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(Presented on 17 July 1991)

At the end of February 1991, a "triple-axis" high-resolution diffractometer for on powder sample measurements with synchrotron radiation was put in operation on the Adone wiggler line BX1 at Frascati. The diffractometer is based on a Seifert goniometer, designed according to our specifications. During the project, particular attention was paid in assuring the highest reliability together with great flexibility in the use. In fact, the diffractometer can also be used in a "medium resolution" configuration. For preliminary alignment and data collection, it is possible to operate with a traditional x-ray tube, too. The alignment procedure of the diffractometer to the x-ray beam is very easy. Powder samples can be measured both on the flat holder and on the capillary. An IBM PC computer is used for the instrument actuation and preliminary on-line data collection, while a large software package has been developed for the data analysis performed by a Macintosh IIcx. The instrument performance has been tested with a standard Si sample and quartz and Ni oxide samples. For the two possible resolution configurations, a test on a NiO sample gave FWHM values of 0.16° and 0.04°, respectively, for the [012] peak.

I. INTRODUCTION

According to Bish and Post,[1] the availability of a high intensity and well-collimated x-ray source such as synchrotron radiation has caused the "renaissance" not only in the diffraction as a whole, but particularly in powder diffraction. Pioneer works in this field are due to Thompson,[2] Hastings,[3] and Parrish,[4] among others. At the end of 1988, in order to increase the impact of synchrotron radiation on crystallography, a joint program was started in Europe between various Italian, French, and Spanish scientific institutions, under the supervision of Professor Marezio of the CNRS, Laboratoire de Cristallographie (Grenoble).[5] The aim was to set up facilities for x-ray diffraction near the laboratories of Frascati and Orsay. Lauriat and coworkers have put a double-crystal monochromator and a single-crystal diffractometer in operation at LURE. In Frascati, the PWA Group has set up a high-resolution powder diffraction station. After only two years, at the end of February 1991, the high-resolution powder diffraction station was connected to the Adone wiggler magnet line BX1. At the beginning of March, the apparatus was successfully tested by using NBS Si standard powder;[6] while in June, a first set of measurements was made on quartz and Ni oxide specimens.

II. THE HIGH-RESOLUTION POWDER DIFFRACTOMETER

As Adone is not a dedicated machine, only few months per year are available for doing experiments with synchrotron radiation in several research areas. Thus, we have expended considerable effort in setting up a powder diffraction station with the maximum possible reliability, as well as flexibility in use, in order to optimize the time available. Indeed, the diffractometer can operate in two different configurations, (a) high resolution and (b) medium resolution, both of them with synchrotron radiation or with x-ray tube sources. By "high resolution" we mean a value in the full width half-maximum (FWHM) of the diffraction peaks one order of magnitude less than that obtained with a conventional x-ray tube. By "medium resolution" we mean a value in the FWHM equal to or less than that obtainable with an x-ray tube after having applied the Rachinger correction.

In the book *Modern Powder Diffraction*,[1] Finger[7] gives a short review of the different powder diffractometer configurations adopted to enhance the resolution of diffraction spectra by using synchrotron radiation. For our high-resolution powder diffraction station, we have adopted the so-called "triple-axis configuration," similar to that developed at CHESS[3] and at SSRL.[4] In our arrangement, the first axis is given by a Si [111] channel-cut monochromator which allows operation in the range 0.5–3 Å with a resolution $\Delta E/E$ of the order of 10^{-4}. The second axis is formed by the sample mounted on the goniometer Θ axis. The third axis refers to the Ge [111] flat single-crystal analyzer mounted on the $2\,\Theta$ diffractometer arm. The high spectral resolution is essentially due to the quality of this crystal analyzer, i.e., the smaller the FWHM of its rocking curve, the greater the resolution.

Our diffractometer is based on a special version of the Seifert MZ-VI goniometer[8] constructed according to our

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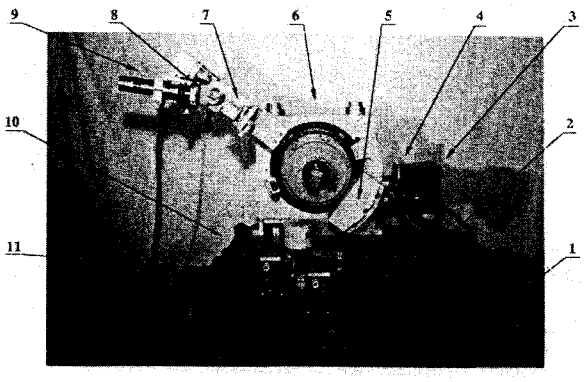

FIG. 1. General view of the high-resolution powder diffractometer. 1: Motor for the longitudinal motion; 2: rotation axis; 3: input slit system; 4: intensity beam monitor; 5: counterweight; 6: Seifert MZ-VI goniometer; 7: output slit system; 8: crystal analyzer and one-axis goniometer; 9: signal detector; 10: supporting table; and 11: motors for the transverse motion and for the rotational motion.

specifications. This goniometer, which is characterized by very good mechanical quality and by the use of microstep technology in the stepper motor actuation, assures an angular resolution better than 0.001°. The diffractometer consists mainly of three parts: (i) a removable primary slit system; (ii) the $\Theta/2\Theta$ goniometer MZ-VI; and (iii) the 2Θ arm with the crystal analyzer (Fig. 1).

The primary slit system consists of the following: (1) A horizontal slit driven by a stepper motor. As the slit width, controlled via software, can vary continuously in the range 0–6 mm, it is possible to program a constant area irradiation of the sample. (2) A manually controlled vertical slit working in the range 0–10 mm. (3) A thin, scattering, 45° sheet of mylar or kapton and a scintillation detector to monitor the incoming x-ray radiation.

The MZ-VI goniometer, vertically placed, supports the 2Θ arm, which is 343.8 mm in length, thus permitting a recording scale of $1° = 6$ mm. A heavy counterweight, placed symmetrically with respect to the 2Θ arm, assures the best mechanical balance and increases the reproducibility in the positioning of the whole $\Theta/2\Theta$ circles. Either a flat sample holder or a goniometer head with capillary can be mounted on the Θ circle. By removing a circular cover disk, a central opening (135 mm in diameter) allows special holders, such as cryostats or high-temperature chambers, to be placed on the Θ circle.

The 2Θ arm supports:

(1) A secondary slit system, formed of a set of removable input/output slits, plus a Soller to be used only for operation with x-ray tubes.

(2) A goniometer head, which holds the crystal analyzer in the $(n, -n)$ setting mode with respect to the sample, standing on a small one-axis goniometer consisting of a stepper motor connected to a 1/20 gear reduction system. This configuration allows an angular resolution of 0.0014° in crystal positioning. Following Hastings,[3] we have chosen a flat Ge[111] crystal analyzer as a compromise between the higher resolution obtainable with a Si[111] crystal or the higher integrated intensity offered by a LiF[220] crystal. The rocking curve of our crystal, as shown in Fig. 2, has a FWHM value of 0.006°, according to our measurements with synchrotron radiation. Thus, the crystal analyzer acts as a narrow receiving slit of about 21 arcsec at 1.54 Å.

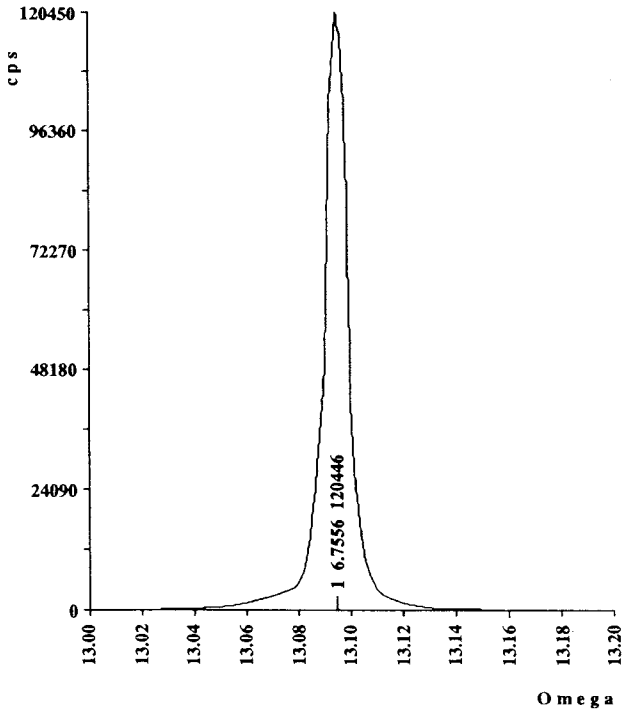

FIG. 2. Rocking curve of the Ge[111] crystal analyzer. FWHM = 0.006°; lambda 1.54 Å; step 0.002°; time 1 s.

(3) A signal scintillation detector, with a manually controlled vertical input slit, standing on a small graduated arm.

In order to work at medium resolution, it suffices to remove the crystal analyzer mounted on the goniometer head and place the signal scintillation detector in the 2Θ arm direction. The operator takes advantage of this configuration, which is characterized by a higher intensity, by using only the resolution of the channel-cut monochromator $\Delta E/E = 1.4 \times 10^{-4}$. In addition, the use of synchrotron radiation means that all the problems connected with the presence of the $K\alpha_1/K\alpha_2$ doublet are avoided.

When Adone is not available for synchrotron radiation experiments, an x-ray tube can also be put into operation for preliminary alignment and data collection. The primary slit system must be removed, while the 2Θ arm, supporting the secondary slit system and the signal detector, can be put into operation at the right-hand side of the goniometer. An x-ray tube can be located vertically on the left-hand side by means of a special stand, as shown in Fig. 3. In this configuration it is possible to work either at "medium" or at high resolution. In the latter case, it is sufficient to change the flat analyzer crystal with a curved analyzer.

An IBM PC controls all the goniometer movements and the complete data collection. The experimental data processing is managed by a Macintosh IIcx through a special large software package named "Mac Dust," which is continuously updated.[6]

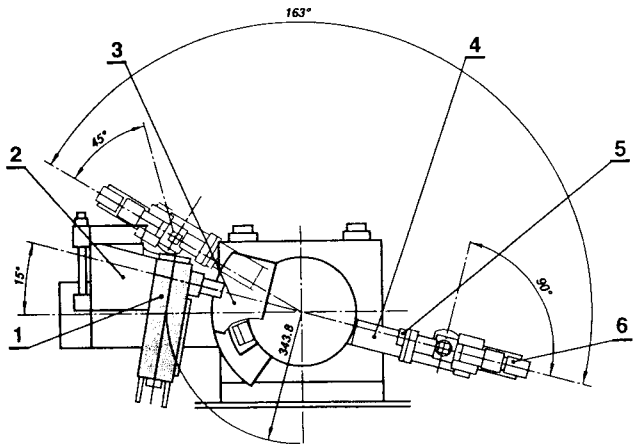

FIG. 3. The powder diffractometer with an x-ray tube mounted on the left-hand side. 1: x-ray tube; 2: adjustable support for tube housing; 3: counterweight; 4: 2Θ arm; 5: input/output slit system with Soller; 6: signal detector.

III. THE SUPPORTING TABLE AND THE DIFFRACTOMETER ALIGNMENT

Figure 4 shows the special supporting table[9] that has been built in order to guarantee six degrees of freedom in positioning the diffractometer with respect to the synchrotron radiation beam, which is 10-mm wide and 2-mm high. The following movements are allowed: longitudinal and transverse motion with respect to the beam direction; up-down tilting on three points plus a rotation of all the diffractometer around an axis that passes across the geometrical center of the horizontal input slit. The movements of the table platforms are performed by means of ac motors, with a 1/80 gear reduction system coupled with relative encoders, under the remote control of a programmable logic control unit,[10] which is separate from the goniometer

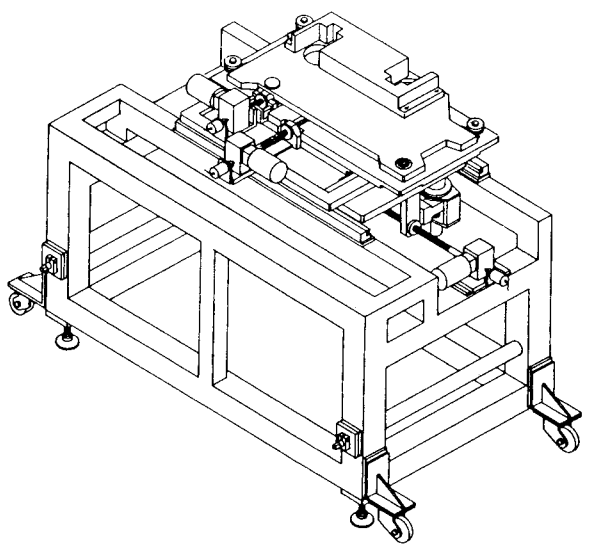

FIG. 4. Drawing of the supporting table which has six degrees of freedom for the alignment of the diffractometer with respect to the synchrotron radiation beam.

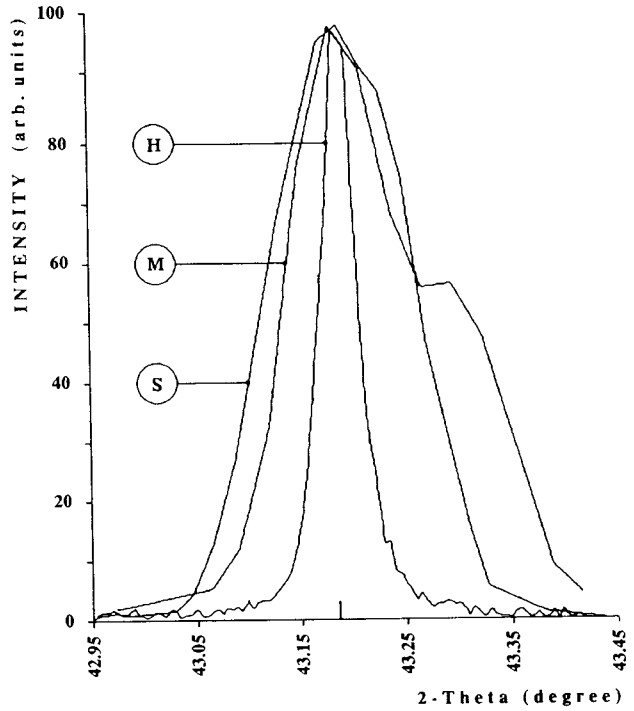

FIG. 5. The diffractometer resolution test. The three overlapped NiO[012] peaks were measured at high (H) and medium resolution (M) with synchrotron radiation. The profile (S) was measured with a Cu tube. The FWHM values are 0.039°, 0.156°, and 0.188°, respectively.

and data acquisition electronics. The positions of the encoders are displayed by counters with nonvolatile memory.[11] Since the diffractometer has been prealigned in the factory, the station alignment procedure and control are quite simple. By using a yellow fluorescent screen with reference marks, located on the sample holder, it is only necessary to assure perfect parallelism between a narrow image of the source, which passes across the input slit, and the central mark on the screen, which is remotely viewed by a CCTV. Then, fast movements around the theta axis ensure that no change in position occurs between the central mark and the source image.

IV. FIRST EXPERIMENTAL RESULTS

At the end of February 1991, the powder diffraction station was connected to the wiggler magnet beam line BX1. At the beginning of March, the apparatus was successfully tested. We first evaluated the FWHM value of the rocking curve of Ge [111] crystal, since the instrumental resolution depends on the quality of the crystal analyzer. Then, by using a NBS Si standard, several diffraction peaks were collected in high resolution, e.g., the peak [111] gave a FWHM of 0.014° against the value of 0.127° for the same peak collected with a Cu tube.[6]

In June a first set of measurements was made on quartz and Ni oxide samples in order to define the best collecting condition. In order to give a global view of the resolution achievable with our diffractometer, Fig. 5 shows three overlapped profiles of the NiO peak [012], $2\Theta = 43.186°$. The curve (S), which was collected using an x-ray Cu

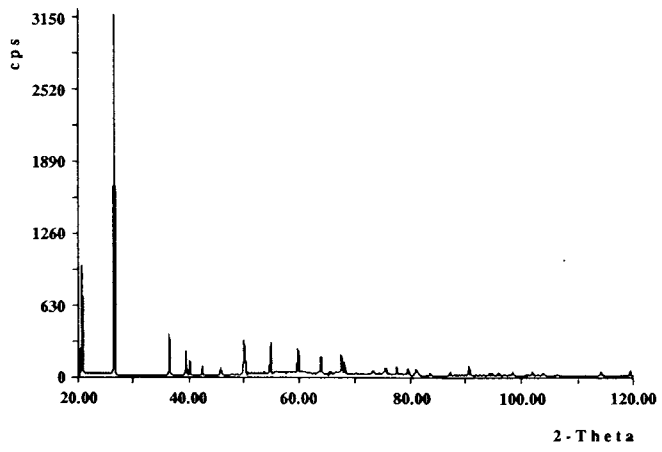

FIG. 6. The quartz powder diffraction spectrum collected at medium resolution during a 5-h run. The intensities are not according to the standard values because no correction was made on the data.

tube, gives a FWHM = 0.188°; the curve (M), collected in the medium resolution configuration, gives a value of 0.156°; while the last curve (H), collected with the highest resolution, gives the smallest FWHM value of 0.039°.

Figure 6 shows, without any smoothing or background correction, the quartz diffraction spectrum collected at medium resolution during a 5-h Adone run. The sharp spectrum points out the very good signal/noise ratio achievable during instrument operation. The intensities are not according to the standard values because no correction was made, e.g., (a) for the decrease in synchrotron radiation light during the run; (b) for the polarization of the incident beam; and (c) for the air absorption, etc.[12]

Finally, Fig. 7, again collected at medium resolution, shows how it is possible to increase the resolution itself by decreasing the width of the primary and secondary slit system for the couple of peaks [116] and [122]. The smallest slit combination, again without any data manipulation, gave $2\Theta = 111.007°$ and $2\Theta = 111.133°$, respectively, for the peak positions, with a separation of 0.0126°. A best fit, e.g., with a pseudo-Voigt profile,[7] would probably give angular positions close to the standard values 111.032° and 111.175°, respectively, with a separation of 0.143°.

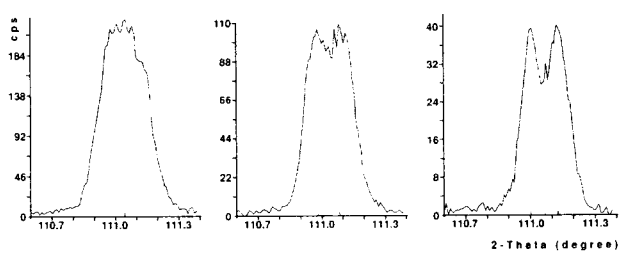

FIG. 7. Resolution vs width for different combinations of the primary and secondary slit systems. NiO peaks [116] and [122]; lambda 1.54 Å; step 0.01°. From left to right: time 5 s, I/O slit 0.6–0.5 mm; time 8 s, I/O slit 0.4–0.5 mm; and time 12 s, I/O slit 0.4–0.3 mm.

V. CONCLUSIONS

We have presented a "triple-axis" powder diffractometer that can be used in two possible configurations: medium or high resolution. Both of them take advantage of the use of synchrotron radiation as a highly collimated and intense x-ray source. The quality of the medium resolution spectra is assured by the "channel-cut" monochromator, which is on the BX1 Adone wiggler line, while the quality of the high resolution spectra is enhanced by the flat Ge crystal analyzer, placed on the 2Θ arm, which acts as a 21-arcsec narrow slit. For preliminarily alignment and data collection, it is also possible to operate with a traditional x-ray tube for both the above-mentioned configurations. The successful experimental tests on a Si standard sample, followed by measurements on quartz and Ni oxide samples prove that the final aim of realizing a sophisticated, multiform apparatus has been reached. Extensive experimental activity is now in progress compatibly with the running time dedicated to synchrotron radiation experiments at Adone.

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