DEUTSCHES ELEKTRONEN-SYNCHROTRON DESY

DESY SR-77/19
November 1977

A Spectrometer for the Investigation of Ultra-Soft X-Ray Emission Spectra Excited with Synchrotron Radiation

by

N. Kosuch, E. Tegeler, G. Wiech, and A. Faessler
Sektion Physik der Universität München

NOTKESTRASSE 85 · 2 HAMBURG 52

To be sure that your preprints are promptly included in the
HIGH ENERGY PHYSICS INDEX ,
send them to the following address (if possible by air mail) :

DESY
Bibliothek
Notkestrasse 85
2 Hamburg 52
Germany

A Spectrometer for the Investigation of Ultra-Soft X-ray Emission Spectra Excited with Synchrotron Radiation

N. Kosuch, E. Tegeler, G. Wiech, and A. Faessler
Sektion Physik der Universität München, 8000 München 22,
West Germany

Abstract

An instrument has been developed for the investigation of ultra-soft x-ray emission spectra which are excited in fluorescence using the synchrotron radiation of the storage ring DORIS at Hamburg.
The 2m grazing incidence concave grating spectrometer and the experimental layout are described.

A. Introduction

X-ray emission spectra provide valuable information for molecular orbital and solid-state-band analysis. For the initial ionisation both electron impact (primary excitation) and irradiation with photons (fluorescence or secondary excitation) are applied.

As is well known, many compounds are chemically changed or decomposed by electron bombardement, contrary to fluorescence excitation which is frequently used in modern x-ray spectroscopy. However, in the region of ultra-soft x-rays (λ>20 Å) it is difficult to obtain sufficient intensities with fluorescence excitation, as long as conventional sources are used. Only a very few measurements of this kind have been carried out so far (1)(2). New possibilities are offered by utilizing the continuous radiation of electron synchrotrons, and especially of storage rings, which are superior in intensity to any conventional source. Additionally, the radiation of storage rings is free of short-time intensity fluctuations.

Some years ago we started the experiment FLEUR (Fluorescence Excitation of Emission Spectra in the Region of Ultra-soft Röntgenrays) using the synchrotron radiation of the Deutsches Elektronen-Synchrotron DESY at Hamburg for fluorescence excitation of ultra-soft x-ray spectra. Much better results were obtained after the spectrometer was operated at the storage ring DORIS and the experimental set up has been gradually improved in many respects. In the following, the present state of the experiment FLEUR is described.

B. The experiment FLEUR

The experiment FLEUR (Fig. 1) is positioned at a distance of about 40 m from the source point.

The fluorescence radiation is analysed by a 2 m concave grating spectrometer housed in a HV tank. The samples to be studied are mounted on special holders in the sample chambers; they are excited by the undispersed synchrotron radiation. The characteristic radiation of the samples passes through a differential pumping stage and then enters the spectrometer. The sample chamber, mounted inside the spectrometer tank, and the mirror case, located between the sample chamber and the ion getter pump (GP 2), are operated at UHV. Details of the experimental set up, designed according to the special requirements of operation at the storage ring, are described in the following sections.

a) Vacuum system

The ion getter pump, GP 1, and the filter wheel, FR, work as a pressure stage. The wheel has several openings covered with thin foils. Since the wheel is pressed against a metal seal a difference in pressure of 2 -- 3 orders of magnitude e.g. from 10^{-6} to 10^{-9} torr can be maintained. By an appropriate choice of the foil material the long wavelength radiation can be reduced in intensity. Two windows opposite each other allow one to observe whether the foils are undamaged.

The ion getter pump, GP 2, maintains a pressure of about 10^{-9} torr in the sample chamber, P, and in the mirror case. With the help of the cryopump, KP, the vacuum can be further improved by one order of magnitude.

The spectrometer tank (10^{-6}torr) is evacuated by a turbo molecular pump, TVP, and the ion getter pump, GP 3.

b) Sample chamber

The sample chamber contains different holders for the samples according to the particular experimental requirements, and an evaporation source.

For comparatively stable samples as SiO_2, BN or $CaCO_3$, two wheel-shaped holders, R, rotable around a horizontal axis are used. Each wheel permits the mounting of several samples suitable for any take-off angle of the fluorescence radiation. A vertically mounted second holder, PS, (for 12 samples) permits a continous change of the take-off angle, e.g. for studies of the anisotropic emission. The temperature of a third holder to be used at a fixed take-off angle can be varied from liquid nitrogen temperature up to more than 300°C.

For precise adjustment, the samples can be observed through a window. The visible fluorescence light emitted by the samples when exposed to the synchrotron radiation indicates the stability of the substances or the progress of their decomposition.

The characteristic radiation of the samples passes the diffusion channel, DS, (height 12 mm, depth 8 mm, variable width) in the wall of the sample chamber and then enters the separately mounted entrance slit, ES, of the spectrometer. By this diffusion channel, a difference of pressure of several orders of magnitude is maintained. Normally, a width of 400 µ is used.

The special construction of the sample chamber guarantees that, in case of thermal expansion, the diffusion channel moves only in the direction towards the entrance slit and not parallel to it.

c) Focusing mirror

The focusing mirror is a thin metal coated glass plate (360 x 35 mm^2), mounted in a specially profiled frame and bent to a cylindrical form. The focal distance is variable. By turning the mirror around a vertical axis the angle of incidence can be varied. The width of the focal line on the sample (2 - 3 mm) is matched to the acceptance of the spectrometer. To avoid local overloading of the samples it should not be narrower.

With the focusing mirror, a gain of intensity of more than a factor two is obtained in the energy range of 200 eV to 500 eV. At lower energies, a greater gain is to be expected, because a larger section of the beam of the synchrotron radiation can be utilized due to an increasing of the critical angle of total reflection. - By choosing an appropriate reflection angle it is possible to cut off the short wavelength radiation.

The focusing mirror and the sample holders can be removed from the beam without breaking the vacuum in order to clear the light path for experiments positioned behind FLEUR.

d) Spectrometer

The 2 m grazing incidence concave grating spectrometer was constructed by Feser (3).It was designed for energies from about 30 eV to 600 eV.

Part of the Rowland circle functions as a guide track carrying the entrance slit, ES, the grating holder, G, the exit slit, AS, and the detector, M (Fig. 1). The exit slit and the detector are mounted on a carriage which is moved along the Rowland circle by means of a lead screw, SP, driven by a step motor, Mo. The plane of the exit slit is always

adjusted perpendicular to the radiation. The position of the exit slit is registrated by a mechanical counter coupled directly with the lead screw. Fig. 2 shows a photograph of the spectrometer unit.

Additional facilities for external adjustment of the detector (4) and the entrance slit, for variation of the slit width, and for radial movement of the grating are installed.

The detector is an open parallel-plate multiplier. It has been developed in our laboratory for the special requirements of a slit shaped optical mapping (5).

e) Electronics

The spectra are recorded automatically in the step-scanning mode. The measuring process is shown in the schematic diagram of fig. 3.

During the course of the measurement the number of exciting photons per step is automatically kept constant. Since the beam current and consequently the intensity of the synchrotron radiation decrease exponentially with time, the time per step has to be increased accordingly. For this purpose a frequency proportional to the beam current is fed into the preset scaler.

The number of photons detected and the position of the exit slit are read into the spectrum buffer. All information stored there is simultaneously shown on a monitor. After the spectrum has been measured, the contents of the spectrum buffer are transferred to the disk storage of the PDP 11/45 computer. For further evaluation of the individual spectra, special programs are available for the PDP 11/45 - IBM 360 set up.

C. Examples of measurements

So far mainly the K-emission spectra of the second period elements lithium (228 A or 54 eV) to oxygen (24 A or 525 eV) were studied.

In the initial experiments at the electron synchrotron the K-fluorescence yield of the elements Li, Be, B, and C was determined from the integral intensities of their K-emission bands and the known absolute intensity of the synchrotron radiation (6).

By measuring the intensity distribution as a function of the take-off angle, the anisotropic emission of the C K-emission band of graphite (7) and of the N K-emission band of hexagonal BN (8) was studied.
Detailed investigations have been made on the electronic structure of the occupied states of numerous oxyanions. Among others, the K-emission spectra of nitrogen and oxygen in NO_2^- and NO_3^-, and of carbon and oxygen in CO_3^{2-} were measured (9). Another study was made of the PO_4^{3-}-ion (10).

For the last mentioned spectra, which ranged from energies of about 200 eV to 500 eV, a gold coated grating, blazed at 1° with 2400 lines/mm was used. At storage ring energies of 2 GeV and a beam current of 150 mA, typical counting rates are 10^3 counts/min obtained for the maxima of the oxygen, nitrogen, and carbon K-spectra for $LiNO_3$ and Li_2CO_3 at a resolution of 0.5 eV. Typical counting times per step lie between 400 mAs and 8000 mAs (i.e. between about 3 s and 60 s) dependent on the stability of the samples and the intensity of their characteristic radiation.

Recently the measurements were extended to organic compounds. Attempts to obtain the K-spectra of carbon in condensed benzene and in naphtalene have been successful.

Acknowledgements

This work was supported by the Deutsche Forschungsgemeinschaft and the Bundesministerium für Forschung und Technologie. The use of the facilities of the Deutsches Elektronensynchrotron DESY and the help of staff members are gratefully acknowledged.

References

(1) A.S. Koster, J.Chem.Phys.Sol. 32, 2685 (1971)

(2) B.L.Henke and M.A.Tester, Advances in X-ray analysis (Plenum, New York, 1975), Vol. 18; B.L.Henke and K.Taniguchi, J.Appl.Phys. 47, 1027 (1976)

(3) K.Feser, Thesis Univ. München 1971; DESY F41-71/3

(4) J.Müller, Thesis Univ.München 1973; DESY F41-74/1

(5) W.Schnell and G.Wiech, Mikrochimica Acta Suppl.7, 323 (1977)

(6) K.Feser, Phys.Rev.Letters 28, 1013 (1972); Phys.Rev.Letters 29, 901 (1972)

(7) J.Müller, K.Feser, G.Wiech, and A.Faessler, Phys.Lett. 44A, 263 (1973)

(8) E.Tegeler, N.Kosuch, G.Wiech, and A.Faessler, submitted for publication in phys.stat.sol., DESY SR-77/07

(9) N.Kosuch, E.Tegeler, G.Wiech, and A.Faessler, submitted for publication in J. Electron Spectrosc.; DESY SR-77/14

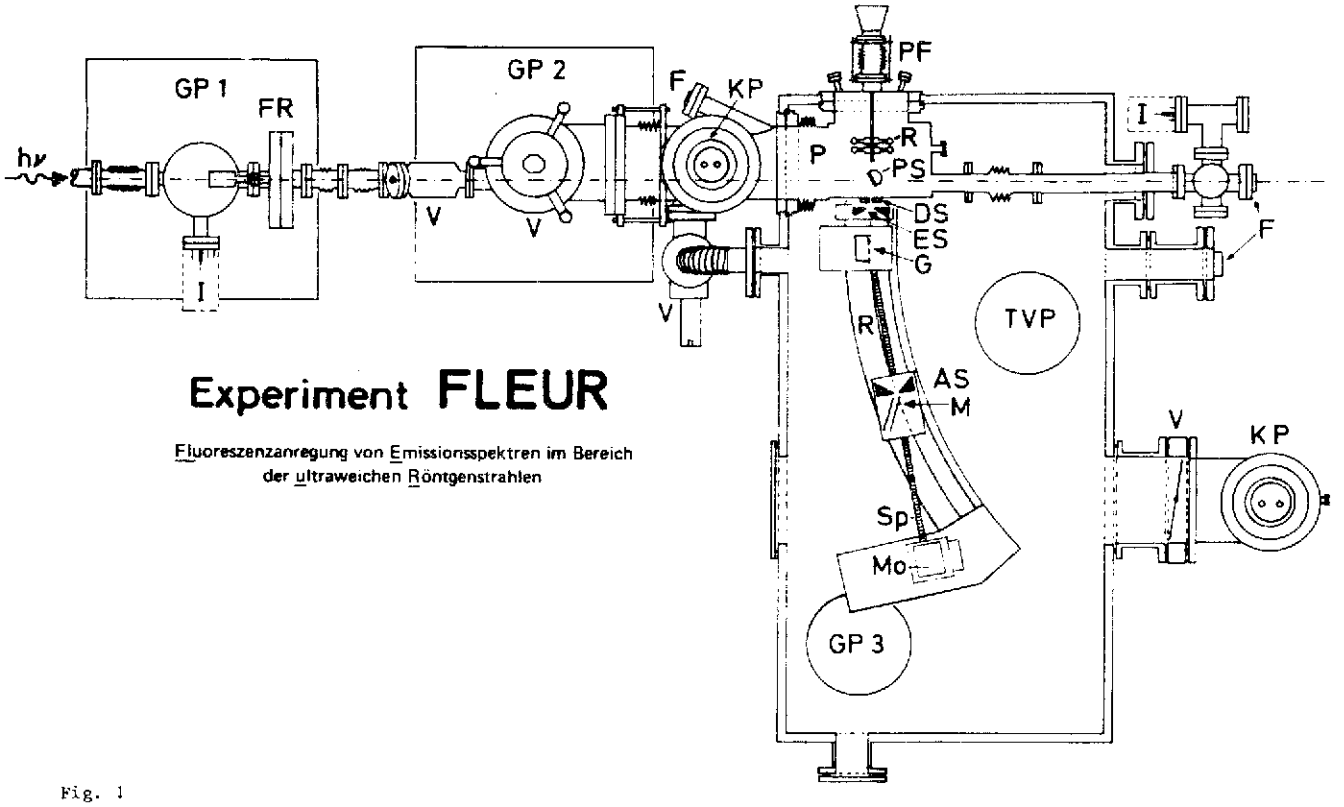
(10) N.Kosuch, E.Tegeler, G.Wiech, and A.Faessler, Chem.Phys.Letters 47, 96 (1977)

Figure captions

Fig.1 Experimental set-up of the experiment FLEUR
(GP ion getter pump, TVP turbo molecular pump, KP cryo pump, I ionisation gauge, V valve, F window, FR filter wheel; P sample chamber with sample holders R and PS, PF manipulator, DS diffusion channel; R rowland track, ES entrance slit, G grating, AS exit slit, M multiplier, SP wavelength drive, Mo step motor).

Fig.2 Photograph of the 2 m concave grating spectrometer unit with (from the right) entrance slit, grating holder, detector carriage with exit slit, wavelength drive with step motor and mechanical counter.

Fig.3 Schematic diagram of the electronic equipment.



Experiment FLEUR

Fluoreszenzanregung von Emissionsspektren im Bereich der ultraweichen Röntgenstrahlen

Fig. 1

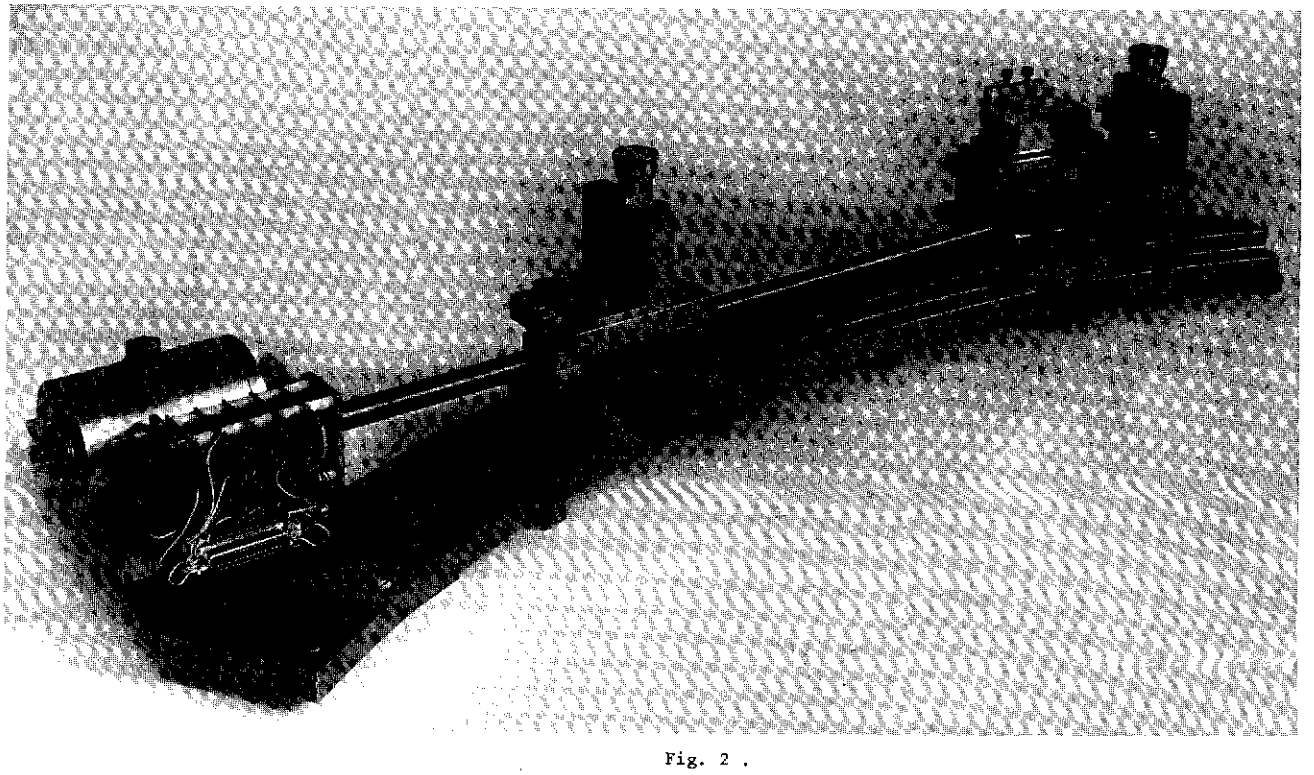


Fig. 2 .

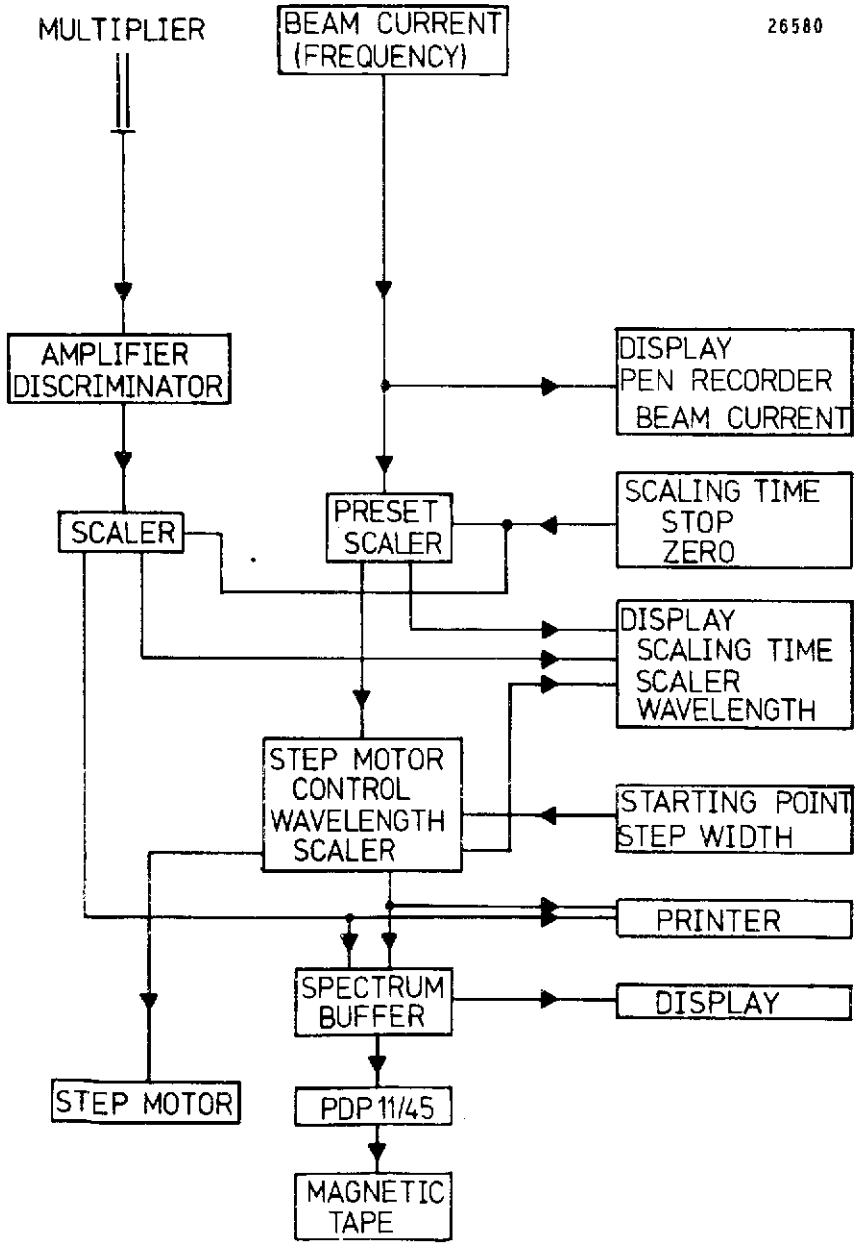

MULTIPLIER
BEAM CURRENT (FREQUENCY)
AMPLIFIER DISCRIMINATOR
DISPLAY PEN RECORDER BEAM CURRENT
SCALER
PRESET SCALER
SCALING TIME STOP ZERO
DISPLAY SCALING TIME SCALER WAVELENGTH
STEP MOTOR CONTROL WAVELENGTH SCALER
STARTING POINT STEP WIDTH
PRINTER
SPECTRUM BUFFER
DISPLAY
STEP MOTOR
PDP 11/45
MAGNETIC TAPE

Fig. 3