JUSIFA—A new user-dedicated ASAXS beamline for materials science

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The fully computer-controlled user-dedicated small-angle scattering facility JUSIFA was set up for general use at the DORIS Synchrotron Radiation Source in DESY (Hamburg). This beamline is especially designed for anomalous scattering studies of anisotropic samples in materials science and enables ASAXS studies to be undertaken on the length scale 0.5 to 100 nm in an energy range of 4.5 to 35 keV.

INTRODUCTION

Small-angle x-ray scattering (SAXS) is widely applied in materials science to study variations of density or chemical composition over distances above 0.5 nm; in the past SAXS on in-house instruments had become a classical method with characteristic fixed-energy radiation from x-ray tubes. The continuous synchrotron radiation from storage rings, however, offers an exciting new opportunity: tuning the incident photon energy close to an absorption edge of a selected element results in a strong anomalous scattering which changes its atomic scattering amplitude typically by about 20%. This results in variations in the scattering intensity which probe its distribution in the sample.

In an anomalous small-angle scattering experiment (ASAXS) both the strength and nature of the varied scattering contrast can be examined, independently of all other unknown parameters. In many cases, this allows us to retrieve more detailed structural information from the SAXS or to reduce a larger number of structural models in the interpretation of classical fixed energy SAXS measurements. ASAXS thus opens the way for a very reliable and more precise structural analysis. Moreover, the possible enhancement of scattering contrasts in ASAXS experiments, for instance between neighboring elements in the periodic table, becomes especially important for studies of many technically relevant microstructures, e.g., precipitates in Fe-Cr-Ni-Ti alloys.

To apply this technique for general use in materials science, it has to be made available to a community the large majority of whom are inexperienced users in synchrotron radiation experiments. The emphasis of Jülich's user-dedicated small-angle scattering facility (JUSIFA),[1] at the DORIS synchrotron radiation source in DESY (Hamburg), was therefore to provide a user-friendly fully computer-controlled ASAXS beamline specifically designed for that purpose, which can be easily operated in a routine manner by the nonspecialist.

I. DESIGN PARAMETERS

ASAXS makes use of the rapid variation of atomic scattering amplitudes (form factor) for x-ray energies close to absorption edges. In JUSIFA experiments the accessible energy range is

$$4.5\ \text{keV} \leqslant E \leqslant 35\ \text{keV}. \quad (1)$$

This allows the K edges of elements with atomic numbers between $Z = 21$ (Sc) and $Z = 55$ (Cs) to be reached and covers the L III-edges for atomic numbers larger than 55. A

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further accessibility of the K edges for the low-Z materials would require lower energies than 4.5 keV. The limit of $E > 4.5$ keV rather than smaller values may be justified due to the rising absorption in vacuum windows and increasing detector problems in this latter regime with regard to the fact that increased absorption in most materials science samples would require measurements with very thin samples, which often are not easy to prepare.

In order to take advantage of the maximum change of the atomic scattering amplitudes in the immediate vicinity of absorption edges and to avoid background problems with intense fluorescence radiation on their low energy side, the JUSIFA monochromator delivers a very narrow and stable energy bandpass,

$$\Delta E/E \leqslant 2\times 10^{-4}, \tag{2}$$

with suppressed higher harmonics. In addition to ASAXS experiments this energy resolution enables measurements of x-ray absorption for those samples, especially in the near-edge region (XANES and EXAFS), which may be used to obtain experimentally determined values for the atomic scattering amplitudes applying both the optical theorem and the Kramers–Kronig relation.

Materials science samples are often not isotropic. To cope with anisotropic scattering patterns, JUSIFA measurements are performed in pinhole collimation with a two-dimensional position-sensitive detector, a multiwire proportional counter (MWPC) with 256×256 resolution elements (pixels) and a 200×200 mm^2 active area.[2] To cover an extended region in reciprocal space, these samples, with typical size $\leqslant 1\times 1$ mm,[2] are tilted relative to the incoming beam around two perpendicular axes in a diffractometerlike rotation stage.

In materials science, typical SAXS studies cover a range in real-space resolution on the length scale,

$$D = 1/Q \approx 0.5,\ldots,100 \text{ nm}, \tag{3}$$

which requires accessible scattering vectors

$$Q = (4\pi/\lambda)\sin \epsilon/2$$

(ϵ = scattering angle, λ = x-ray wavelength). In JUSIFA, we are using the most straightforward scattering geometry, the nonfocusing pinhole collimation system of Fig. 1. As best resolution one obtains

$$D_{\max} \approx 1/Q_{\min} \approx 1/\Delta Q \approx 100 \text{ nm}, \tag{4}$$

which can be achieved for 4.5-keV x-ray energy at the maximum sample-detector distance of 3.3 m, the proper setting for studies of extended structures. To cover scattering patterns on a smaller length scale with the 200×200 mm^2 active detector area, the detector distance can be shortened in five steps down to 0.9 m under computer control or to a minimum of 0.6 m manually. In this way, a sufficiently large region in reciprocal space becomes accessible.

The mechanical design was governed by the requirement to eliminate as much parasitic background as possible: any parasitic air scattering is avoided by measurements under high-vacuum conditions, whereas the storage ring x-ray background was shielded using a large 160-mm vertical displacement of the monochromatized primary beam. For great flexibility in the choice of custom-designed equipment as, e.g., special ovens for *in situ* low or high temperature measurements or special cells for liquid samples, a sufficiently large sample chamber is supplied.

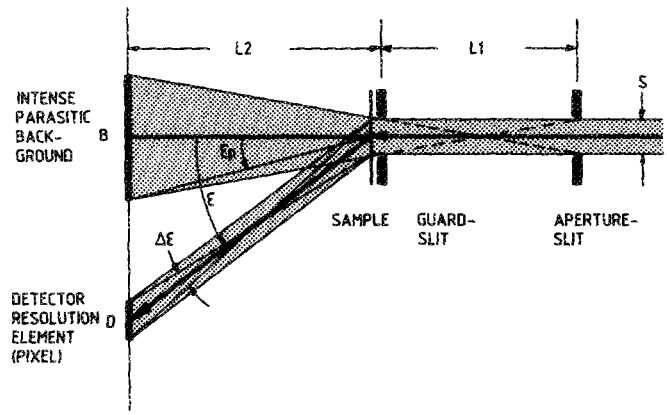

FIG. 1. Optical parameters of the JUSIFA scattering geometry with nonfocusing pinhole collimation. From $B = 2S\,(L2/L1 + 0.5)$, one gets with $L1 \geqslant L2/1.5$ and $L2 = 0.6,\ldots,3.3$ m for $E \geqslant 4.5$ keV: $Q_{\max} \geqslant 3$ nm^{-1}, $Q_{\min} \approx 1/\Delta Q \geqslant 10^{-2}$ nm^{-1}.

II. PRINCIPAL LAYOUT

The principal layout of the JUSIFA beamline is shown in Fig. 2. Three computer-controlled slit systems collimate the primary beam from a bending magnet to typical sample sizes between 0.1×0.1 and 1×1 mm^2. The monochromator consists of two symmetrically cut perfect Si 311 crystals, which are held on two separate rotation stages. If desired they can be replaced by prealigned Si 111 or Si 511 crystals, respectively. For the suppression of higher harmonics, the second crystal is detuned by a fraction of its Darwin width out of the maximum reflectivity via a MOSTAB[3] controlled piezodrive. The reflectivity is monitored by the scattering from Kapton foils via two NaI-scintillation detectors. In the same way, the primary beam intensity is monitored by a third detector behind the beam-size defining second slit.

Energy tuning is performed fully under computer control by two independent crystal rotations (monitored with an accuracy of 10^{-3} degrees by Heidenhain ROD-800 encoders on the two rotation axes) and a horizontal translation of the second crystal for a fixed-exit operation.

Two beam height monitors, large-area Hamamatsu S 1723-06 1-cm^2 photodiodes which are coupled to scintillation screens via glass prisms, are used to control the positions of both the incoming and outgoing beam with an accuracy of ± 2 μm. Beam height variations of the DORIS beam are corrected by a vertical translation of the first rotation stage, and exit beam displacements via additional horizontal translations of the second crystal.

Small angle scattering intensities are measured by a two-dimensional position-sensitive multiwire proportional counter.[2] For ease of measurement, up to ten samples with typical sizes $\leqslant 1\times 1$ mm^2 are mounted on preadjusted standardized multispecimen holders and positioned under computer control. This allows us to run time-saving measurement programs, for instance with a series of differently

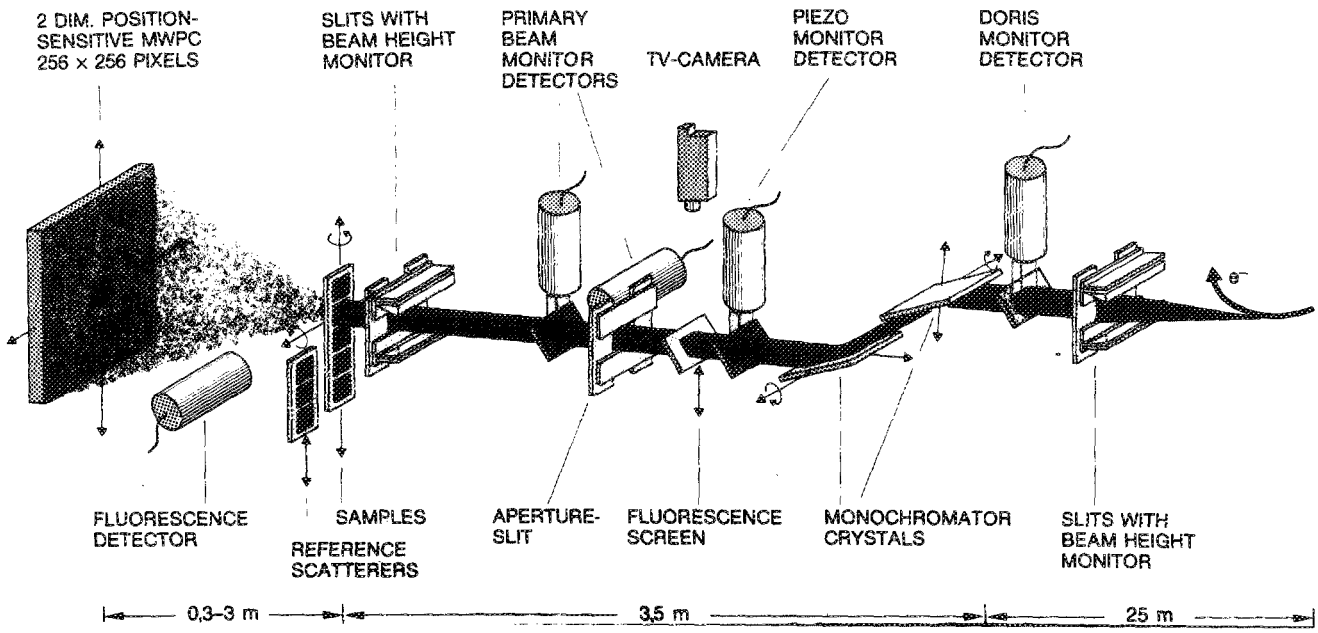

FIG. 2. Principal layout of the JUSIFA ASAXS beamline.

prepared samples. The sample changer stage is an integral part of a sample rotation stage. Its two rotations around perpendicular axes are used to study anisotropic structures under various view angles. At a second sample changer stage a set of reference samples can be brought into the primary beam for calibration purposes and for determinations of sample absorptions as intensity ratios of incoming and transmitted primary beams: Whereas direct primary beam measurements would overload the 2-D detector (this is prevented by a beam stop), smaller fractions are measured as total SAXS intensities from an additional scatterer, e.g., porous glassy carbon. For absorption measurements it is brought into the primary beam on the reference sample changer stage. The ratio of the detector anode counts with and without the sample in the beam is then monitored as a measure of the sample absorption. The onset of fluorescence radiation and resonant Raman scattering is monitored by a high-resolution Ge detector.

III. MECHANICAL DESIGN

The schematic view of the beamline is given in Fig. 3. As shown, the complete front flange of the monochromator box can be lowered for service work. During operation it is closed electropneumatically and the beam path is entirely evacuated to avoid air absorption and parasitic scattering. Three independent vacuum sections: monochromator box, sample chamber, and the back-end beamline are separated

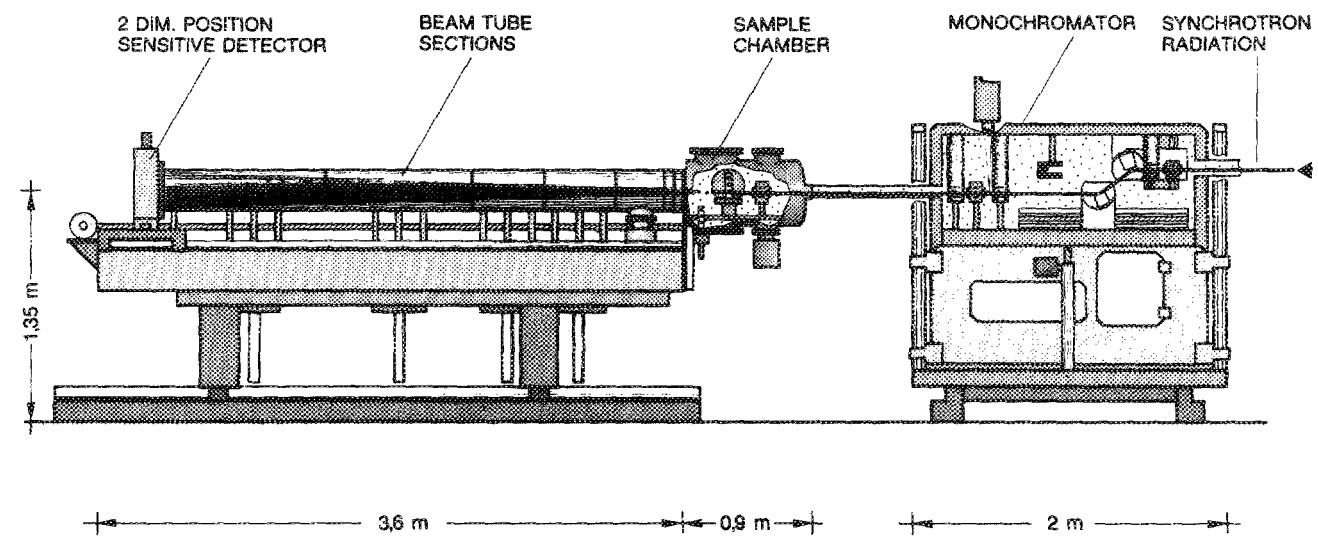

FIG. 3. Schematic view of the JUSIFA beamline: Monochromator box, optical bench with sample chamber, and two-dimensional position-sensitive detector.

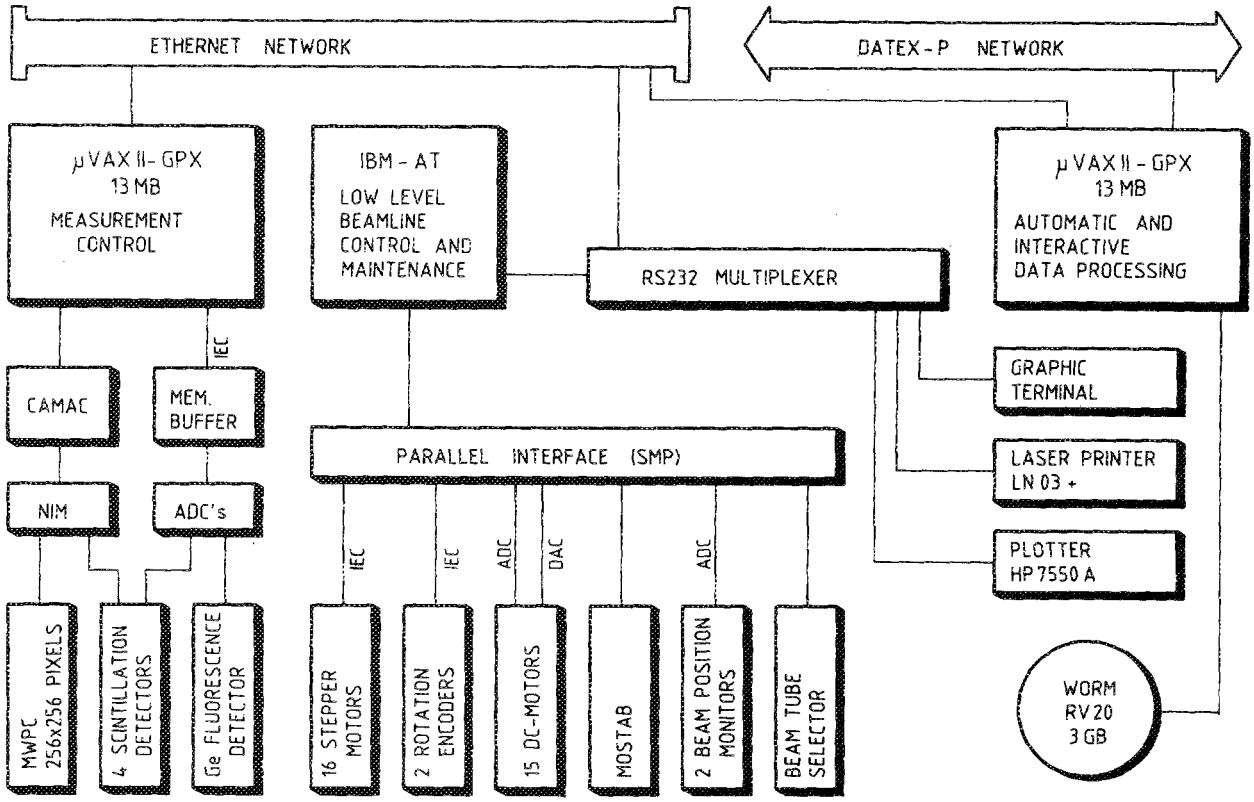

FIG. 4. JUSIFA data-acquisition and computer hardware.

by 25-μm Kapton windows (monochromator box) and a computer-controlled in-line valve (located between the sample chamber and the downstream beam tube section). They are pumped down by three 360 ℓs^{-1} turbomolecular pumps (roughing pumps 25–40 $m^3 h^{-1}$) to some 10^{-5} Torr.

The complete mechanical part was manufactured as a custom design by Huber Diffractionstechnik GmbH,[4] mainly by using vacuum compatible commercial equipment.

The optical bench is shown with the detector in its outmost position for studies of larger structures. For examinations on a finer scale, the sample-detector distance can be reduced. In order to shorten the beam tube length between the sample chamber and the back-end x-ray window, the beam tube is composed of four removable smaller tube sections, each of which can be moved out electropneumatically down into the optical bench. The full procedure: closing of the sample chamber by the in-line valve, vacuum breaking in the beam tube, selection and positioning of the required beam tube sections, movement of detector and x-ray window, vacuum pumping in the shortened beam tube section, and reconnecting of the sample chamber is completely under computer control and initiated either manually or by the software running the experiment.

IV. DATA ACQUISITION, COMPUTER SYSTEM, AND SOFTWARE

An overview of the data-acquisition and computer system is given in Fig. 4. The setup of x-ray energy, sample-detector distance, as well as the change and orientation of samples and internal references is controlled by a personal computer which itself is governed by programs running on two μ-VAX II GPX work stations. The user-friendly software allows us to run a preselected program of measurements from several samples, internal references and scattering backgrounds at x-ray energies near absorption edges and to perform automatic or interactive data reduction to the structural parameters with the possibility of an optional remote log-in via the Datex-P network. For later use all data are permanently stored on 2 GB optical WORM disks.

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[4]Huber Diffraktionstechnik GmbH, D-8219 Rimsting, West Germany.