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# DOE PHASE I STTR - FINAL TECHNICAL REPORT

## Contract No. DE-SC0020555

Adelphi Technology, Inc.
2003 East Bayshore Road
Redwood City, CA 94063

**Principal Investigator:** Dr. Jay Theodore Cremer, Ph.D.

**Project Title:** *Multiplexing Focusing Analyzer for Efficient Stress-Strain Measurements*

**Topic:** 18c

**Proprietary Data Legend**
Not Applicable

# Project Summary

*Company:* Adelphi Technology, Inc.
*Title:* Multiplexing Focusing Analyzer for Efficient Stress-Strain Measurements
*PI:* Dr. Jay Theodore Cremer
*Topic:* 18c

**Statement of the problem or situation that is being addressed.**

Although thermal and cold neutron scattering is widely used and is critical for success in many areas of materials science and engineering, relatively low neutron fluxes severely limit applications of not only laboratory neutrons generators, but also large national neutron facilities.

State-of-the-art thermal and cold neutron sources are large expensive national facilities, which serve diverse community of scientific and industrial users. The constant need to improve the instruments performance, stems from the fact that neutron methods are gaining in popularity, and becoming more and more powerful, while new neutron sources are not being constructed to keep pace with the developments and needs of the scientific community.

Small research reactors at universities and National Labs, and laboratory-based neutron generators, are necessary not only for education and training, but also when samples cannot be transported to other facilities. However, the standard neutron techniques, which were developed for high-flux facilities, require much higher efficiencies to be used effectively with the low fluxes of small sources. Thus, the efficient use of neutron sources, such as with our proposed analyzer, is important for the progress and broader use of these neutron techniques.

**General statement of how this problem is being addressed.**

We propose to design and demonstrate novel diffractive optical device, which will enable very efficient residual stress neutron diffractometers. The proposed device will be a multi-foil analyzer, where each foil is constructed of focusing bent single crystals of Si. Such device will enable polychromatic residual stress neutron diffraction. At large national facilities, such as at Oak Ridge National Laboratory, these analyzers would enable very fast measurements for determining residual stress tensors, raster large samples or screen multiple samples.

**Commercial Applications and Other Benefits**

The outcome of this project would be the demonstration of commercial devices, novel neutron optical components, which could be utilized to improve the performance of existing instruments or build novel neutron scattering instruments at DOE neutron facilities and commercial laboratory neutron sources. These new devices will widen the scope of research conducted using neutrons and enable measurements not feasible at present.

**Key Words** Bragg diffraction optics, residual stress measurements, neutron diffraction

**Summary for Members of Congress**

Thermal and cold neutron beams are a powerful materials science probe, which provide unique information about the structure of matter. The proposed innovations expand the reach of neutron-based investigations to new materials and industries by enabling new instrumentation capabilities, thereby greatly enhancing and expanding the role of small, laboratory-based neutron instrumentation, and improving education and training of neutron users.

In Phase I, we conducted feasibility studies of the focusing perfect-crystal analyzer and the applicability of the bent single-crystal devices for use in polychromatic powder diffraction, **Fig. 1**.

Our goal is to utilize the so-called "thickness focusing" based on previous theoretical developments, especially in Ref. [6] where it was shown that under certain conditions bent perfect-crystal analyzers can be rather thick to increase the number of reflected neutrons without degrading the resolution.

We conducted a combination of analytical calculations, ray-tracing simulations, and preliminary experiments to demonstrate the focusing. We concluded that the combination of bent crystals and position-sensitive detectors can be used very effectively with a polychromatic beam when certain geometrical conditions are satisfied.

We determined that such an arrangement, called "dispersive diffraction imaging" has not been implemented before but is feasible. Our project is to develop multiplexing double-focusing analyzers suitable for polychromatic neutron diffractometers, namely a **Bent perfect crystal multi-wafer analyzer.**

Proposed analyzers are made of bent multi-wafer single crystals following the developments of the so-called "thickness focusing " [6,7].

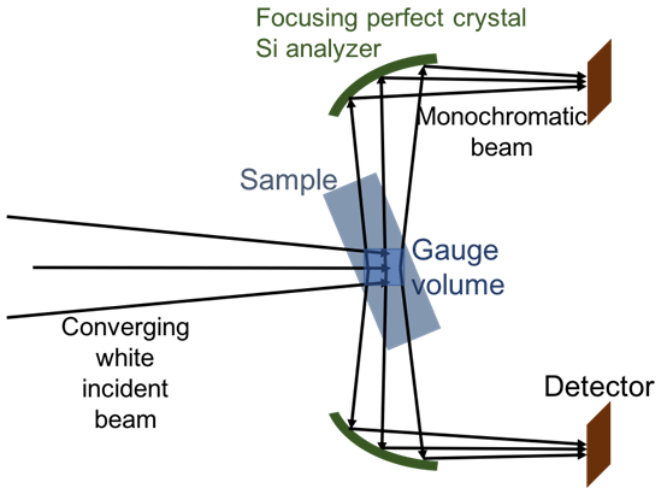


**Fig. 1** Schematic drawing of the focusing bent Si crystal analyzers for residual stress diffraction.

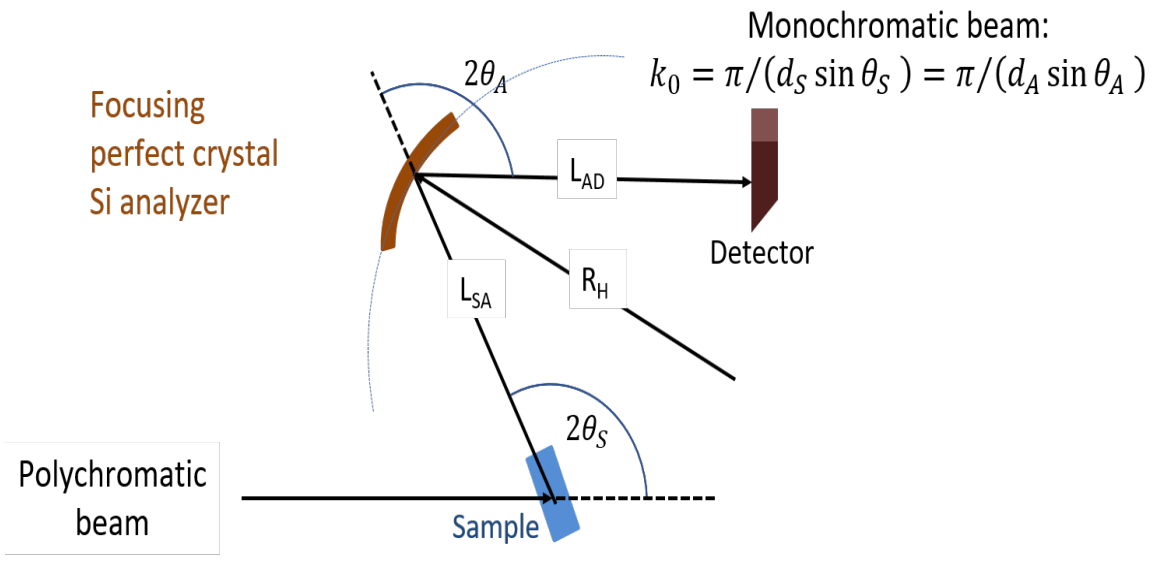


**Fig. 2** Schematic illustration of the polychromatic diffractometer showing diffraction angles, distances between the sample, analyzer, and detector (L_AD,L_SA), and the radius of curvature (R_H) of the analyzer.

The proposed analyzers are made of bent multi-wafer single crystals following the developments of the so-called **"thickness focusing "** [6,7].

One particularly successful application of Bragg optics is in residual-strain measurements. The monochromators for this application are made from stacks of thin Si wafers (multi-wafer monochromators), which allow elastic bending up to 1 m radius of curvature [8]. Since Bragg reflection is specular, reflection from curved crystallographic planes can be approximated by geometrical optics principles, but dispersive (wavelength-dependent) diffraction properties in thick crystals offer new possibilities of manipulating the neutron beams.

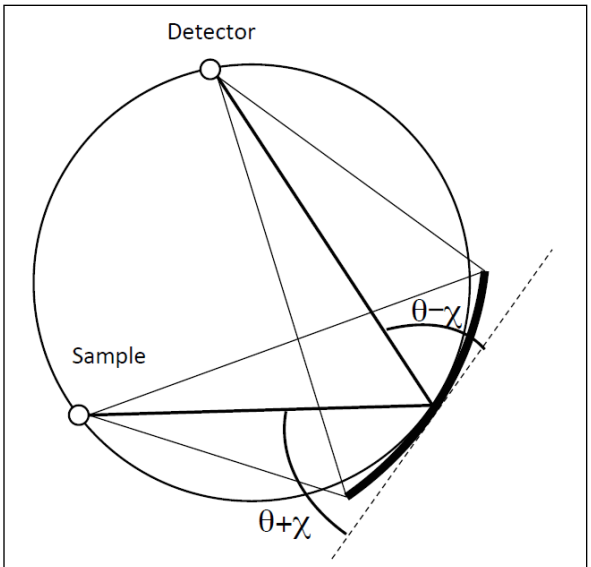


**Fig. 3.** Focusing analyzer schematic, after [4].

In curved monochromators, the horizontal radius of curvature creates a convergent neutron beam at the sample position with a tight correlation between the neutron's wavelength and direction. In particular conditions, the width of a diffraction line can be minimized as detected by a position-sensitive detector [9]. Many engineering diffractometers are using such monochromators at major research reactors: HIFR (Oak Ridge), NIST, ILL (Grenoble, France), FRM-II (Munich, Germany), Prague Research Reactor, etc.

We note that bent perfect crystals can be used as analyzers in various dispersive or non-dispersive arrangements [6]. The feasibility of energy transfer analysis with a Si double-bent device was demonstrated in [7] but not implemented in neutron facilities.

Here we propose to develop another application: focusing diffraction analyzers. The inspiration for this design is found in Ref. [6], chapter 3.3 "Dispersive imaging in diffraction". The bent silicon analyzer will reflect neutrons diffracted by a sample in a limited sampling volume and reflected neutrons will be detected by a PSD. Each diffraction line will be detected at a particular location at the PSD, according to the instrument calibration curve.

Following the general approach described in [6], we describe the general properties of such focusing analyzers. As shown in Fig. 2, the matching of diffraction angle of sample and analyzer enforce the obvious relationship:

$$d_A \, sin\theta_A = \, d_S \, sin\theta_S \tag{1}$$

And since the sample and analyzer d-spacings are not the same, a difference parameter is defined:

$$\kappa_{SA} = 1 - \frac{tan\theta_A}{tan\theta_S} \tag{2}$$

The two angles are absolute values and the expression corresponds to the parallel setting where the two angles are changing the sign. As the family of diffracting planes can be inclined relative to the face of the crystal ( $\chi_A$ - cutting angle), a reflection asymmetry parameter is defined:

$$\xi_A = \frac{cos(\theta_A - \chi_A)}{cos(\theta_A + \chi_A)} \tag{3}$$

When this parameter is less than 1, the setting implies beam compression, usually named the Fankuchen cut. Using eq. (27) from Ref. [6], the radius of curvature necessary for focusing in diffraction has to satisfy the following equation:

$$R_H = \frac{2\,cos(\theta_A+\chi_A)}{(1+\kappa_{SA})\,sin2\theta_A}\,(\kappa_{SA}L_{AD} + \xi_A L_{SA}) \quad (4)$$

The $L_{SA}$ is the distance between sample and analyzer and $L_{AD}$ is the distance between analyzer and detector, and $R_H$ is the horizontal (in the scattering plane) radius of curvature of the analyzer. The schematic diagram describing the geometry is shown in **Fig. 2** and **Fig. 3.**

The range of d-spacing, which can be analyzed by a single exposure, is determined by the crystal length ($l_A$):

$$\frac{\Delta d}{d_0} = \frac{l_A}{2R_A} \quad (5)$$

Here $d_0$ is the d-spacing values corresponding to the middle of the analyzer and the scattering angle value from (1). The range of d-spacings projected along the detector coordinate $y_D$ will be:

$$\Delta y_D = \frac{(\kappa_{SA}L_{AD}+\xi_A L_{SA})}{1+\kappa_{SA}} \times tan\theta_A \frac{l_A}{R_A} \quad (6)$$

A simple expression for the resolution can be also written in linear approximation:

$$\left\langle\left(\frac{\Delta d}{d}\right)^2\right\rangle = \frac{1}{4}\,cot\theta_S\,\langle\gamma_0^2\rangle + \frac{1}{4}\left(\frac{1+\kappa_{SA}}{L_{AD}-\xi_A L_{SA}}\right)^2\,[\langle(\delta y_D)^2\rangle + \xi_A^2\,\langle(\delta y_S)^2\rangle] \quad (7)$$

The brackets are symbolizing standard deviations from the average values; $\langle\gamma_0^2\rangle$ – represents the divergence of the incident white beam; $\delta y_D$ and $\delta y_S$ refer to the extension of detector spatial resolution and the sampling expansion range, respectively.

Analyzing this expression, we will expect to obtain better results if the magnification factor *M* is high and the analyzer is in beam compression, where

$$M = L_{AD}/L_{SA} \quad (8)$$

Interestingly, $d_A < d_S$ configuration will be also possible, as $\kappa_{SA}$ becomes negative. However, the preferred scattering angle for strain scanning instruments is $2\theta_S \cong 90°$ and the practical choice for $2\theta_A$ is usually limited to the $70° - 110°$ range.

Three wafer orientations are readily available on the market: <100>, <110> and <111>. With the first type of wafers the (511) and (311) Bragg reflections are possible and the corresponding $\chi_A$ values are 15.8° and 25.3°.

These two reflections allow covering a sample d-spacing range spanning from 0.8 to 1.9 Å by rotating and tuning the radius of the analyzer. For each setting, the d-spacing window is expected to reach 0.1 – 0.2 Å.

One question which must be addressed is the optimal slope (or even shaping) of the detector relative to the nominal direction of the reflected beam, to flatten the d-spacing resolution along the detector.

The last equation for $\left\langle\left(\frac{\Delta d}{d}\right)^2\right\rangle$ determines the reciprocal space resolution, but in strain scanning experiments the spatial resolution is also of concern.

There are two usual ways to restrict $\delta y_S$ by either using a radial collimator or a slit close to the sample. The radial collimator should oscillate to homogenize the transmission and, for practical reasons, cannot provide sampling widths smaller than 2 mm.

On the other hand, the slit cannot be close enough to the sample position due to sample environment limitations or in the case of bulky samples. (The slit can be displaced away from the sample position and pinhole diffraction-based imaging can be employed [10] that was recently demonstrated for a TOF diffractometer.)

In our case, a scan along the incident beam direction is required to map the entire sample, but the spatial resolution will be determined by the slit opening. The method extension to using coded aperture could be possible. Another condition arises to enable vertical focusing. In the vertical direction the lens law must be satisfied:

$$\frac{f_V}{L_{SA}} + \frac{f_V}{L_{AD}} = 1 \tag{9}$$

The focal length $f_V$ is related to the vertical radius of curvature $R_V$ as follows:

$$f_V = \frac{R_V}{2\,cos\chi_A\,sin\theta_A} \tag{10}$$

It is a restrictive condition, which cannot be satisfied exactly because the vertical curvature is usually achieved by segmentation and cannot be reset for different pairs of $\chi_A$ and $\theta_A$. A wafer segmentation of 5 mm is achievable and, with a suitable $R_V$ selection, the vertical spread at the detector location can be maintained inside the 1 – 2 cm range.

Such design allows for a novel way of measuring either powder diffraction or residual stress, in a compact high-resolution instrument. The focusing could be used to increase the flux on the small gauge volume. Most importantly, the major benefit of such an arrangement is the opportunity for multiplexing, as described in **Fig. 1**.

Simultaneous measurements at $2\theta_S = \pm 90°$ would enable efficient measurements of components of the stress tensor. Another option for multiplexing uses the fact that Si crystals, even thick ones, are relatively transparent for neutrons and thus allow for several analyzers to be placed in consequent order, spanning a large range of reciprocal lattice [6].

The signal rate increase is due to the use of a thick focusing analyzer, which reflects a broad band of neutron energies into the same pixel on the PSD. Note that since the illuminating beam is polychromatic, the analyzers can be used at time-of-flight facilities, such as SNS, and steady-state reactor and laboratory neutron sources.

**Goal 1. Feasibility of bent Si analyzers for residual stress and powder diffractometers.**

Focusing conditions determine the distances between the sample, analyzer, and detector to achieve both horizontal and vertical focusing, as described above. We made a Python-based script that encodes the focusing conditions using equations (4), (9), and (10).

**Fig. 4** on the next page shows an example of a screenshot generated by this script. The example is for Si[400] analyzer crystal, which we will use. The sample is Fe [110].

Some other examples are listed in the **Table 1** on the next page.

| Si | | Sample | *(hkl)* | d | $L_{SA}$[m] | $\theta_S$ | $\theta_A$ | $L_{AD}$[m] | $R_H$[m] | $x_D$[m] | $y_D$[m] |
|---|---|---|---|---|---|---|---|---|---|---|---|
| [400] | | Diamond | [220] | 1.2610 | 1.24 | 66.68 | 67.03 | 1.90 | 1.33 | 1.90 | -0.02 |
| | | | | | 1.27 | 67.50 | 67.87 | 1.80 | 1.36 | 1.80 | -0.02 |
| | | PG | [006] | 1.1190 | 1.27 | 67.50 | 55.28 | 2.47 | 1.96 | 2.24 | 1.02 |
| | | Fe | [211] | 1.1660 | 1.24 | 66.68 | 58.36 | 2.35 | 1.76 | 2.25 | 0.67 |
| | | | | | 1.27 | 67.50 | 58.93 | 2.19 | 1.74 | 2.10 | 0.65 |
| [511] | F | Fe | [220] | 1.0097 | 1.24 | 66.68 | 62.53 | 2.33 | 1.94 | 2.30 | 0.34 |
| | AF | | | | 1.24 | 66.68 | 62.53 | 2.33 | 1.09 | 2.30 | 0.34 |
| | F | Diamond | [311] | 1.0754 | 1.05 | 60.71 | 63.83 | 3.30 | 1.83 | 3.28 | -0.36 |
| [311] | F | PG | [004] | 1.678 | 1.08 | 61.85 | 64.63 | 3.86 | 2.48 | 3.84 | -0.37 |

**Table 1** Example of parameters for Si analyzer, samples, and distances as described in **Fig. 3**. Not shown in **Fig. 3** are $x_D$ and $y_D$ (two last columns), coordinates of Bragg peaks at the PSD.

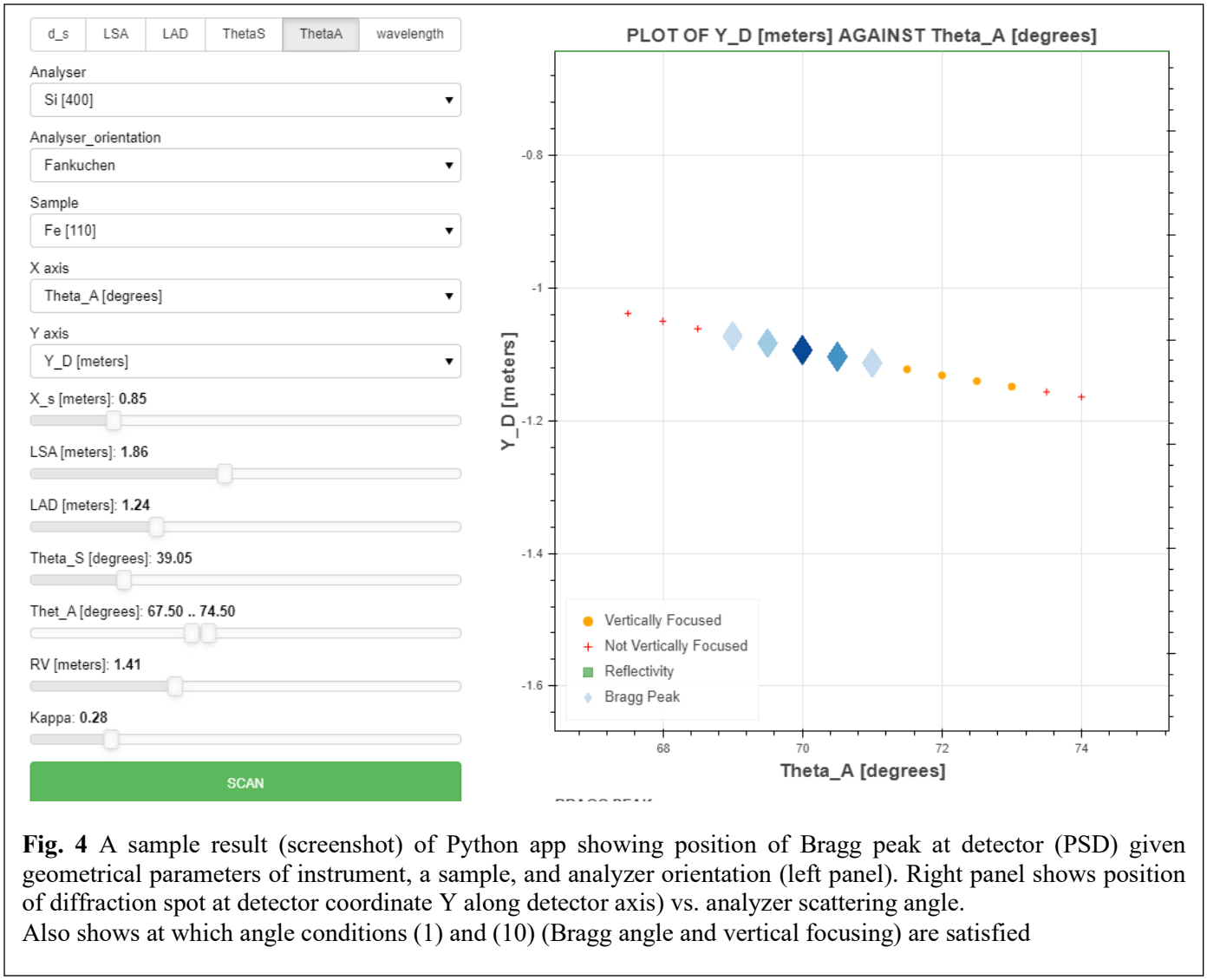


**Fig. 4** A sample result (screenshot) of Python app showing position of Bragg peak at detector (PSD) given geometrical parameters of instrument, a sample, and analyzer orientation (left panel). Right panel shows position of diffraction spot at detector coordinate Y along detector axis) vs. analyzer scattering angle.
Also shows at which angle conditions (1) and (10) (Bragg angle and vertical focusing) are satisfied

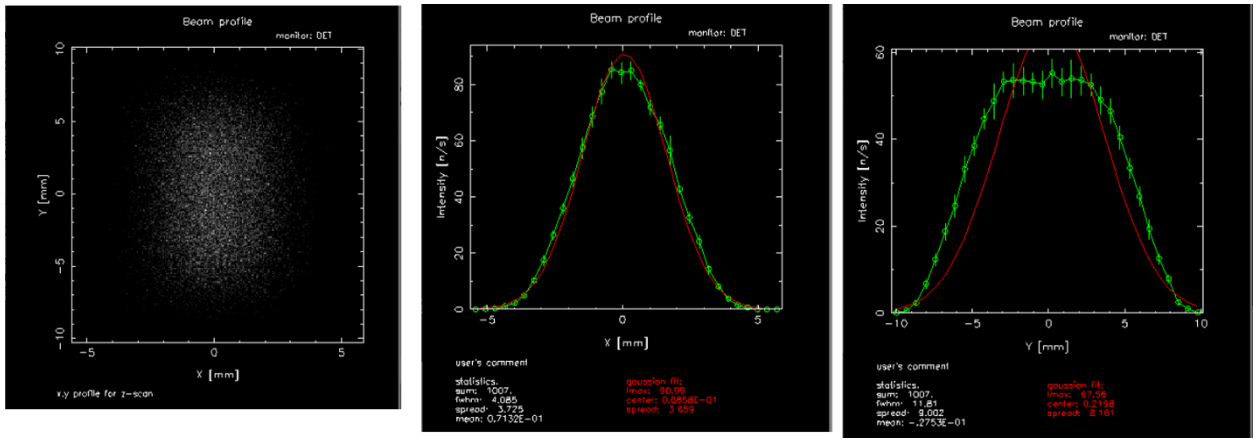


**Fig. 5** Ray-tracing simulation of the diffracted beam profile for Fe [110] sample (dS=2.0269 Å), Si [400] analyzer, LSA=LAD=2.4 m, RH=4.2 m, and RV=1.38 m

## Goal 2. Optimized specifications of focusing analyzers for stress-strain and powder diffractometers.

We conducted ray-tracing simulations using SIMRES, a neutron ray-tracing package used to simulate bent perfect-crystal optics (http://neutron.ujf.cas.cz/restrax) [11]. We have simulated the system that has the polychromatic thermal beam (center wavelength of 1.55 Å). We chose a sample of Fe [110] ($d_S$=2.0269 Å) using Si [400] analyzer; $L_{SA}$=$L_{AD}$=2.4 m, $R_H$=4.2 m, and $R_V$=1.38 m.

**Fig. 5** shows the image of the simulated diffraction spot on the detector.

To demonstrate the concept of dispersive imaging in diffraction, we simulated the diffracted beam while scanning the horizontal curvature $R_H$. **Fig. 6** shows that we achieved the focusing at the predicted $R_H$=4.2 m.

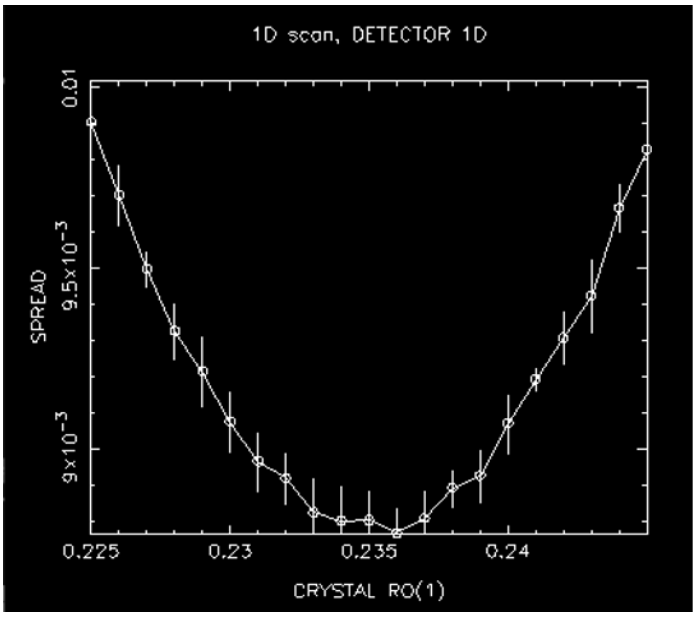


**Fig. 6** Ray-tracing simulations of the diffracted spot size (vertical axis) as a function of the horizontal radius of curvature of the analyzer RH (horizontal axis).

**Goal 3. Feasibility and technology of manufacturing of the analyzers**

To implement the proposed instrument and to build the required double-focusing analyzers we identified the facility, University of Missouri Machine Shop, which has the expertise in manufacturing such multi-wafer analyzers, similar to the existing monochromators at HFIR (HB-2 diffractometer) and NIST, and we identified the supplier to provide properly cut Si crystals. The quotes from the University of Missouri and other potential suppliers (Mirrotron and AVS|US) showed that the University of Missouri is preferred.

**Goal 4. Testing**

A preliminary experimental demonstration has been carried out VULCAN beamline at the Spallation Neutron Source. We have participated in the planning of this demonstration, but we could not participate in the experiment due to pandemic restrictions.

As shown in **Fig. 7**, this experiment demonstrated the diffraction focusing by measuring the (006) reflection of pyrolytic graphite (PG).

By rotating the sample, the analyzer is effectively scanned. The (006) peak remains at the same detector pixel independent of the crystal's angular position because the diffraction focusing (or dispersive imaging in diffraction) is achieved according to Eq. (4).

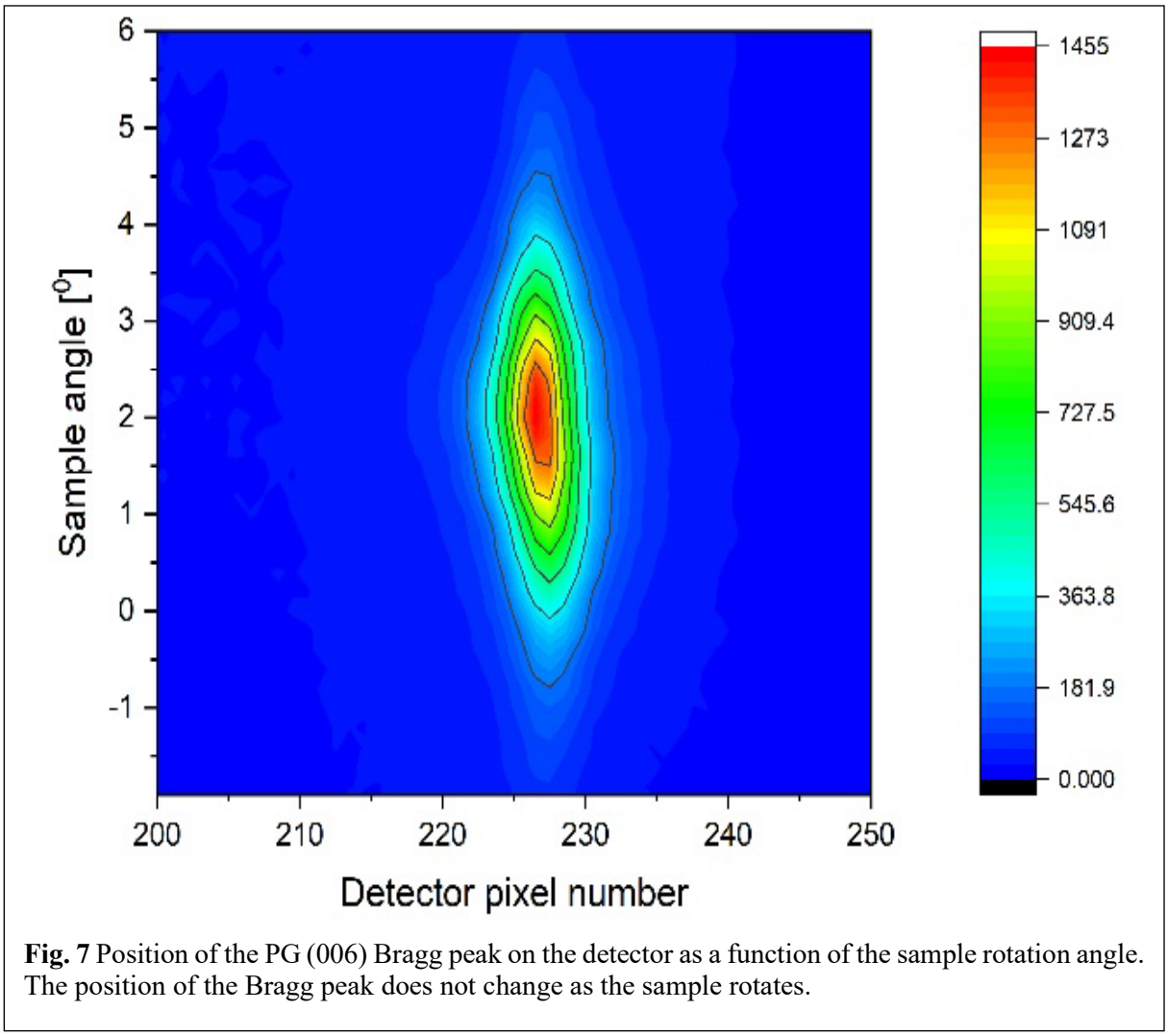


**Fig. 7** Position of the PG (006) Bragg peak on the detector as a function of the sample rotation angle. The position of the Bragg peak does not change as the sample rotates.

The main promise of such analyzers is that several can be combined in one diffractometer to increase the signal rate or to determine different components of a stress tensor. This is the goal of the demonstration during Phase II.

We determined that devices with fixed vertical curvature and variable horizontal curvature (both radii of about 1 m) can be manufactured for the demonstration during Phase-II. The demonstration setup illustrated in Fig. 1 can be built at the MIT Nuclear Reactor Lab (while it cannot be accommodated at VULCAN or other instruments at Oak Ridge or NIST).

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