the first record and the first 20 characters of the title line will be filled with the name of the program used to generate the output.

While the program runs, the display on the terminal screen will tell you what is happening. The subroutines that control the scanning, show the accumulated count and the accumulated counting time as each point in the scan is measured. The display is updated every 10 s as the count is accumulating. To facilitate multiple data collection runs, it is possible to "stack" up to ten 2θ or χ scans to be measured sequentially without operator intervention. If the FDD is being used, the scans can be made with the sample under tension.

Preparation of Fiber Samples

Multifiber samples will give diffraction patterns comparable to the pattern for a single fiber only if all the fibers in the sample are parallel. This condition is difficult to achieve but it can be approximated very well with fiber samples prepared for use with the FDD as discussed in another report (Lenhert and Adams, 1990). Since preparation of FDD

samples is tedious we describe another method which will give acceptable results in most experiments.

The fiber yarn is threaded into a glass capillary of the type used for powder samples. Match the yarn size to the capillary so that you have a snug fit. The capillary with the yarn inside is mounted on a metal frame made from a medium size paper clip. The paper clip is bent to the shape shown in Fig. 1. The lower end can be held in a regular goniometer head and the capillary cemented to it as shown. Both ends of the capillary are broken and the fiber yarn pulled partly through. The lower end of the yarn is cemented to the holder. When the cement is set, put cement on the opposite end of the fiber yarn and hold it under tension until that cement is also set. Five minute epoxy works well for these steps. After the holder is in the goniometer head and mounted on the diffractometer, the final step is to carefully break the glass at the point where the X-ray beam will strike the sample. The slight tension and the snug fit in the glass should hold the yarn fibers very nearly parallel.

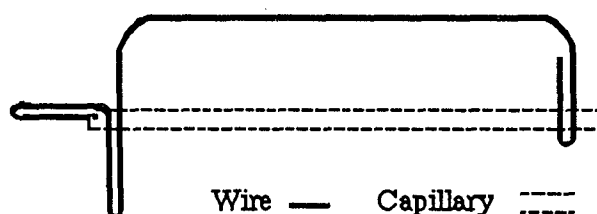


Figure 1. Diagram showing a simple sample support.

Samples prepared in this way are suitable for 2θ and layer line scans. If fiber orientation is to be studied with χ scans, it may be necessary to use the FDD and apply slight tension to ensure that the fibers are parallel. In such cases, the method of mounting the fibers in a capillary is not acceptable.

Note also that no matter what method you use, the diffraction pattern of a twisted yarn will not be the same as the pattern from an untwisted yarn with parallel fibers.

The Menu

- IC -- *Insert Comment*. This command is used to insert comments into the output file. The comment is entered and stored until the next data collection procedure is started, then it is written into the output file.
- DF -- *"Manual" Mode for the Diffractometer*. This command allows the experimenter to manipulate the diffractometer using a subroutine which is a version of PIKMAN.
- MD -- *"Manual" Mode for the FDD.** This command allows the experimenter to drive the micrometer motor, check the motor status and read the strain gauge bridge output. It is useful for checking the *Crystal Logic* FDD interface function and the motor and strain gauge operation. The subroutine used is a version of MOTOR.
- TH -- *Two Theta scan*. This command initiates a 2θ step scan. After the output file initialization procedure, the input requested is: a) counting time in seconds for each step in the scan, b) ω , ϕ , χ angles to be used for the 2θ scan, and c) 2θ values and 2θ step

* Not currently available in HUBER

size. The program then displays the time required to make the scan with the parameters selected. The time of day for the run to finish is also shown. If all is satisfactory, parameters for another scan can be entered, if not, the parameters just entered can be replaced with new values. Up to ten 2θ scans can be entered, *i.e.*, 2θ scans at ten different ω , ϕ , χ settings. There is no tension setting option. Output is written to TTSDAT.TMP

The step size and counting time used for the 2θ scans will depend on the count rate and the accuracy needed. If the peaks are to be fit analytically using FIT (see below), the 2θ step size for polymer fibers will normally be 0.1° or 0.2° in regions where peaks are expected. Larger steps may be used for background regions. Counting times of 200 s per step are advisable if weak peaks contain significant information. SVA settings of 2x6 mm or 3x6 mm will give satisfactory resolution unless the peaks are very sharp. The Ni filter should be used with the 1.0 mm incident beam and the 1.5 mm diffracted beam collimators unless fiber samples are significantly smaller. Be sure that the fiber axis is aligned with the ϕ axis before beginning data collection. (See CZ command below for checking sample alignment.) In order to allow a subsequent check for the zero point in 2θ it is advisable to scan at least one strong peak at negative 2θ .

Note that if you are measuring or plan to measure an air scan to make an air scatter correction, you must be sure to use identical collimators, filter and KV settings as differences between the air scan and the data scan cannot be easily scaled.

- OM -- *Omega scan*.* This command initiates an ω scan which is then fit with a Gaussian to determine the center of the ω peak. After the output file initialization procedure the input is entered as: a) counting time in seconds for each step, b) 2θ , ϕ , χ , c) ω values and ω step size and d) the background option. The background option choices are: measure the background at the specified ω settings or enter known background count rates at specified ω settings. A tension setting can be specified if the FDD is in use and up to ten scans can be entered before the run is started. When each scan is completed, the linearized least-squares procedure is used to fit Gaussian parameters to the scan just measured. Output is written to OMSDAT.TMP.
- CZ -- *Chi Zero scan*. This command is used to make a χ scan and fit the result with a Gaussian in order to determine the center of the χ scan. This is useful in determining the alignment of the fiber sample. The true χ zero value, thus determined, is written into SCAN.IN to be used with the *Chi Scan* function, CH. Input requested is the same as the *Omega Scan* except that 2θ , ω and ϕ are entered as fixed angles and χ is stepped. The same background options apply as in the *Omega Scan* except that χ is specified instead of ω . No tension setting option is available and only one scan may be entered at a time. Output is written to CHSDAT.TMP.
- CH -- *Chi Scan*. This command initiates a χ scan. After the output file initialization procedure the input is entered as: a) counting time in seconds for each step, b) 2θ , ω , ϕ and c) χ values and χ step size. There are several modes available for the χ scan and the next input depends on which of the three cases listed below that you want to use.
 - Case I. Simple χ scan: Input only counting time, 2θ , ω , ϕ and then χ with χ step size (negative limits allowed).
 - Case II. Integrated intensity χ scan. In addition to the data for Case I, the 2θ points for measuring the X-ray background must be supplied. These are each counted for a total time equal to 1/2 the peak counting time and the total is subtracted from the peak count to approximate the integrated intensity.

Case III. χ scan with orientation factor calculation: Same input as Case II but, Herman's orientation factor is calculated from the integrated intensities for each χ scan. These scans must start at $\chi = 0$, and the orientation factor calculation assumes that all points between the final χ measured and $\chi = 90^\circ$ have zero intensity.

When the input for the selected case has been entered you will be presented with the estimated time of the scan and the time of day for it to finish. If all is not satisfactory, new parameters can be entered, otherwise you will be queried about the use of tension and if you choose to use it, the value will be requested. More scans can then be entered if you wish. Note that the χ values to be used in the scan will be relative to the true χ zero determined with the CZ function for this sample or the value entered when the output file is initiated. Output is written to CHSDAT.TMP.

Measurement conditions for a χ scan should normally be the same as those for a 2θ scan of the same material except that the SVA should be set to a square opening. If a 3x6 SVA setting is used for 2θ scans, the χ scan should be made with a 3x3 SVA. Use the drill bits to set the opening. Be careful when you set the χ limits and choose the points for measuring the background. Diffraction in adjacent layer lines may cause problems if the scan or the background limits overlap any of the adjacent layer lines. It is very useful to have a flat film WAXS photograph of the material being studied to visualize the diffraction pattern when you set up the χ scan. A preliminary scan with shortened counting times is also useful to check your choice of scan parameters.

If you plan to correct the scan for instrumental broadening, be sure to make the χ scan with fine enough steps. For reasonably sharp peaks (FWHM of 3 to 6°) the χ steps should be 0.1° from the center to the point where the scan begins to level off. For example, a peak with FWHM = 5° was satisfactorily measured with χ steps of 0.1° from $\chi = 0$ to 4°, 0.2° from $\chi = 4$ to 10° and 0.5° from $\chi = 10$ to 13°. The scan was terminated at 13° to avoid including intensity from the first layer line which gave significant diffraction at the 2θ of the peak and the background being measured.

An instrumental broadening correction can be made with the programs FITSM and UNFOLD discussed below. It will require as input the χ scan for the fiber sample and a similar χ scan for a standard material. The standard must have a strong reflection at the same 2θ as the peak of interest from the fiber sample. The standard χ scan must have a width which is due only to instrument factors and the scan must be made with the same instrumental settings, collimation, SVA, *etc.*, as the fiber sample χ scan. Details are described in the following section on the UNFOLD program.

The scan data section of the output file, CHSDAT.TMP, will contain five columns of numbers if case II or case III are used. The first column is χ , the second is the intensity (CPS) corrected for background, the third is the raw intensity, the fourth and fifth are the two background measurements in CPS, all measured, for each step of the scan. Herman's orientation factor, if calculated, is also in CHSDAT.TMP. The orientation factor is given by

$$H = \frac{\sum_i I_i (3 \sin^2 \chi_i - 1) \cos \chi_i}{2 \sum_i I_i \cos \chi_i}$$

where I_i is the measured intensity at χ_i .

- **LL -- Layer Line scan.** This command is used to measure the intensity along the layer lines of the fiber diffraction pattern. After the usual file initialization procedure, the input is entered as: a) layer line spacing in Å, b) counting time for each step in seconds, c) a 2θ limit on the data to be measured if you want 2θ rather than the layer line index and the running index to limit the data, d) layer line index values to use: start, increment and stop, and e) running index along the layer line: start, increment and stop. The estimated run time and the time of day for the run to finish will then be presented. If all input is ok, the tension will be requested and it can be entered if the FDD is in use. Additional scans cannot be entered. An accurately measured complete layer line scan can take one or more days. Output is written to LLSDAT.TMP.

Normally one would measure a quadrant corresponding to positive indices with scans along and between the layer lines, *i.e.*, at $l = 0.0, 0.5, 1.0, 1.5$ *etc.* The optimum k spacing will depend on the sharpness of the peaks. Use a k increment of 0.05 for a good quality scan. (Note; l is the layer line index, k is the running index along the layer line.) A 3x3 mm SVA is to be preferred but if the layer line and peaks are broad, you may be able to use a larger opening. The Ni filter should be used with the 1.0 mm incident beam collimator and it may be advisable to use the 1.0 mm diffracted beam collimator to reduce background. Good quality scans require fairly long counting times, say 100 to 200 s per point. Layer line data taken as suggested may be processed with LLPLOT.

An air scatter correction is advisable if intensities at low 2θ values are to be reliable. Make the scan with the TH option of PICKER. Be sure not to move the X-ray beam stop. (Low angle air scatter intensities are very sensitive to the beam stop position.) All settings, collimation, filters, *etc.* must be identical to those used for the layer line scan but don't forget to remove the sample! The 2θ intervals should be small (0.1°) at low angles and may gradually increase to 1.0° at 15° and 2.0° above 45° .

When the layer line scan and the air scatter scan are finished, copy them together in the same file for safe keeping. Use a descriptive name for the file. You are now ready to prepare input files for analysis and plotting with the LLPLOT program.

- **SS -- Standard scan.** This command is intended to make 2θ scans of a standard powder sample to determine the instrumental broadening so that crystallite size can be determined for fiber samples. It has not been fully implemented.
- **EX -- Exit.** This command causes an exit from PICKER after saving the current values of the diffractometer angles in DIFF;1.

When the scan is finished, the file name of the data file, either TTSDAT.TMP (for a 2θ scan), OMSDAT.TMP (for an ω scan), CHSDAT.TMP (for a χ scan) or LLSDAT.TMP (for a layer line scan) is displayed along with the final values of the diffractometer angles. Note also that DIFF;1 is updated after each scan is finished. Table 2 shows a portion of a TTSDAT.TMP file which is similar to the other output files.

These output files can be processed for analysis, plots, *etc.* The plotter program, PLOT1, can be used to plot 2θ , ω and χ scans. LLPLOT can be used to make an air scatter correction and plot the layer line scan data. FIT can be used to carry out data reduction on 2θ scans and fit the peaks using a non-linear least-squares procedure. An air scatter correction can be made with LLPLOT and FIT if you have an air scatter data file. The FIT, LLPLOT and PLOT1 programs are described below.

Table 2. TTSDAT.TMP file showing the diffractometer set up and sample information generated by the initialization request followed by data collected, in this case, 2 θ settings and counts per second.

2 THETA SCAN PGL POLYESTER

SAMPLE FROM SLUTZKER

Incident beam collimator diameter is: 1.0mm.

The diffracted beam collimator diameter is: 1.5mm

The diffracted beam aperture is 3.0mm wide and 6.0mm high.

The filter is NI . It is 0.0010 in. thick.

X-ray generator is set at 45 Kv 70 ma.

Correct sample orientation is at 0.000 deg.in CHI

File initialized: DATE IS 22-MAR-90 AT 16:11:05

EXPECTED TIME FOR SCAN IS 172. min:

FINISH at 19: 3, 0 Days hence.

2 THETA SCAN PGL POLYESTER

Each point in the scan is counted for 100 sec.

For each 2 Theta, OMEGA = 0.00, PHI= 0.00, CHI= 91.00

Scan started: DATE IS 22-MAR-90 AT 16:17:00

35.00	31.500
35.20	31.860
35.40	33.340
35.60	32.390



54.60	27.780
54.80	28.100
55.00	27.530

Scan ended: DATE IS 22-MAR-90 AT 19:10:26

ANGLES SAVED IN FILE DIFF. ARE:

TI = 55.00 OM = 0.00 PH = 0.00 CH = 91.00

5. THE X-RAY MODULUS PROGRAM

The X-ray Modulus Program, XRYMOD, facilitates all operations related to X-ray tensile modulus measurements. It is the software interface between the automated diffractometer, FDD and the experimenter. It requests the information needed and issues instructions to the experimenter *via* the terminal screen for calibration of the FDD and for mounting the sample. It can carry out the measurements, adjust the tension as requested by the experimenter and calculate the X-ray modulus based on the tension and the 2θ values observed. A plot file of d spacing vs tension which is compatible with a Calcomp plotter, is also generated. A brief description of the workings and features of XRYMOD is given below. Detailed information on the FDD and on XRYMOD is in another report (Lenhert and Adams, 1990).

Information about the material being studied and the particular sample currently mounted is entered from the terminal and stored in the VAX file, MOD.IN. The experimenter responds to requests from the computer for the required information, such as: title of the experiment, identifying information about the substance and sample history, number of fibers in the sample, fiber density, denier per fiber, expected modulus (used to predict the approximate position of diffraction peaks as a function of strain), 2θ at zero tension, step scan parameters, counting time, X-ray tube settings, *etc.* The MOD.IN file is read by XRYMOD any time data on the sample is needed. Any data stored in MOD.IN can be modified by the IO function. The calibration function, CS, stores the FDD calibration in MOD.IN.

The experiment log file, XRYMOD.TMP, is initiated by the IO function. This command causes sample data from MOD.IN to be entered into XRYMOD.TMP. The log file serves as a record of the experiment. It is updated at each measurement to record all tension and scan data as well as the modulus calculation. All measurements performed with XRYMOD will be recorded in the current log file until a new file is initiated.

The calibration procedure determines the relation between the strain gauge output and the tension applied by the FDD. The tension is proportional to the strain gauge bridge output which is available as a digital display. The bridge output is read by the VAX computer through a subroutine, which returns the value displayed (in mv). Lead weights of known mass are hung from the sample holder to determine the relation between the strain and the bridge output.

The menu of XRYMOD is discussed briefly below. XRYMOD carries out calibration and modulus measurements with the FDD. It guides the experimenter through the steps which result in the desired calibration and tension measurements.

- EP -- *Enter sample or scan Parameters.* This command is used to create or update an existing MOD.IN sample data file which includes scan parameters for the peaks to be measured as well as the tension values.
- IO -- *Initialize Output.* The IO command opens a new XRYMOD.TMP file, reads and displays the MOD.IN file containing sample and experiment data and allows changes to MOD.IN if any are requested. The X-ray generator Kv and ma, filter, SVA and collimator sizes are also written in XRYMOD.TMP along with the current date read from the VAX clock.

- **AL -- *Make FDD Alignment Adjustments.*** This command drives the FDD tension arm to the alignment position and provides a prompt to start the alignment procedure.
- **CS -- *Calibrate Strain gauge.*** This command initiates the strain gauge calibration. Prompts request operator intervention in the calibration procedure when they are required. A linear least-squares fit of force vs strain gauge output determines the calibration. The calibration, slope and date are recorded in XRYMOD.TMP.
- **DF -- *"Manual" Mode for the Diffractometer.*** This command allows the experimenter to manipulate the diffractometer using a subroutine which is a version of PIKMAN.
- **MD -- *"Manual" Mode for the FDD.*** This command allows the experimenter to drive the micrometer motor, check the motor status and read the strain gauge bridge output. It is useful for checking the *Crystal Logic* FDD interface function and the motor and strain gauge operation. The subroutine used is a version of MOTOR.
- **MS -- *Mount Sample.*** This command lowers the tension arm to allow convenient mounting of the sample. Then, with the sample in place the tension arm is positioned for tension measurements. The experimenter receives reminders as to what adjustments must be made to mount the sample.
- **MR -- *Modulus Run.*** This command initiates a modulus measurement. The diffractometer angles are initialized, data from MOD.IN is read and XRYMOD.TMP is opened. A list of run specific input is requested before tension adjustments and scanning are started. The first peak is scanned by the 2θ step scan subroutine, the step scan data are fit to the intensity and the d -spacing corresponding to the center of the peak is determined. The tension is adjusted to the next value in the list, the peak is rescanned and fit until all tension settings have been finished. The modulus and its uncertainty are calculated and printed along with all observed points and their residuals. A plot of tension vs d -spacing is stored in PLOT.PLT. When the file is plotted, the plot shows the experimental points, the line and the modulus calculated by least-squares. The run title, date and data on the sample are also shown.

6. DATA ANALYSIS PROGRAMS

Utility Plotting Program, PLOT1

This program will plot any file of x,y data. The first line of the file must be a title, all subsequent lines must start with the x variable followed by the y variable. Additional numbers on the line will be ignored.

The program requests the name of the file containing the data. If the file contains more than 1500 data points, the "use every 5th point" option must be chosen. When the data file is read, the program reports the number of points, minimum and maximum x and it requests the first and last point to be used for the x -axis. The axes used can be greater than the range of the data set, or less. The choice depends on the plot you want. The maximum and minimum values for y are reported and again your choice for the y -axis limits depend on the plot you want. If you want to expand the scale to show small peaks, you may truncate the large peaks by setting a lower value for the y -axis limit without causing problems with the plot. Next enter the appropriate axis labels as requested.

The number of places to be printed after the decimal point is requested for both axes. If you want the axis values to correspond to those in the data file, *i.e.*, not to be scaled by a power of 10, enter the number of places as a negative number. If you want the axis values to be whole numbers with no power of 10, enter -99. The size of the plot is controlled by the values you enter for the length of the axes. At the "END OF FILE" message, your plot is in PLOT.PLT ready to be copied to TXC7:.

FORTTRAN files required for this program, in addition to PLOT1 are CCM88, the Calcomp subroutine package which drives the plotter on the TXC7: port.

20 Scans - Data Reduction and Curve Fitting, FIT

Data collected with the TH option of PICKER or HUBER can be analyzed by FIT. Details of the measurement procedure are discussed above. The TTSDAT.TMP output file is modified to become the input file for FIT. The 20 scan should have the title, run information, date and time written by PICKER condensed to two lines (other heading information should then be deleted). No blank lines are allowed and the file ends with the last 20 and count. The input file with the air scatter data, also a modified TTSDAT.TMP file, should have a one line title and the measurement conditions should be summarized on that line. Except for the title, the format of the air scatter file is identical to the 20 scan file for the fiber sample. Since a linear interpolation is used to find values corresponding to the 20 scan for the fiber sample, one need not repeat an air scan to obtain data points that coincide with the data points of the sample scan.

The options available in the FIT program are presented as questions when the program is run. If an Air Scatter correction is elected, the SVA area and the X-ray tube current used for the data scan and for the air scan must both be entered. The air scatter intensities are scaled to match the conditions for the data file before they are subtracted. The air scatter correction is made before any other corrections are applied. Air scatter is not significant at high angles but can be substantial at low angles and at very low angles, the beam stop position is critical.

The polarization correction depends only on 2θ and can be applied to any 2θ scan. The absorption correction requires sample density, size and chemical composition. The volume of sample material exposed to the X-ray beam is a function of 2θ for meridional scans but it is essentially constant for a cylindrical fiber sample if equatorial scans are made. If the absorption correction is requested for a meridional scan, both absorption and variation in irradiated volume will be included in the correction factor. The Compton scattering correction requires the sample composition and one measurement of sample scattering at a point where there is no coherent (Bragg) scattering. For most samples you should include an intensity measurement at $100^\circ 2\theta$ if possible. The Lorentz correction, as presently coded, may contain an error. Do not use it. When the options have been selected, the data reduction phase of the calculation is carried out.

You may now respond that you wish to fit the data. If you are using the program to carry out data reduction only, respond with "N" and your corrected data will be in a file named FITTST.DAT. Otherwise, the fitting portion of the program is entered and input is requested. It is best to fit resolved peaks separately but unresolved peaks must be fit in groups. Select the 2θ range so that several points of background intensity are included on each side of the peak(s). Only the measurements between the limits you enter will be included in the present calculation. Enter the number of peaks and the number of background points to be used for this fit. Experience shows that two background points at the limits of the data interval should normally be used if the background intensities are to be refined. All peak and background information is entered as requested by the program prompts.

If you are working on a resolved peak of reasonable size, all variables, including the background intensities, can be refined on the first pass. Remember that this is a non-linear least-squares problem so the calculation may not converge if your initial parameter values are too far off. Satisfactory starting values can be estimated from a printout of the data file or from a preliminary plot. If you are working on a group of unresolved peaks, it is usually best to start by refining only the most prominent features and adding the others as your description gets better. The output from an example calculation with unresolved peaks, as written into FOR007.DAT is shown in Table 3. Note that the position, height and width of the center (unresolved) peak in this example are not refined on the first attempt. After the second set of iterations has converged, the result is plotted. All parameters are written in the summary which shows that small shifts in some parameters were made on the last cycle. Each least-squares fit is completed before the 2θ range and peak parameters are entered to fit the next segment of data. If you are not satisfied with the current fit, ask to "refit this data".

The final prompt from the program allows you to fit another 2θ range (or change the present 2θ range). Each set of parameters generates its own output to FOR007.DAT and its own plot, if requested. Each plot and the corresponding printed output shows the time the plot was created. That way you don't get the plots mixed up. The last page of the printed output is a summary of the refinement which lists the final values of all parameters. This listing is shown at the bottom of Table 3 for the three peak example. A record of the progress of the refinement for each set of parameters is in FOR008.DAT. If the plot option is selected, the hard copy plot can be obtained by the command, COPY PLOT.PLT

TXC7: Fig. 2 shows the plot corresponding to the output in Table 3.

FORTTRAN files required, in addition to FIT, are: FIT1, FIT2, FIT3 and CCM88.

Table 3. Selected portions of the output file, FOR007.DAT of FIT, for a two step fit of the parameters for three overlapping peaks. The fiber sample is UV irradiated silk.

THE AREAS UNDER THE VARIOUS PEAKS ARE GIVEN BELOW:

AREAS = 2.08769 0.89490 1.39713
 INTEGRAL BREADTH = 0.02135 0.02983 0.04953

BASELINE AREA = 10.83509

FINAL FITTING PARAMETERS USED

PEAK POS	INT AT MAX	FWHM	GAUSS (PEAK POS & WIDTH IN deg)
1 0.44136	1 97.775	1 0.01620	0 0.50
39.784		1.432	
0 0.45322	0 30.000	0 0.02264	0 0.50
40.900		2.000	
1 0.48224	1 28.210	1 0.03759	0 0.50
43.649		3.321	

BASELINE POSITIONS AND INTENSITIES

0.4116 37.00 1 95.594

0.5379 49.00 1 75.941

ITERATION NUMBER 5, SE = 0.109986E-07

THE AREAS UNDER THE VARIOUS PEAKS ARE GIVEN BELOW:

AREAS = 2.24441 0.90063 1.47027
 INTEGRAL BREADTH = 0.02269 0.02809 0.04760

BASELINE AREA = 10.67530

FINAL FITTING PARAMETERS USED

PEAK POS	INT AT MAX	FWHM	GAUSS (PEAK POS & WIDTH IN deg)
1 0.44125	1 98.938	1 0.01605	1 0.31
39.773		1.418	
1 0.45363	1 32.062	1 0.02369	1 0.76
40.939		2.093	
1 0.48404	1 30.887	1 0.03105	1 0.07
43.820		2.743	

BASELINE POSITIONS AND INTENSITIES

0.4116 37.00 1 93.611

0.5379 49.00 1 75.394

ITERATION NUMBER 11, SE = 0.589611E-08

TWO THETA	S(1/A)	++++ OBSERVED	++++ CALCULATED	++++ OBS-CALC	BASELINE	INDIVIDUAL PEAKS PK 1	PK 2	PK 3
37.00	0.4116	98.820	100.090	-1.270	93.611	4.655	0.568	1.255
37.10	0.4127	101.085	100.334	0.752	93.456	4.988	0.598	1.292
37.20	0.4137	98.968	100.620	-1.651	93.301	5.358	0.631	1.330
37.30	0.4148	101.630	100.955	0.676	93.147	5.771	0.668	1.369
39.60	0.4394	201.481	199.246	2.234	89.602	94.433	12.119	3.093
39.70	0.4405	202.322	204.590	-2.268	89.448	98.109	13.802	3.230
39.80	0.4415	206.007	207.091	-1.084	89.295	98.822	15.596	3.377
39.90	0.4426	206.942	206.610	0.333	89.141	96.454	17.480	3.535
40.00	0.4437	202.845	203.511	-0.665	88.988	91.397	19.423	3.703

49.00 0.5379 77.425 78.201 -0.775 75.394 0.466 0.148 2.192

PLOT MADE DATE IS 31-JAN-90 AT 13:00:43

SUMMARY

*** MARY B UV S3 SILK COLL. 1.0-1.5 SVA 3X6 NI 45KV 70ma 200 s

*** MER SCAN 4-JAN-90 06:09:42

FINAL PARAMETERS FOR EACH OF THE PEAKS FITTED. FIRST ROW IS PARAMETERS,
 SECOND ROW OF EACH PAIR IS FINAL CYCLE NUMBER AND THE PARAM SHIFT ON THAT CYCLE.
 PEAK AREA AND INTEGRAL BREADTH ARE ALSO SHOWN.

PK	2 TH	d-SP	S(1/A)	MAX INT	FWHM	FWHM(2TH)	XGAUS	PK AREA	INT WID
PK 1	39.773	2.2663	0.44125	98.94	0.01605	1.418	31.2	2.24441	0.02269
CY 11	0.000			-0.10		0.000	-0.2		
PK 2	40.939	2.2044	0.45363	32.06	0.02369	2.093	76.1	0.90063	0.02809
CY 11	-0.001			0.02		0.009	-0.9		
PK 3	43.820	2.0659	0.48404	30.89	0.03105	2.743	7.4	1.47027	0.04760
CY 11	0.003			0.04		-0.002	-1.6		

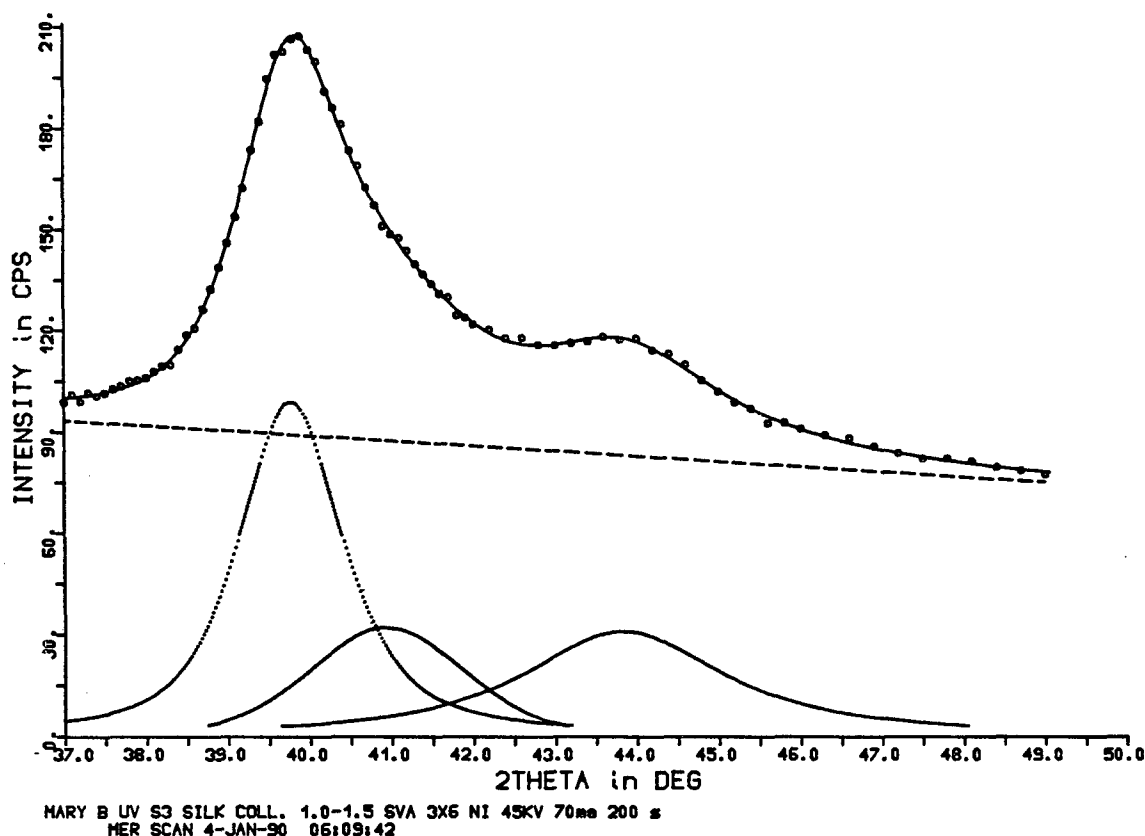


Figure 2. Plot generated by FIT. Experimental points are shown as small circles and the composite calculated curve as a solid line (top curve). The background is a dashed line and the individual calculated peaks are the lower dotted lines. The title information for the data file is below the 2θ axis and the time the plot file was written is in the top right corner.

FIT was used to analyze data from fiber samples of PBO and PBZT to obtain the d -spacings and intensities reported by Fratini, Resch, Lenhert and Adams (1989).

Layer Line Scans and Plots, LLPLOT

Data collection with the LL option of PICKER can be analyzed by LLPLOT. Details of the measurement procedure are discussed above. The file with the layer line scan thus measured, LLSDAT.TMP, should be modified as follows. The title, run information, date and time written by PICKER must be condensed to two lines (other heading information should then be deleted). The numerical data headings for each set of layer line counts are retained. No blank lines are allowed and the file ends with the last layer line count. Give the file a convenient, easy-to-remember name.

The required format of the Air Scatter file is also similar to the PICKER output in TTSDAT.TMP. Title, run information, and data must be condensed to one line, the lines with 2θ and intensity values must be retained and any non numerical lines at the end of the file must be deleted. Give the file a convenient, easy-to-remember name.

Data reduction and layer line plotting are carried out by running LLPLOT. Input prompts by the program are self explanatory for the most part.

If the Air Scatter correction option is selected, a least-squares polynomial fit of the air scatter data is used to interpolate air scatter to 0.01° intervals in 2θ . The interpolation is controlled by subroutine PINTP and the least-squares fit is carried out by subroutine POLFIT. A standard option of a 3rd degree polynomial fit to 20 data points is offered. If this option is rejected the user must specify the polynomial degree and the 2θ range for the fit. In either case, the results of the fit are displayed and the user either accepts or rejects it.

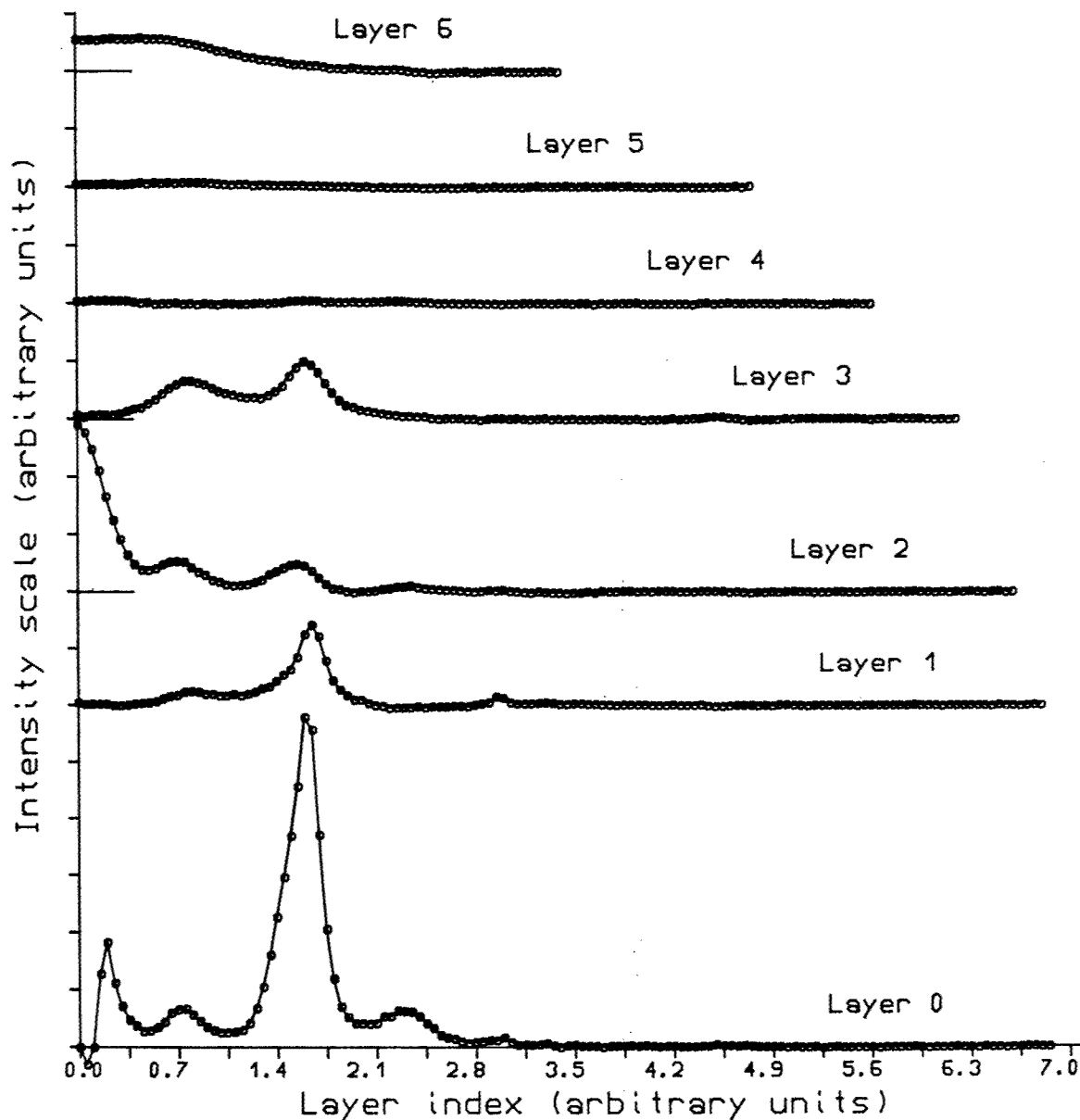


Figure 3. Layer line scan of degummed silk (*Bombyx mori*) showing layers 0 through 6.

If the fit is rejected, new values for degree and 2θ range are requested and another fit is calculated. If the fit is accepted, interpolated values of the air scatter for 2θ in steps of 0.01°

are stored. The next segment of the measured air scatter step scan is then considered. The new range will overlap the previous range in 15 of the 20 points if the standard option is used, otherwise the overlap is specified by the user.

It is good practice to overlap the segments, as the standard option does, so that three or more interpolated values are calculated for each point. When the point to point change in air scatter becomes small enough so that the standard option gives a good fit, the interpolation process can be speeded up by accepting the range extending option. This is equivalent to accepting the remaining fits in advance. When the end of the 2θ range is reached, all the overlapping values are averaged to give the air scatter curve used for the correction.

Processing of the layer line scan data now begins. The specified file is read, the air scatter is subtracted point by point and then the background scans at $l = 0.5, 1.5$ etc. are subtracted from the layer lines ($l = 0, 1$ etc.). If the layer lines are to be indexed, the a and b unit cell translations are entered. Note that the indexing option provided assumes an orthogonal lattice. If the data are indexed, the layer line coordinates, intensity (less corrections), the d -spacing and the indices are written in the OUT.DAT file. The plot is stored in PLOT.PLT and the command, COPY PLOT.PLT TXC7: will give you a hard copy plot an example of which is shown in Fig. 3.

FORTTRAN files required for this program, in addition to LLPLOT, are: PINTP, POLYF, CCM88, MATRIX, and BBMB.

χ Scans, an Instrumental Broadening Correction

The Programs, FITSM and UNFOLD

The FITSM program takes the raw data from a χ scan, smooths out statistical fluctuations in the count rate and interpolates where measurements were originally made at intervals larger than 0.01° . The smoothing and interpolation is done by a least-squares polynomial fit to segments of the data. The number of data points in the segment and the degree of the polynomial are selected when the program is run. Successive overlapping segments are fit and the resulting calculated values are averaged to give the final smoothed curve with count rates for each 0.01° in χ . Satisfactory agreement between the experimental points and the polynomial functions is obtained in most cases with 20 points and a 3rd degree polynomial. For the tails of the scan, where the count rate varies slowly, polynomials of zero to first degree can be used. Comments on this technique are given in the discussion of Air Scatter curves in the section on LLPLOT.

The polynomial subroutines used in FITSM are subroutines POLFIT and PVALUE both from the SLATEC library.

The UNFOLD program takes input from both a standard scan and an experimental scan. The standard scan represents the instrumental and sample size broadening of the χ scan at the 2θ angle and the instrument settings used. The experimental curve, measured with the fiber sample, has a finite width in χ due to imperfect sample orientation as well as sample size and instrumental factors. The output of UNFOLD is the Fourier coefficients of the χ scan that would be obtained for perfect collimation and an infinitesimal sample with the orientation of the fiber sample.

Both the fiber scan and the standard scan must be symmetrical about $\chi = 0$, have values proportional to counts per second and have the same χ range and constant χ interval for

both scans. These requirements are normally met by proper choice of scan parameters when the χ scan is measured and by subsequent use of FITSM (see above). If the scans have N points, $N/2$ Fourier coefficients are calculated for each. The coefficients of the unfolded curve (the experimental curve with the standard curve deconvoluted from it) are calculated from the following relationships.

$$A = \frac{A_s A_e + B_s B_e}{A_s^2 + B_s^2} ; \quad B = \frac{A_s B_e - A_e B_s}{A_s^2 + B_s^2}$$

Where A and B stand for the real and imaginary parts of the complex Fourier coefficients of the unfolded curve. The subscripts s and e denote Fourier coefficients for the standard and experimental scans, respectively. The coefficients for the corrected curve must be scaled to match the magnitude of the experimental curve and the odd terms have their signs changed before the corrected curve is synthesized.

A problem is observed when the coefficient for the standard is small. As the above relationships show, the coefficient of the unfolded curve then becomes inaccurate and usually too large. This introduces significant ripple, with a frequency corresponding to the term in error, into the resulting function. The Fourier series should be truncated at the point where the terms for the unfolded curve drop to a minimum (usually less than unity). This will give a ripple free unfolded curve. The program displays the coefficients for the standard, the experimental curve and the deconvoluted or unfolded curve so the truncation point can be selected.

The Fourier subroutines used in UNFOLD are subroutines EZFFT1, EZFFTB, EZFFTF, EZFFTI, RFFTB, RFFTB1, RFFTF, RFFTF1, RADB2, RADB3, RADB4, RADB5, RADBG, RADF2, RADF3, RADF4, RADF5, and RADFG, all from the SLATEC library. They are in the file FOUR.FOR.

The UNFOLD program was tested by unfolding known functions. Gaussian curves of various widths were convoluted with the program FOLD, then fed into UNFOLD. The narrower of the two combined peaks was treated as the standard scan and the convoluted peak was the experimental scan. The unfolded peak matched the original peak (before folding). It should be noted that rounding errors in the calculation prevented the use of more than 30 to 40 terms in the Fourier series for the unfolded peak even though we were dealing with calculated curves free of experimental error. Higher order terms were probably inaccurate because the accumulation of rounding errors was significant in the small coefficients in the Fourier series for the standard curve. A double precision calculation might improve the result but this was not tried.

An Example, KEVLAR 149 χ Scans

Some comments on the treatment of an actual data set seem in order as an example. The data set was measured on the KEVLAR 149 equatorial reflection at 20.55°. The sample contained 233 12 μ diameter fibers with a density of 1.47 gm cm⁻¹ and a denier per fiber of 1.44. The diffracted beam aperture was 2x2 mm, collimators were 1.0 incident and 1.5 mm diffracted beam with 0.001 in Ni filtered Cu K α radiation at 45 Kv and 70 ma. Counting time was 200 s per step. In this experimental set up, the effective sample size is determined by the incident beam collimator size for both the fiber sample and the standard. The raw χ scans extended from -13° to +13° with measurement intervals from 0.1° to 0.5°.

FITSM was used to smooth and interpolate the χ scans. When FITSM is run, the output file contains the X-ray intensity in counts per second (CPS) at each value of χ , in 0.01° steps between -13° and $+13^\circ$, the original range of the scans. A utility program, TESTCH, was used to make a least-squares fit of the center portion of the χ scan with a Gaussian function. This determines the center of the distribution and if it is not at $\chi = 0$, it is displaced by $\Delta\chi$ when the utility program SYM is run. SYM averages each pair of intensities at $+\chi_i$ and $-\chi_i$ to give a perfectly symmetrical curve. If $\Delta\chi$ is not zero, the averaged curve does not end at $\chi = \pm 13^\circ$ and it must be fixed with the aid of EDIT, the VAX editor. Curves thus prepared are ready for UNFOLD.

The standard χ scan was made with a Potassium Ammonium Phosphate (KAP) monochromator crystal. It was run at $19.93^\circ 2\theta$ with the same instrumental settings and χ range as the KEVLAR scan. The step size ranged from 0.01° to 0.5° , depending on the change in the count rate. FITSM was used to smooth and interpolate the KAP χ scan. The tails were fit with a straight line and the remainder of the curve was fit in segments with 3rd order polynomials. A slight asymmetry in the tails (high and low χ) was corrected by averaging count rates at equivalent positive and negative χ values to give a continuous curve. The smooth interpolated curve is therefore symmetrical and gives the count rate for every value of χ in the range measured at steps of 0.01° . UNFOLD was then used as described above.

The KEVLAR curve, the KAP standard and the unfolded curve were plotted. Fig. 4 shows the χ scan of the KEVLAR 149 equatorial reflection at $20.55^\circ 2\theta$ as recorded with an applied stress of 1.0 GPa. Fig. 5 displays the KAP scan at $19.93^\circ 2\theta$ which shows the width of a scan with perfect orientation. Fig. 6 displays the unfolded curve which shows

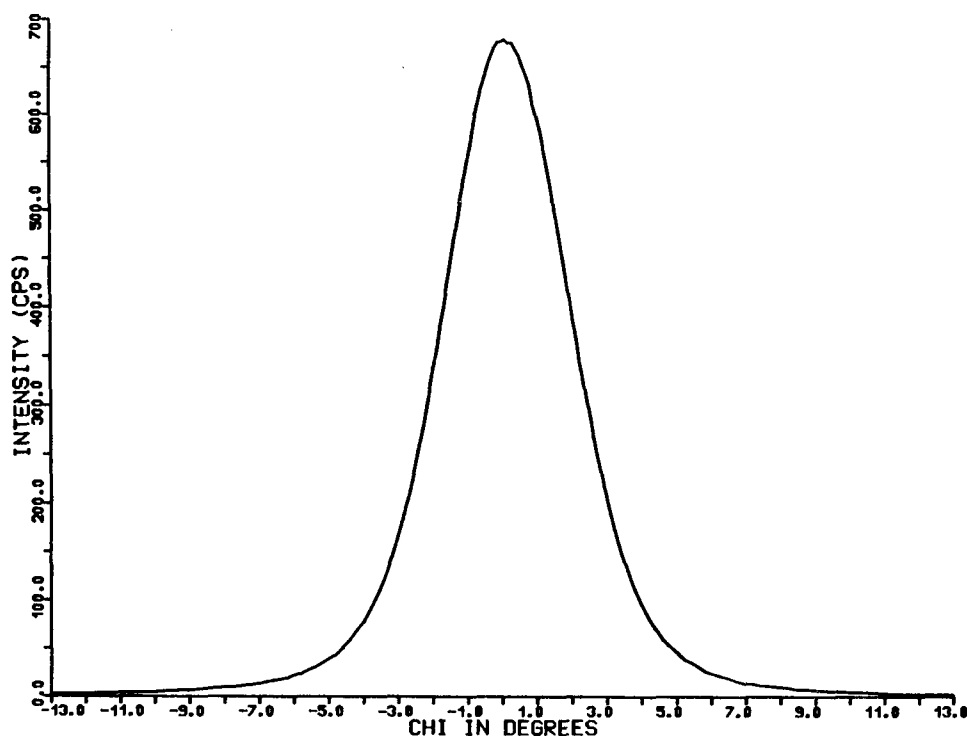


Figure 4. χ scan, KEVLAR 149 equatorial reflection at $20.55^\circ 2\theta$. Tension is 1.0 GPa.

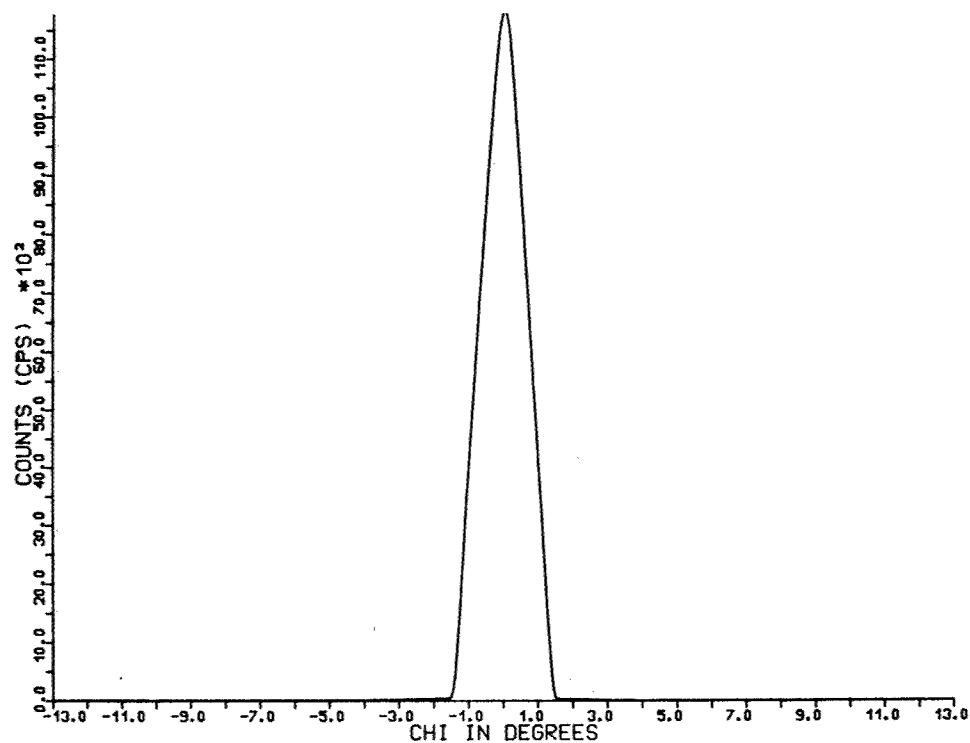


Figure 5. χ scan of the KAP reflection $19.93^\circ 2\theta$.

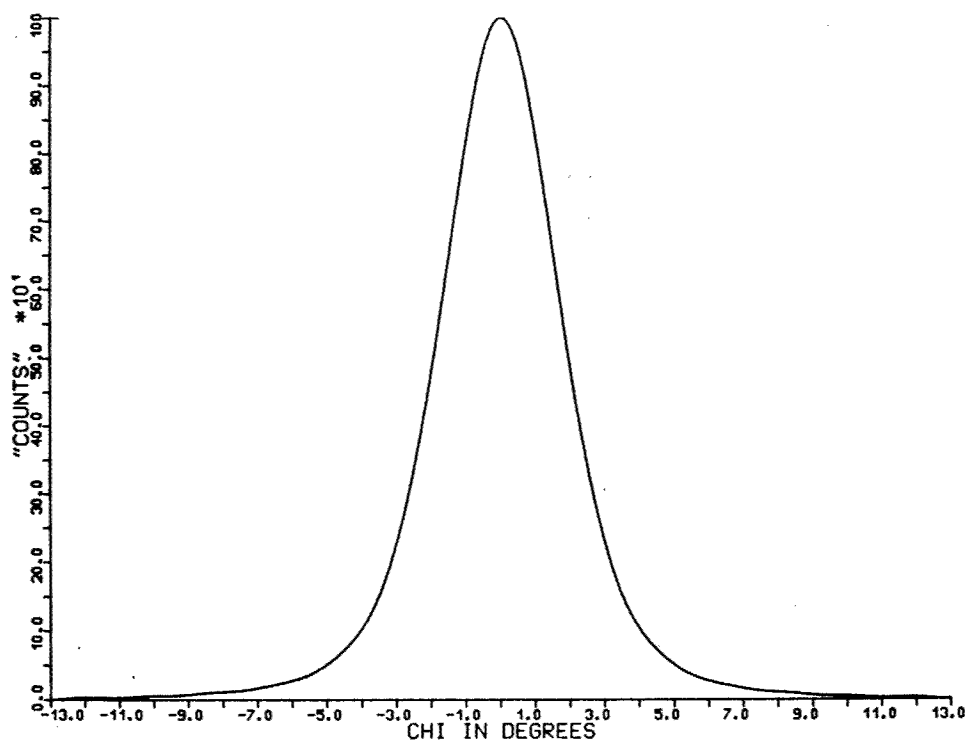


Figure 6. χ intensity distribution, KEVLAR 149 equatorial peak at $20.55^\circ 2\theta$ with instrumental and sample size factors removed. Tension is 1.0 GPa, 14 Fourier terms used for the synthesis.

the intrinsic width of the $20.55^\circ 2\theta$ scan with the instrumental broadening removed by the Fourier method of Stokes.

Finally, XHERM was used to calculate the Herman's orientation factors and χ_{ave} for the experimental curves of KEVLAR 149. Results are shown in Table 4 at three different tensions for the experimental and the corresponding UNFOLDED curves. This illustrates the use of the method in studying orientation as a function of tension. The full width at half maximum (FWHM) was taken by inspection from the data files for both sets of curves.

Table 4. KEVLAR 149 χ SCANS at $20.55^\circ 2\theta$

Stress	FIT-INTERPOLATED			UNFOLDED		
	Orient. Fact.	AVE χ	FWHM	Orient. Fact.	AVE χ	FWHM
0.2 GPa	-0.496696	87.310°	4.61°	-0.497066	87.465°	4.31°
0.5 GPa	-0.496911	87.465°	4.42°	-0.497283	87.561°	4.11°
1.0 GPa	-0.497170	87.511°	4.22°	-0.497545	87.681°	3.89°

7. REFERENCES

- Fratini, Albert V., Lenhert, P. Galen, Resch, Timothy J. and Adams, W. Wade, *Mat. Res. Soc. Symp. Proc.*, **134**, 431-445 (1989).
- Jiang, H., Eby, R. K., Adams, W. W and Lenhert, Galen, *Mat. Res. Soc. Symp. Proc.*, **134**, 341-350 (1989).
- Lenhert, P. Galen, "Diffractometer Alignment, 4-Circle Instruments," ACA Tutorial, University of Oklahoma, *Am. Cryst. Assoc. Tutorial Lecture Notes*, (1978).
- Lenhert, P. Galen, O'Brien, Joseph F., and Adams, W. Wade, "A User's Guide to the Picker Diffractometer for Polymer Morphology Studies," Materials Laboratory, Air Force Wright Aeronautical Laboratories, Final Report 1980-84, AFWAL-TR-86-4024 (1986).
- Lenhert, P. Galen and Adams, W. Wade, *Mat. Res. Soc. Symp. Proc.*, **134**, 329-340 (1989).
- Lenhert, P. G. and Adams, W. W., "Tensile Modulus by X-ray Diffraction: Instrument and Method," Materials Laboratory, Wright Research and Development Center, Final Report 1984-90 WRDC-TR-90-4070 (1990).

8. APPENDICES

A. THE CRYSTAL LOGIC HARDWARE SYSTEM

Picker and Huber Adjustments

The *Crystal Logic* interface connects the diffractometer hardware (motors, shutter, scalar, etc.) to the VAX computer through an RS-232 terminated optical cable. The VAX sees a "terminal" which receives information sent in response to FORTRAN statements of the form WRITE (1,*) 'ALPHA' The "terminal" sends information to the VAX when requested by READ (1,99) LIST.

The diffractometer angle shafts are driven by stepping motors controlled by the *Crystal Logic* interface which provides motion in either direction in 0.01° steps at selected speeds between preset base rates and slew speeds. In order to provide smooth operation, an acceleration parameter controls the starting up and slowing down of each motor.

The slew speed and acceleration register settings are especially important on the Huber instrument since both angles are often driven simultaneously even though the θ and 2θ motions are independent. In order to maintain the Bragg condition approximately, the drive rates must have a 1:2 ratio.

The high speed oscillator frequency for each of the four axes is set by subroutine SLEW. High speed settings are in the SLEW data statement, however, subroutine SETSPEED, which actually sets the registers, doubles the values for each axis (except ϕ) before transmitting them to the interface register. The minimum increment accepted by the interface is 40, i.e., all values specified by SLEW should be multiples of 20 except for ϕ which should be a multiple of 40.

High speed settings for the four angles (in SLEW) are: 2θ , 2400; ω , 2400; ϕ , 2400; χ , 1200. Since the 2θ oscillator is modified to run at 1/4 normal speed, 2400 causes 2θ to run at 3 deg/sec. The ω oscillator runs at 1/8 normal speed so 2400 causes ω to run at 1.5 deg/sec. The ϕ oscillator runs at normal speed but the SLEW setting is not multiplied by two in SETSPEED and, unlike the other axes, the ϕ axis runs at two degrees per motor revolution, therefore; 2400 produces a ϕ speed of 12 deg/sec. The χ oscillator also runs at normal speed so that χ is driven at 6 deg/sec.

The original motor controllers for the Picker and Huber diffractometers reset each angle from one direction to eliminate the effects of backlash. Since backlash does not appear to be a problem with either of the diffractometers, angle resetting has been eliminated. Details of the technical change have been recorded in the *Picker Log Book*.

The stepping motors are mounted on the old Picker motor shafts except for ϕ , which is on the old encoder shaft. The zero positions of three of the four diffractometer axes (2θ , ω , χ) are fixed by goniostat geometry. Zero sensors are installed on the new Picker to allow the computer to determine the zero position for each axis (including ϕ). Each angle requires a coarse (degree) and fine (0.01°) sensor. Each degree sensor consists of a detector (fixed to the diffractometer frame) and a marker (a wire fixed to a diffractometer shaft or circle). The fine sensors are associated with the stepping motor shafts. The marker wire is part of the coupling clamp and the detectors are fastened to the motor mounts. The detectors and markers on the Picker diffractometer were installed in the Materials Laboratory, in

some cases after modifying or redesigning the mounts. The Huber diffractometer was delivered with all sensors and markers installed.

The ϕ coarse detector must ride on the χ circle. In order to allow the required free motion of the connecting cord and also place the detector on the Picker so collision possibilities would be minimized, the detector was mounted on the base of the ϕ axis in the plane of the χ circle. The ϕ base was drilled and tapped (after removing the bronze gear) to receive the detector unit. The wire connecting the detector to the interface was cut and a standard (small) 4-terminal phone cord and plug was inserted. This allows the ϕ sensor to be disconnected after the zero position of ϕ has been determined so there is no chance the cord will become tangled during a data run. The ϕ coarse marker was attached using a preexisting screw hole but in order to accommodate the marker, the goniometer base plate had to be modified.

Finally, adjustments were made to the software parameters of the zero sensing subroutines (ZERO) in PIHARD and HSHARD to match them to the respective diffractometers. When the diffractometer is aligned, true zero for the angles is set by driving the shafts in the negative direction until the coarse and fine sensors are both detected. The direction is reversed to correct the overshoot and the angle is driven slowly back to the fine sensor. Finally true zero is set as specified in subroutine ZERO. This is much easier than adjusting the position of the fine sensors to make them correspond exactly to true zero. If adjustments are required when the instruments are aligned, refer to subroutine ZERO. Find the comments in the program which explain the changes required.

The collision protection circuitry on the old Picker is no longer functional. It is replaced (partially) by a system of limit switches which suspend motor motion if they are activated. The Huber has a similar system of limit switches. If a limit switch is activated, the interface must be powered down and the switch released manually before the diffractometer can be driven again by computer control.

FDD Adjustments and Hardware

The FDD interface was also supplied by *Crystal Logic*. The device itself was designed and constructed at the Materials Laboratory. Details are given in a separate report (Lenhert and Adams, 1990).

B. COMPUTER PROGRAM ORGANIZATION

Hardware driver subroutines written in FORTRAN were supplied by *Crystal Logic* for both the Picker and Huber systems. In order to facilitate writing applications programs for the diffractometer, the subroutines were studied and a summary statement written describing the function of each of the main subroutines. The subroutines were then divided into groups and each group was stored in a separate file. The **hardware** routines issue commands to the interface and receive data such as counts, time and other signals. The **utility** routines prepare information for and call the hardware routines. Subroutines for control of the FDD were written in the Materials Laboratory. They have also been divided into hardware (MHARD) and utility (MUTIL) groups.

The following is a list of these files. The specific subroutine names and their functions are given for each file.

GHARD.FOR	Hardware routines for both Picker and Huber
ECHO	Check and verify motor drive complete
OPEN	Open shutter
CLOSE	Close shutter
HOME	Drive all angles to zero
ANGSAV	Save current angle values in DIFF.;1
COUNT	Count for specified time (sec)
RECOVER	Reset stepping motor controller
SETINC	Sets angle increments for some scans
SETMOVE	Sets micro steps per 0.01° for specified axis
PIHARD.FOR	Hardware routines for the Picker only
ACC	Set acceleration rates
BASE	Set base speeds
DDRIVE	Set drive command & target to select angle
DRIVE	Set drive command & target to select angle
ZERO	Determine true zero for all or selected angle
SETSPEED	Set high speed limit for selected angle
DAA	Drive all angles to specified target <i>via</i> DDA
DDA	Set move registers for setting angle
SCAN	Scan selected angle and return counts
TTSCAN	Scans 2θ and returns counts
HS HARD.FOR	Hardware routines for the Huber only
ACC, BASE, DDRIVE, DRIVE, ZERO, SETSPEED, DAA, DDA, SCAN & TTSCAN	Functions as in PIHARD but specific for Huber.
MHARD.FOR	Hardware routines for the FDD
STARTFDD, FDDLIM, STVAL, TENS N, RSTAT, RLIMIT, MSTEP, SETTNS & FDDMAN	These are discussed in detail in another report.
GUTIL.FOR	Utility routines for both the Picker and Huber
TDATE	Finds current time and date
TFIN	Converts estimated run time to finishing time
ACK	Check for angle out of range & allow temporary change
BRAG2T	Drives θ and 2θ to maintain Bragg condition for Huber. For Picker, drives 2θ in normal manner.
CRATE	Drives to selected angles and returns CPS for counting time specified
START	Opens the VAX Port & initializes angles and sets up angle limits
INITM	Sets angle limits and motor speeds
INIT	Initializes angles and calls INITM
R100	Multiply by 100 and round
SPH360	Scans a complete revolution of φ
CHECKP	Creates stop file
NORM	Normalize angles to ±180°

PIUTIL.FOR	Utility routines for the Picker only
LIMITS	Limits on diffractometer angles are specified in this subroutine
SLEW	Sets slew speeds as specified here
PMAN	Subroutine version of PIKMAN
DA	Drives selected axis to specified angle
SA	Scan angle to pick up counts during scans
HSUTIL.FOR	Utility routines for the Huber only
LIMITS, SLEW, HMAN, DA	As in PIUTIL but specific for Huber.
MUTIL.FOR	Utility routines for the FDD
RMODIN, WMODIN and LINFIT	are discussed in detail in another report (Lenhert and Adams, 1990).

This grouping allows all routines to be stored and accessed in a convenient way so that only those needed for a particular EXE file are included.

Some routines required modification. The speed at which the various goniometer angles are driven was adjusted and angle limits, which prevent collisions, were also changed to correspond to the requirements of the experiments to be done on the Picker and Huber systems. The routines that detect the zero points of the four goniostat angles required special attention to adjust both the speed and direction of the zero search.

A subroutine, BRAG2T, has been written and included in the GUTIL.FOR file. On the Huber, BRAG2T drives ω and 2θ together (at slew speed) so that the Bragg condition is approximately maintained while 2θ is changed. If the VAX port indicator is set for the Picker, *i.e.*, to TXA0, BRAG2T changes 2θ without driving ω , thus the subroutine can be called in programs for either diffractometer system.

The BRAG2T subroutine has been tested and best results are obtained with the speed and acceleration factors presently set in the Huber hardware routines. For the details of changing slew speeds, see page 11 of "Instructions for *Modulynx* Motor Control Interface Card, Type I0D004A".

C. UTILITY PROGRAMS

Stability Test of the Picker Mount, STABLE

A computer program which makes repeated measurements of the X-ray source intensity (STABLE) was written and used to test the stability of the Picker-RU-200 mount. First the incident and diffracted beam apertures were adjusted so small movements of the diffractometer relative to the X-ray source would produce large changes in the measured X-ray intensity. STABLE was started and the X-ray intensity was monitored over night. At first there were large changes in intensity. The diffractometer "legs" were suspected as the main source of the instability. They were modified so the height adjustment screws could be clamped with a lock nut, and the system stability improved to a satisfactory level.

STABLE works by repeatedly counting for a specified number of seconds at a specified 2θ . At intervals, *i.e.*, after a specified number of observations, a step scan is made across the peak (usually the direct beam, *greatly attenuated!*) to check for any shift of the

diffractometer relative to the X-ray source. When the run is complete, the variance of the measured counts is calculated and compared with the expected variance based on normal counting statistics. The results are in VODAT.TMP. An example of a result obtained with STABLE is given in Appendix F.

STABLE requires subroutines in GUTIL, GHARD, PIUTIL and PIHARD. The program has not been adapted for use with the Huber system.

Angle Driver Test Program, ANGTST

Some slippage was noted at the motor shaft couplings when the *Crystal Logic* couplings were initially installed. This suggested that the couplings (and perhaps the stepping drivers) could be a source of error in setting the angles. In order to verify the reliability of the couplings and stepping drivers, a test program, ANGTST, was written. ANGTST is entered with all angles initially set to zero. ANGTST then drives all motors to their limits and back, repeating the exercise with stops at gradually decreasing intervals. When the test is completed, a visual check of the angle odometers and scales should be made to see if all angles read zero. If any angles show non-zero readings, either the coupling has slipped or the stepping driver has failed.

ANGTST requires subroutines in GUTIL, GHARD, PIUTIL and PIHARD. The program has not been adapted for use with the Huber system.

Alignment Software for the Picker and Huber, PALIGN & HALIGN

Correct alignment of a diffractometer is best accomplished with a small highly symmetrical crystal such as basic beryllium acetate. The alignment procedure includes centering crystal reflections and then, on the basis of the diffractometer angle settings thus determined, adjusting the diffractometer position and/or the angle scales. Computation of the reflection settings from the orientation of the alignment crystal is required as well as calculation of the crystal orientation, given the angle settings which center the crystal reflections. The alignment process requires several steps. After preliminary adjustment to maximize the X-ray intensity in the incident beam collimator, one must determine the best settings of the diffractometer angles for a series of crystal reflections, *i.e.*, those that center the reflections in the detector aperture. For a more detailed discussion of diffractometer alignment, see Lenhart (1978).

These centering angles may indicate changes in the relation of the diffractometer to the X-ray source and/or error in the zero settings of the diffractometer shafts. Indicated changes should be made. Various subroutines find the angles which center the reflections and carry out other required calculations.

The main program, PALIGN (or HALIGN), provides a menu and calls subroutines to carry out the requested operations. They aid in aligning the respective diffractometers and they can also be used to orient samples for various types of diffraction measurements.

Centering angles for reflections are found with the intensity optimization subroutines in CRSUBS. The three diffraction angles of interest, 2θ , ω , and χ are optimized by finding the settings on each side of the peak which give half of the peak count rate. These are averaged to give the best value for the angle. A series of reflections can be optimized with

approximate setting angles stored in the file PEAKS. Optimized settings are written into CPEAKS.

One subroutine, MT, calculates the orientation matrix and cell constants of the crystal given three non-coplanar reflections. MT was translated from the PDP 8 assembly language of the old Vanderbilt FACS-I program system. Another subroutine, called ANGLES, calculates diffractometer setting angles when the Miller indices (h,k,l) are input. It uses the cell constant-orientation matrix stored in OMMAT.DAT. If more than three reflections are used to determine the cell constants and/or crystal orientation, the excess information can be included in the calculation by using a linearized least-squares calculation. The subroutine REFINE, which does this, requires as input, preliminary values for the cell constants and setting angles for two orienting reflections. A list of setting angles, treated as observations, is read from a file, LEAST., and subjected to the least-squares calculation. Examples of lines from the files, HKL.DAT, PEAKS., CPEAKS. and LEAST. are shown below in Table 5.

Table 5. *The four lines in this table show the format of the files HKL.DAT, PEAKS., CPEAKS. and LEAST., respectively.*

4.	2.	-3.						
200	-4620	-0000	27000	9000				
0	0	0	22.56	0.00	-90.00	90.02	16011	
2	-10	0	28.98	359.94	334.57	287.30	0	

PALIGN and HALIGN require subroutine files FIND, CRSUBS, REFINE, BBMB, MATRIX, GHARD and GUTIL along with the diffractometer specific subroutine files PMAN or HMAN, PIUTIL or HSUTIL and PIHARD or HSHARD.

D. ADJUSTING the AMPLIFIER and PHA SETTINGS

Follow the steps given below for correct pulse discrimination settings. Beware that the specific values may change as the circuits age or if a radiation other than $\text{CuK}\alpha$ is used. If the settings are to be adjusted, use a strong single crystal reflection, not a polymer sample. Avoid count rates too high because coincidence losses can fool you into thinking you have optimum settings when in fact, you are off the peak.

1. Connect channel 1 of the laboratory oscilloscope (*Tektronix 465M*) to the AMP output port on the 590A *Ortec* module. Channel 1 on the oscilloscope should be set to DC and the gain should be 2 for the 1X probe setting. This will make the one cm divisions on the CRT screen equal to 2 V.

2. Select a strong single crystal reflection (a KAP monochromator crystal works well) and adjust the Kv and ma settings on the X-ray generator so the count rate is 30,000 cps or less. This will avoid problems with coincidence losses.

3. The High Voltage Power Supply, *Ortec* unit 556, should be set so the coarse HV is 500, mid range HV 100.

4. The Amplifier gain should be set at 100 with the fine adjustment near mid scale.

5. The HV and Amplifier fine adjustments can now be set so the $\text{CuK}\alpha$ pulse height is about 5 V (2.5 cm on the oscilloscope screen). Make most of the required adjustment with the HV setting. Recent settings were with HV at 2.35 and Amp fine setting centered in its range.

6. The Window and Lower Level settings should now be adjusted. The window must be centered on the peak presented by the amplifier, about 5 V (step 5). The width of the window should be sufficient to include all pulses from $\text{CuK}\alpha$ photons. For statistical reasons these vary by about $\pm 35\%$. A minimum width setting of 3.5 V or more should therefore be used. To avoid problems with small amounts of drift, the width should be increased above the 3.5 V minimum to about 4.5 V for $\text{CuK}\alpha$. 4.45 V is the current setting.

7. The lower level (LL) setting is the voltage at the low voltage end of the window. If LL is adjusted, the window is moved but its width is not changed. The current LL setting is 3.1 V.

8. These settings can be checked by testing to see if an increase or decrease in width gives a minimal change in count rate. Finally, the LL setting should be checked by moving it systematically up and down to verify that it is set to give the maximum count rate.

E. DEAD TIME DETERMINATION and CORRECTION

Method of Chuck Strouse

The following is a simple procedure for the measuring counter dead time. This procedure is based on the measurement of intensities of two reflections, one strong and one weak, as a function of incident flux. The strong reflection should be one that has significant coincidence losses at high flux. The weak reflection should be one that can be assumed to be unaffected by coincidence losses (about 1000 cps should be OK). If W is the observed intensity of the weak reflection, and R is the ratio of the *true* intensities, then, to a good approximation, RW , is the true intensity of the strong reflection. The observed intensity, S , of the strong reflection should be given by

$$S = RW(1 - DRW) = RW - DR^2W^2$$

where D is the dead time of the counter in seconds. Dividing this equation by W gives

$$\frac{S}{W} = R - DR^2W$$

Thus if S/W is plotted versus W for different values of the incident flux, one should obtain a straight line, the slope and intercept of which should provide values of R and D .

Rates for Strong and Weak "Reflections" on the Picker System

Incident X-ray flux was provided by the reflection from a silicon monochromator crystal at $28.36^\circ 2\theta$. The 0.001 in Ni filter was used in the incident beam with the 0.5 mm incident beam and 1.5 mm diffracted beam collimators. The SVA was set to give count rates such that the Al attenuator filters could be used to give the two "reflections," i.e., a strong and a weaker count rate. Two experiments were carried out, since upon analysis, it seemed probable that the dead time obtained was affected by coincidence losses at the lowest count rates. In both cases, the minimum number of counts measured for each observation was 500,000 so all measurements have σ 's of $\pm 0.14\%$ or less.

For Series I, the X-ray voltage was fixed at 30 Kv and the *S* and *W* measurements were made at 10, 20, 100 ma. The same attenuator was placed in the beam each time between the *S* and *W* measurement. Values obtained (CPS) were:

ma	S (cps)	W (cps)	S/W
10	14340.17	546.76	26.2275
20	27159.90	1113.94	24.3818
30	38374.70	1696.19	22.6241
40	47802.32	2271.97	21.0400
50	55416.05	2832.74	19.5627
60	62224.95	3363.55	18.4998
70	68303.85	3896.49	17.5296
80	73650.25	4426.47	16.6386
90	78530.35	5013.09	15.6651
100	81609.75	5609.83	14.5476

To obtain the dead time, *D*, values of *S/W* vs. *W* were fit by linear least-squares (subroutine LINEFT). Since *S/W* vs. *W* is not accurately linear, one obtains different values of *D* by using the full data set from Series I or parts of it. For example, with all ten points, one gets 3.1 μ s for the dead time. If the first seven points are used, one gets 3.5 μ s, for the first five, 3.8 μ s and for the first three, 4.0 μ s.

It was postulated that coincidence losses in the low count rate "reflection" affected the calculated dead time. In order to correct for this effect, a modified approach was used in which the *S* and *W* "reflections" were corrected for the coincidence loss based on the first dead time calculation, then the new (corrected) ratios, *S/W*, and *W* were used to repeat the least-squares fit. One then gets a larger value of the dead time. If the procedure is iterated, it converges to a dead time of 6.15 μ s. If the first seven, five and three points are used and the iteration is carried to convergence, one finds *D* as 5.92, 5.83 and 5.48 μ s respectively.

The measurements were then repeated (Series II) with the same experimental conditions, except for the attenuation filters, in order to eliminate the effects of coincidence loss from the points with the lowest count rates. The rates below were obtained:

ma	S (cps)	W (cps)	S/W
10	6965.07	256.67	27.1363
20	13129.28	498.09	26.3593
30	18850.4	737.33	25.5658
50	29006.65	1210.65	23.9598
70	37579.00	1622.71	23.1583
90	44381.13	2021.19	21.9579

When all six of these points are fit by least-squares with no iteration one gets 3.78 μ s, for the first five, four and three points one gets 3.86, 4.25 and 4.17 μ s respectively. With iteration and corrections as above, using the same data points one gets 5.06, 4.95, 5.34 and 4.88 μ s respectively.

What is the correct value of the dead time? We reject all values calculated from Series I since Series II should give dead time values less affected by non-linear coincidence losses. In fact, the values for the dead time obtained from Series II do not show a consistent change as the higher count values are eliminated, especially when the iteration method is used.

Consider the result of applying the coincidence loss formula:

$$N_t = N / (1 - ND)$$

where N is the measured and N_t the corrected count rate (cps). If D is taken as 5.0 μ s, the corrected value of W (Series II) for the 10 ma setting is found to be unchanged but the 90 ma value for W is about 1% too low, a violation of the assumptions used in deriving the basic formula of the method. Therefore, we conclude that the best value calculated for the dead time from the above data is 5.06 μ s.

The RU-200 X-ray generator used for these measurements is run at constant potential.

Correction for Coincidence Loss

Subroutine COUNT in GHARD has been modified to correct all counts measured with the Picker system. The dead time used in the above formula for N_t is 5 μ s and all measurements made with COUNT are corrected, i.e., N_t is returned rather than N . If the raw count rate is greater than 50,000 cps, the corrected value is returned as a negative number which can be interpreted as appropriate to the experiment in progress.

F. RU 200 X-RAY STABILITY TEST

The difficulty of obtaining consistent modulus scan results on G30 carbon prompted an investigation of the X-ray output stability. Two experiments were run. Both used the diffracted intensity from the 43.25° peak of G30 carbon (modulus sample G30a). Instrument settings were: SVA 3x6 mm, collimators; 1.0 mm incident, 1.5 mm diffracted, 45 Kv, 70 ma, no filter. The count rate was about 480 CPS. The program STABLE was used to measure repeated counts for a fixed time interval.

In Experiment I, the counting time was 400 s and 168 measurements were made over a period of 19 hr. The results are plotted in Fig. 7. which shows total count accumulation in 400 s plotted against observation number. The standard deviation, based on counting statistics, is 430 counts which is a little less than the distance between tick marks on the count axis in the figure. Fig. 8 shows the result for the 175 measurements made with counting times of 1000 s in Experiment II which extended over 49 hr. In the figure, the COUNTS scale is shortened by a factor of 2.5 so that a given percent change in counts is

represented by the same distance on each plot. Here the standard deviation is 680 counts, less than half the spacing of the tick marks.

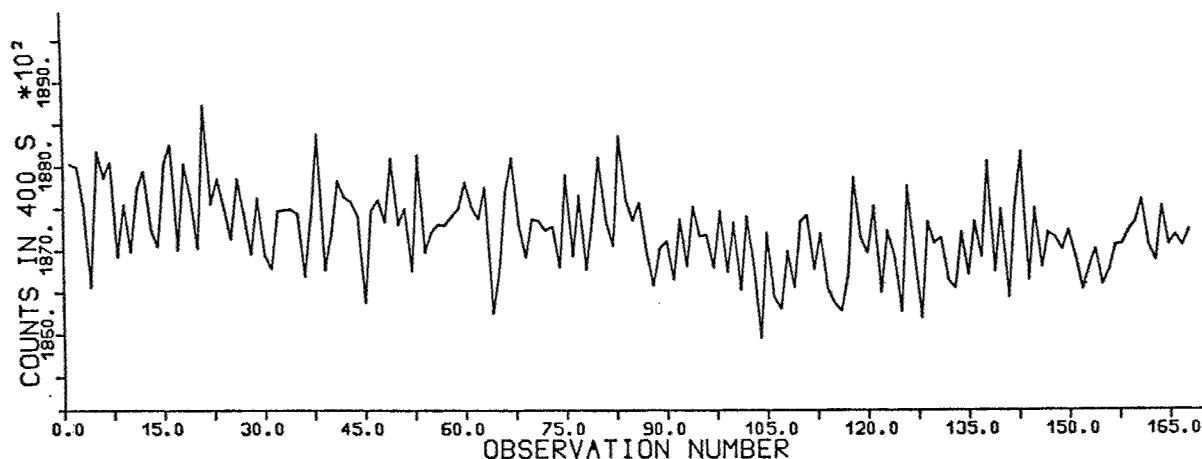


Figure 7. Count rate at 400 second intervals.

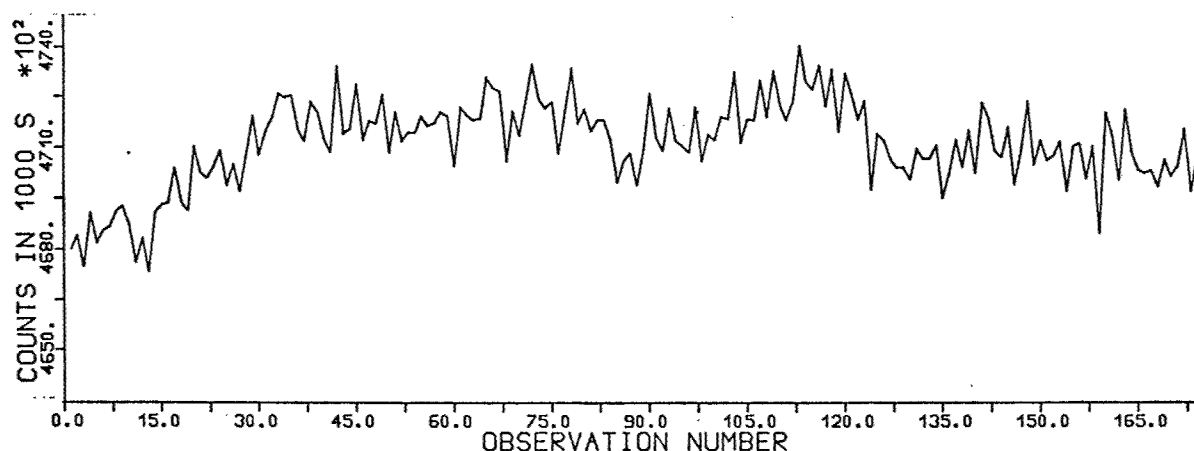


Figure 8. Count rate at 1000 second intervals.

In both figures, the apparent count rate frequently changes by several standard deviations over a time scale of a few observations, *i.e.*, less than the time required to scan one modulus data point with either a 400 s or 1000 s step count time.

G. HUBER ALIGNMENT NOTES

The following steps are a summary of the alignment of the Huber Diffractometer done by PGL with assistance from Dave Anderson.

1. The table was raised and leveled with a high quality level. The table height was set so that the beam tunnel (anode to monochromator) was level.

2. The monochromator crystal was translated until the X-ray beam intersected it in the center with the vertical entrance slits set symmetrically left and right. This was accomplished by opening the monochromator housing and observing the beam with a fluorescent probe. The monochromator baffle was set so that any part of the incident beam that missed the monochromator crystal would be stopped.

3. The monochromator angles, 2θ and ϕ , were set approximately to give a maximum intensity as observed with the probe. The monochromator exit slits were set symmetrically most of the way open.

4. Next, the diffractometer was moved across the beam until counts could be observed. The motion across the beam is actually a rotation about a vertical axis through the incident beam monochromator. Levitation air is on and the magnetic base micrometer is used. The collimator slits at the sample end of the beam tunnels were wide open and the diffracted beam monochromator was placed out of the beam path for this step.

5. When the direct beam of the monochromator is counted, the lowest X-ray settings, 20 Kv, 10 ma and a suitable attenuation filter should be used. The beam stop is removed.

6. Adjustments available for changing the diffractometer position are the 2θ arm (mentioned above) and the diffractometer rotation. The monochromator 2θ arm adjustment is made by the micrometer with the magnetic base. The diffractometer itself can be rotated by adjusting the micrometer in front of the diffractometer which is fixed to the 2θ arm. Levitation air is on for these steps.

7. The collimator beam tunnels have identical support brackets. Three independently adjustable legs provide height adjustment. The track that holds the collimator tube can be rotated about a vertical axis or translated perpendicular to the axis of the collimator. Rotation is about a pivot near the front of the collimator bracket. The adjustment screw is on the right, near the rear of the bracket. Before adjustment, the clamp screw must be loosened. It is under the bracket, near the rear edge. Translation is by means of two opposing screws on the right and left sides just forward of the bracket center. To translate, the two lower clamp screws must be loosened.

8. When the x-ray beam has been located at the counter, maximize it by adjusting the 2θ arm under the diffractometer. Don't use the diffracted beam monochromator.

9. The goniometer base mounted slits are now useful for centering the X-ray beam on the diffractometer axis. The slits must be attached to the goniometer mount on the diffractometer and centered using the diffractometer microscope. After centering the slits by adjusting the translation screws on the goniometer base, open both the vertical and horizontal slits symmetrically to approximate the size of the X-ray beam expected. This now provides a centered target to aim the beam at. Ensure that the collimators are in approximately correct positions and that the collimator slits are open. The beam can now be centered by adjusting the diffractometer position *via* the monochromator 2θ arm. The intensity should also be maximized by translating the monochromator crystal and tuning for maximum intensity, and, when needed, rotating the monochromator crystal with the monochromator ϕ adjustment. Iterate these steps until you have a centered beam with maximum intensity.

10. If the microscope is not known to be centered, a goniometer head with a fine wire in it can be mounted on the diffractometer to use as a target for centering the microscope. The microscope adjustment, if needed, is made with the four small screws near the objective lens.

11. At this point in the procedure, the X-ray beam is centered on the diffractometer axis. All slits down beam from the monochromator entrance slits can be set on the edge of the beam by closing them one at a time until the count rate starts to drop. If the X-ray beam is not centered in a collimator, adjust it by translation, rotation or height adjustment.

12. At this point, the X-ray beam is centered in all collimators and it passes through the diffractometer axis. You must also be sure that it is perpendicular to the diffractometer axis. This can be determined with the help of an alignment crystal and the program HALIGN which is discussed above.

When Huber alignment tune-up was attempted it was decided to move the monochromator support in order to better center the monochromator crystal. The support was translated toward the RU-200 side. After the support was moved, the position of the monochromator crystal was adjusted. The procedure consisted of the following:

1. With the entrance slits open several millimeters the monochromator angle was adjusted to give a strong reflected beam. Lights were out and 40 Kv, 50 ma settings were used to view the beam with the florescent probe.

2. The next step was to get the beam centered in the collimators. Air is on, the collimator scatter slits are fully open and the monochromator exit slit is open several mm. Move the diffractometer using the monochromator pivot arm until the beam is visible at the sample position.

3. Now insert the slits that are mounted on the goniometer base at the sample position and adjust the slits symmetrically. Adjust the diffractometer position further if necessary to get the beam through the sample slits.

4. Now make sure the monochromator entrance slits are closed symmetrically to give a vertical opening with a width of about 1 mm.

5. At this point the diffractometer position should be fine tuned to exactly center the beam in the sample aperture.

6. Rotation of the diffractometer about the center pivot could be used to center the beam in the diffracted beam (SVA) aperture but the problem is that this also affects the position of the receiving end of the incident beam collimator at the mono exit slits. There should be an adjustment for rotation of the outside housing of the monochromator but so far I haven't found it.

7. To make the final adjustments, the sample slit was set to 1 mm by 1 mm and the scatter slits on the collimators were adjusted to the edge of the beam. The incident apertures (before and after the monochromator) were adjusted to give maximum intensity through the 1 mm square sample aperture.

8. After the above adjustments the beam strength was measured. Calibrated Al filters with known mass/unit area were used to attenuate the beam and the direct, attenuated beam was counted. The original beam strength was calculated using the attenuation factors. Filters #7, 9, 10 were used. The attenuation factors were .4827, .2261, .1213 respectively. At 45 Kv 60 ma there were 18,000,000 cps.

H. BUILDING EXE FILES

For XRYMOD.EXE, LINK

XRYMOD, MODIN, CALIBR, MSCAN, PKFIT, CALMOD, MUTIL, CCM88, MHARD, PIHARD, PIUTIL, GH/
GUTIL

For PALIGN.EXE, LINK

PALIGN, MATANG, FIND, CRSUBS, REFIN, GHARD, GUTIL, PIUTIL, PIHARD, BBMB, MATRIX

For HALIGN.EXE, LINK

HALIGN, MATANG, FIND, CRSUBS, REFIN, GHARD, GUTIL, HSUTIL, HSHARD, BBMB, MATRIX

For PIKMAN.EXE, LINK

PIKMAN, PIUTIL, PIHARD, GHARD, GUTIL

For HUBMAN.EXE, LINK

HUBMAN, HSUTIL, HSHARD, GHARD, GUTIL

For PICKER.EXE, LINK

PICKER, PIUTIL, PIHARD, GUTIL, GHARD, SCNIN, THETAS, TTSSCN, CHIS, CHSSCN, CHIZRO, OM
OMSSCN, PEAKFT, LLSCAN, STDSCN, BBMB, MATRIX, MHARD

For HUBER.EXE, LINK

HUBER, HSUTIL, HSHARD, GUTIL, GHARD, SCNIN, THETAS, TTSSCN, CHIS, CHSSCN, CHIZRO, PE
LLSCAN, STDSCN, BBMB, MATRIX, MHARD

For FIT.EXE, LINK

FIT, FIT1, FIT2, FIT3, CCM88

For FOLD.EXE, LINK

FOLD, SPLINE, FOUR

For UNFOLD.EXE, LINK

UNFOLD, SPLINE, FOUR

For FITSM.EXE, LINK

FITSM, POLYF

For LLPLOT.EXE, LINK

LLPLOT, PINTP, POLYF, MATRIX, BBMB, CCM88

For PLOT1.EXE, LINK

PLOT1, CCM88

I. VAX PORT PARAMETERS

```

Terminal:  _TXA0:      Device_Type: Unknown      Owner: PETER
Input:      9600      LfFill:  0      Width:  80      Parity: None
Output:     9600      CRfill:  0      Page:   24
  
```

Terminal Characteristics:

Passall	No Echo	Type_ahead	No Escape
No Hostsync	No TIsync	Lowercase	No Tab
Wrap	Scope	Remote	No Eightbit
No Broadcast	No Readsyc	No Form	Fullcup
Modem	No Local_echo	No Autobaud	No Hangup
No Brdcastmbx	DMA	Altypeahd	Set_speed
No Line Editing	Overstrike editing	No Fallback	No Dialup
No Secure server	No Disconnect	No Passthru	No Syspassword
No SIXEL Graphics	No Soft Characters	No Printer Port	Numeric Keypad
No ANSI_CRT	No Regis	No Block_mode	No Advanced_vide
No Edit_mode	No DEC_CRT	No DEC_CRT2	

```

Terminal:  _TXA1:      Device_Type: Unknown      Owner: No Owner
Input:      9600      LfFill:  0      Width:  80      Parity: None
Output:     9600      CRfill:  0      Page:   24
  
```

Terminal Characteristics:

Passall	No Echo	Type_ahead	No Escape
No Hostsync	No TIsync	Lowercase	No Tab
Wrap	Scope	Remote	No Eightbit
No Broadcast	No Readsyc	No Form	Fullcup
Modem	No Local_echo	No Autobaud	No Hangup
No Brdcastmbx	DMA	Altypeahd	Set_speed
No Line Editing	Overstrike editing	No Fallback	No Dialup
No Secure server	No Disconnect	No Passthru	No Syspassword
No SIXEL Graphics	No Soft Characters	No Printer Port	Numeric Keypad
No ANSI_CRT	No Regis	No Block_mode	No Advanced_vide
No Edit_mode	No DEC_CRT	No DEC_CRT2	

```

Terminal:  _TXA2:      Device_Type: Unknown      Owner: PETER
Input:      1200      LfFill:  0      Width:  80      Parity: None
Output:     1200      CRfill:  0      Page:   24
  
```

Terminal Characteristics:

Interactive	Echo	Type_ahead	No Escape
No Hostsync	ITsync	Lowercase	No Tab
Wrap	Scope	No Remote	No Eightbit
No Broadcast	No Readsyc	No Form	Fullcup
Modem	No Local_echo	No Autobaud	No Hangup
No Brdcastmbx	DMA	Altypeahd	Set_speed
No Line Editing	Overstrike editing	No Fallback	No Dialup
No Secure server	No Disconnect	No Passthru	No Syspassword
No SIXEL Graphics	No Soft Characters	No Printer Port	Numeric Keypad
No ANSI_CRT	No Regis	No Block_mode	No Advanced_vide
No Edit_mode	No DEC_CRT	No DEC_CRT2	