# PROFESSOR BRONISLAW BURAS: THE ENERGY-DISPERSIVE METHOD AND SYNCHROTRON RADIATION

**Leif Gerward***

*Dept. of Physics, Technical University of Denmark,DK-2800 Lyngby, Denmark, email: gerward@fysik.dtu.dk*

**Abstract:** In this note I am giving an account of my collaboration with Professor Bronislaw Buras during the early years of synchrotron radiation in the 1970's and 80's. In particular, I am focusing on the development of the energy-dispersive method for X-ray diffraction and its use in high-pressure structural studies. I also describe Buras' role in establishing the European Synchrotron Radiation Facility (ESRF).

*Keywords: Bronislaw Buras, X-ray diffraction, energy-dispersive method, synchrotron radiation, personal recollections*

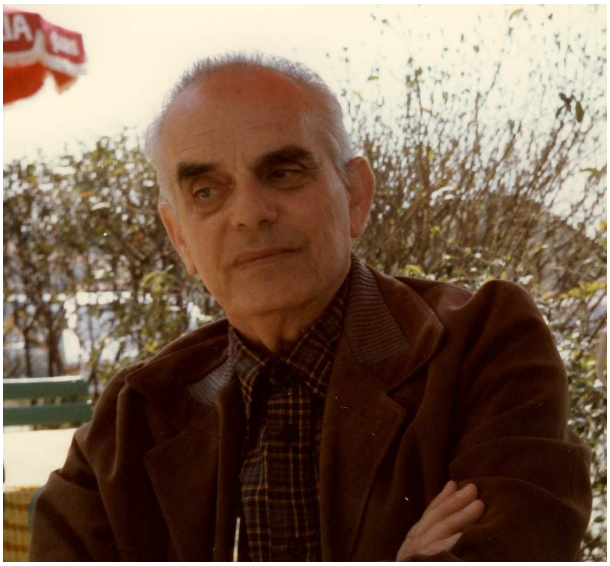

Figure 1. Professor Bronislaw Buras (1915–1994).

*Photo: A. Buras.*

## 1. Introduction

In 1971 Professor Bronislaw Buras (Fig. 1) – then a renowned physicist – immigrated with his family to Denmark. Buras was one of the inventors of the time-of-flight (TOF) method for neutron diffraction [1-3]. In the mid 1960's, he had initiated collaboration between the Institute of Nuclear Research in Świerk and the Danish Atomic Energy Commission Research Establishment Risoe (later Risoe National Laboratory) for building a TOF spectrometer at Risoe, similar to the one in Świerk and another one under construction in Dubna. The International Atomic Energy Agency had supported the project. In the beginning, the Risoe instrument was applied to powders, but the interest shifted to single-crystal methods [4]. Thanks to this earlier collaboration, it was possible to secure an academic position for Buras at the University of Copenhagen with opportunity to perform research work at Risoe.

As for myself – at that time a fresh post doc – I had got a job in November 1970 at the Technical University of Denmark (DTU) in Lyngby, where Professor Asger Lindegaard-Andersen was organizing a new Laboratory for the study of materials using X-ray diffraction methods. It was therefore very natural that we very soon took up discussions with Buras about possible common interests. This was the beginning of a lifelong and fruitful friendship and collaboration.

## 2. The energy-dispersive method

A few years before his arrival in Denmark, Buras *et al*. [5], and independently Bill Giessen and Glen Gordon [6], had invented the energy-dispersive method for X-ray diffraction. In this method the sample is irradiated with polychromatic radiation (the continuous spectrum) from an X-ray tube. The energy spectrum of X-rays scattered at a given angle is observed with a semiconductor detector coupled with a multichannel pulse-height analyser. The energy-dispersive method can be considered the X-ray counterpart of the neutron time-of-flight method.

Buras was of course eager to develop the energy-dispersive method further. We found it an exciting and unconventional way of looking at the Bragg equation. A joint project was set up with me from DTU, and Janus Staun Olsen from the H.C. Oersted Institute at the University of Copenhagen as the main participants. Several other colleagues were involved in the project during the course of time as seen in the list of references below.

To begin with, we repeated the original experiments [7]. We then set out doing a lot of methodological work. We reported the appearance of the silicon Kα escape peak [8], we derived simple relations between the integrated intensities of the various diffraction methods [9], and we studied the influence of polarization [10] and the optimum resolution [11]. Moreover, we extended the method to samples of single crystals [12].

### 3. Early experiments with synchrotron radiation

The potential use of synchrotron radiation for physical research was a much-discussed topic in the first half of the 1970's. We studied with great interest a pioneering paper by T. Tuomi *et al.* [13]. They had used synchrotron radiation in X-ray diffraction topography. It occurred to us that synchrotron radiation would be an ideal X-ray source for the energy-dispersive method in view of its high intensity, high degree of collimation, and the continuous spectrum, extending into the X-ray regime.

Dr. Christof Kunz of the German synchrotron facility DESY in Hamburg was an invited speaker at a Nordic solid-state physics meeting in Gothenburg, Sweden, in June 1975. He talked about the pioneering work with synchrotron radiation done by the DESY group. Staun Olsen and I approached him during a coffee break and mentioned that we had an experiment that seemed to be well suited for synchrotron radiation. It was agreed that we should make a test experiment at DESY. Buras, Staun Olsen and I went to Hamburg in October the same year to discuss the practical arrangements. Buras brought a two-page questionnaire, in his neat handwriting, about everything from beam characteristics to accommodation.

After a formal application, we could perform the test experiment in January 1976. The synchrotron radiation laboratory was situated in a small "bunker" close to the DESY synchrotron. The laboratory was crammed with experimental equipment. In order to get the beam on, one had to call the control room of the accelerator and ask for radiation: "*Strahlung an Bunker eins, bitte!*" In the beginning we were not even allowed to call ourselves, and a DESY technician was present all the time. The people in the control room must have been rather frustrated at our presence, since we needed to open and close the beam shutter very frequently during the test experiment.

Many people had told us in advance that our experiment was doomed to failure. The high level of background radiation at the synchrotron, so they said, would completely saturate the detector, making it impossible to record any meaningful spectrum. Therefore, we had brought a lot of lead plates to shield the detector and the diffractometer from the alleged background radiation. We started building a heavy wall of lead bricks. In one of the bricks, we had drilled a hole for a pinhole collimator. The powder sample was contained in a glass capillary placed on top of a goniometer. Burning a dark spot on a glass plate localized the incident X-ray beam (later, we used so-called green paper). A sketch of the experimental set-up is shown in Fig. 2.

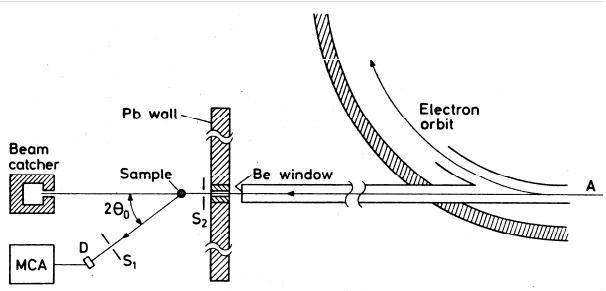


Figure 2. Experimental arrangement for the test experiment at the DESY synchrotron. $2\theta_0$ - fixed scattering angle, $S_1$ and $S_2$ - slits, A - focus point of radiation, D - semiconductor detector, MCA - multichannel pulse-height analyser.

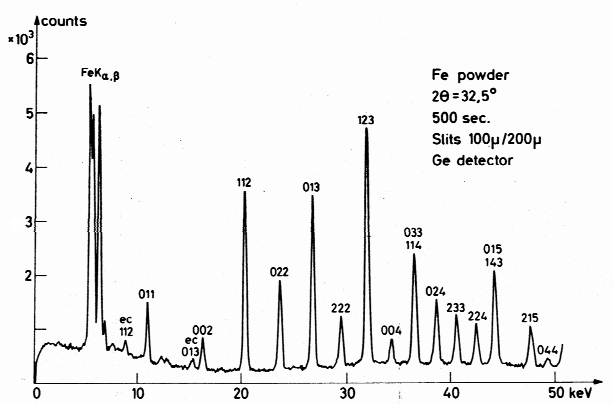


Figure 3. Diffraction spectrum of iron powder. Counting time 500 s. The Bragg angle is $\theta = 16.25°$, ec - escape peak.

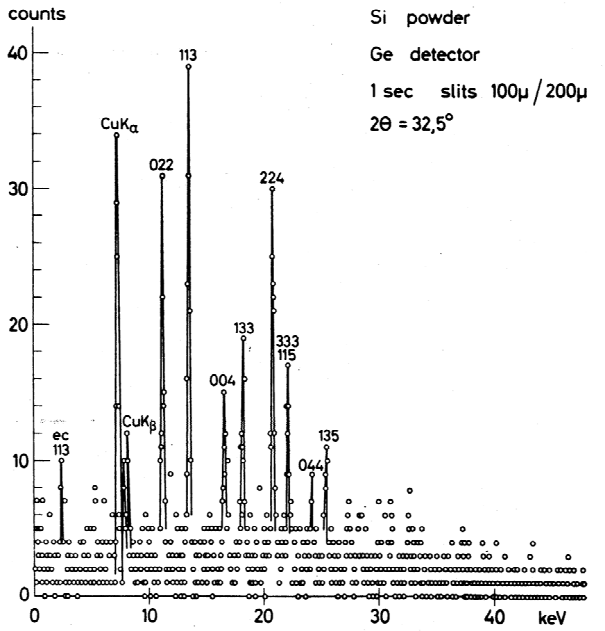


Figure 4. Diffraction spectrum of silicon powder. Counting time 1 s. The notation is as for Fig 3.

Much to our surprise, we got some rather nice diffraction spectra after a few trials with the alignment of the equipment. At the end of our beam time we boldly removed some of the lead shielding – and the spectra became even nicer! An example is shown in Fig. 3. As it turned out, we had created some background radiation ourselves with our excessive lead shielding. A one-second shot for a silicon powder (Fig. 4) demonstrated the sensitivity of the energy-dispersive method. The diffraction peaks are clearly visible, albeit with poor counting statistics, indicating that it should be possible to follow rapid phase transitions in the sample.

It was a happy group that returned home after this successful test experiment. A first note with preliminary results was quickly submitted to *Nuclear Instruments and Methods* and also published as a DESY preprint [14]. When preparing this note, we became aware of a preprint by J. Bordas *et al.* [15], who almost simultaneously had used the energy-dispersive method for small-angle scattering at the synchrotron NINA in Daresbury, UK.

Based on further work, we discussed the special features of the white-beam energy-dispersive method using synchrotron radiation in two subsequent, more detailed papers [16, 17]. We were now looking for some good physics that could be done with our method. Already in our preliminary paper [14], we had pointed out that the fixed geometry of the energy-dispersive method made it suitable for structural studies at extreme conditions, such as high pressure and high or low temperature. Accordingly, we built an oven for 50–850 ±0.25°C, and G. Will and E. Hinze of the University of Bonn introduced us to the diamond squeezer technique for high-pressure studies [18]. Later, we had our own diamond anvil cells of Syassen-Holzapfel type built at the workshop of the H.C. Oersted Institute.

Much better beam conditions could be obtained at the electron storage ring DORIS where a few beamlines were available. Together with Mike Glazer and M. Hidaka of the Clarendon Laboratory, Oxford, we demonstrated that structural refinement of the Rietveld type could be performed on energy-dispersive diffraction spectra recorded at the storage ring [19]. Figure 5 shows another one-second shot with much improved quality as compared with Fig. 4.

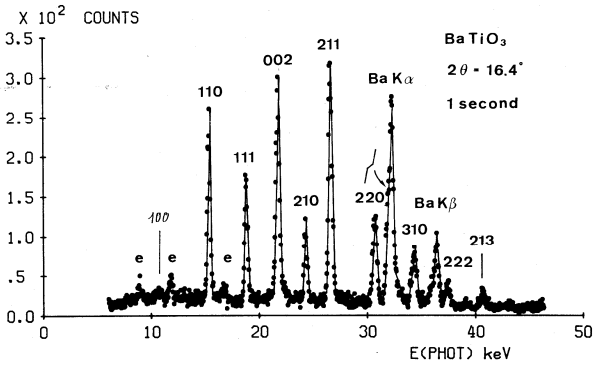


Figure 5. Diffraction spectrum of $BaTiO_3$ obtained at the DORIS storage ring. Counting time 1 s, $\theta = 8.2°$.

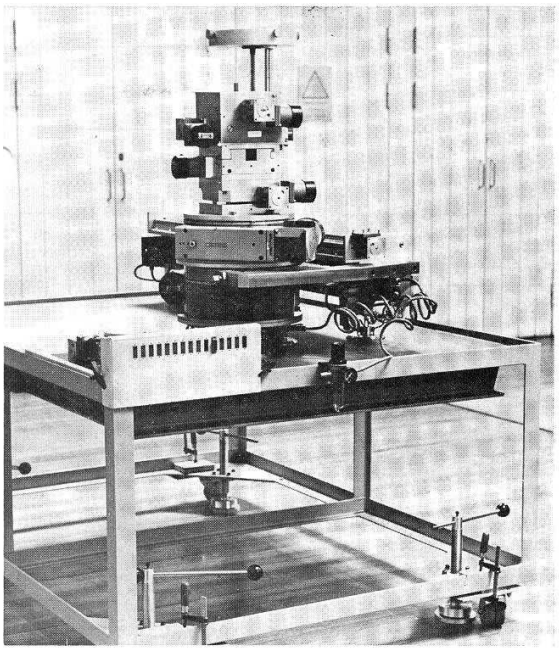

Figure 6. The energy-dispersive spectrometer (EDS).

*Photo: F. Ferrall.*

High-energy physicists ran DORIS, like the other accelerators of DESY, in the search for elementary particles. Synchrotron radiation was from their point of view a waste product, which was used in a "parasitic" way by us and other physicists. However, the demand for synchrotron radiation grew steadily, and it was decided to build a new laboratory hall at DORIS. Moreover, DORIS would run part time dedicated to synchrotron radiation (later it became a fully dedicated radiation source). The new laboratory, which was dubbed HASYLAB (Hamburg Synchrotron Radiation Laboratory), had 15 beamlines when it opened for the users in 1981. Later it was enlarged to 30 beamlines.

Our group became involved in constructing an energy-dispersive spectrometer for HASYLAB (Fig. 6). The main parts of the instrument were a robust Huber goniometer and a strong table with a smooth surface, on which the diffractometer and the detector arm could move on air cushions [20]. The instrument was built in the workshop of the H.C. Oersted Institute and transported to Hamburg on a truck. HASYLAB provided the electronics and a high-purity germanium detector. After some test experiments, the spectrometer was installed at beamline F3 of HASYLAB. It was going to be a workhorse for more than 25 years. In fact, the mechanical parts of the instrument are still in use, although the electronic system and the associated computer have been upgraded several times.

It should be mentioned here that scientists from Risoe National Laboratory also were heavily involved at HASYLAB. They constructed a flexible triple-axis spectrometer (Fig. 7), which was installed at beamline D4 [21, 22]. Later, Risoe implemented several other advanced instruments at the wiggler beamlines.

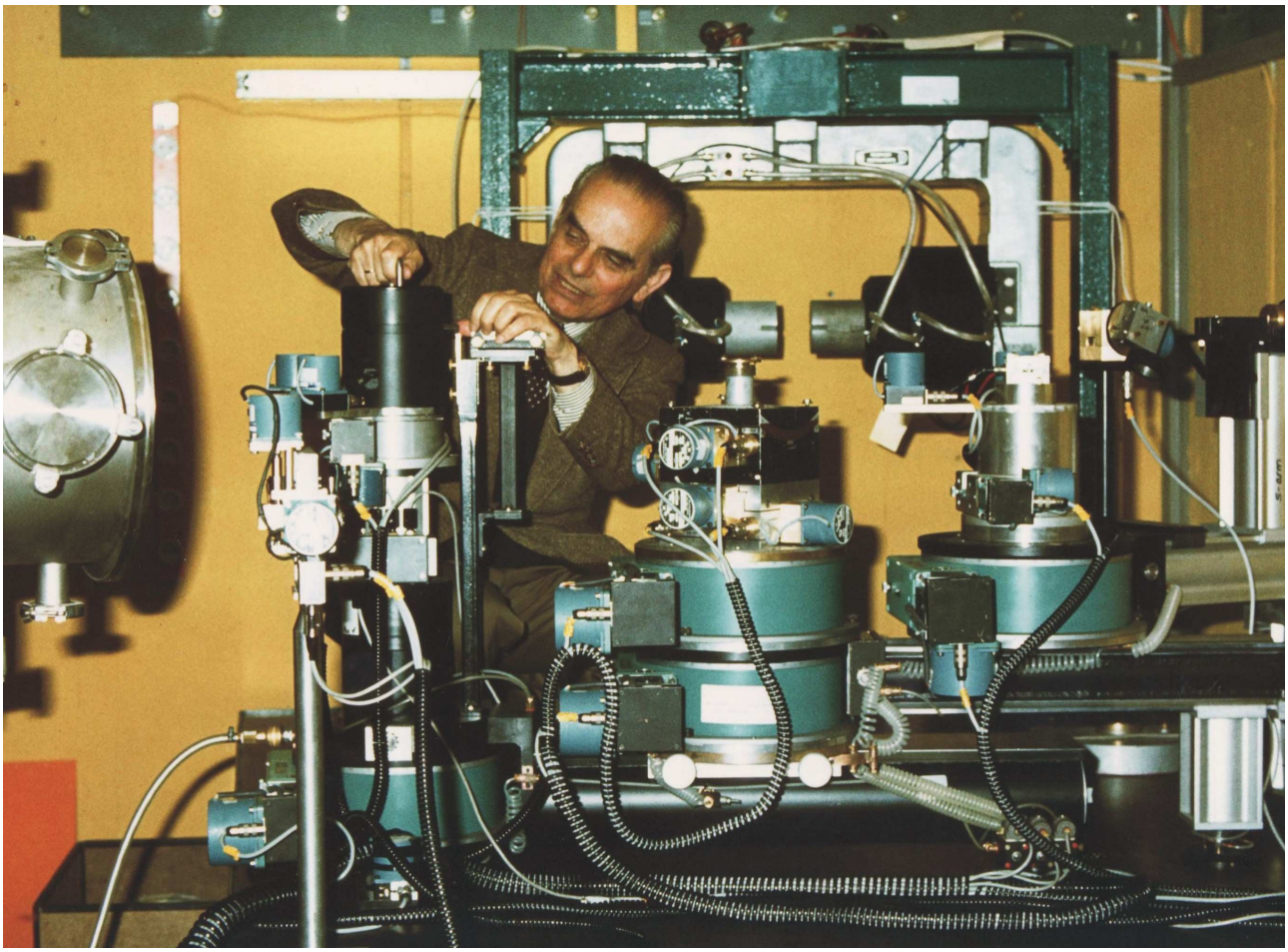

Figure 7. Buras at work, using the triple-axis spectrometer at HASYLAB (in energy-dispersive mode!).

*Photo: B. Lebech.*

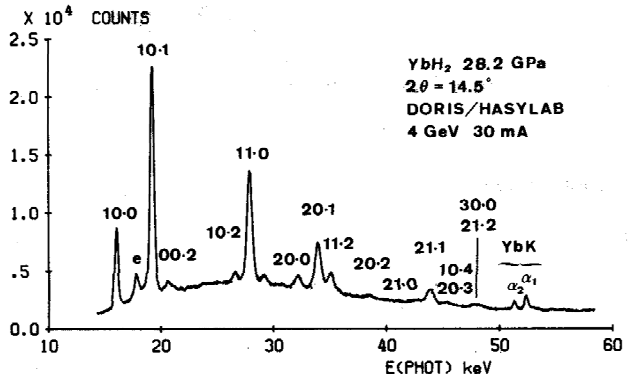


Figure 8. Diffraction spectrum of hexagonal $YbH_2$ at 28 GPa. Counting time 500 s, $\theta = 7.25°$.

Our first full-fledged high-pressure structural study was devoted to $YbH_2$. At ambient conditions, $YbH_2$ crystallizes in an orthorhombic structure with space group *Pnma*. By comparison with other rare-earth dihydrides it was expected that $YbH_2$ should transform into the fcc fluorite structure at high pressure, as the valence state of the Yb atom changes from $4f^n(5d6s)^2$ to $4f^{n-1}(5d6s)^3$.

Buras *et al.* [23] developed a high-pressure cell for neutron diffraction at Risoe, but no phase transformation in $YbH_2$ was observed up to 4 GPa. Using a diamond anvil cell and radiation from an X-ray tube we reached 11 GPa, but there was still no phase transformation. Using synchrotron radiation, however, it was possible to reach 28 GPa, and we clearly demonstrated a phase transformation at about 14 GPa [24].

It turned out that the high-pressure structure of $YbH_2$ was not fcc as expected but hexagonal (Fig. 8). Meanwhile, Bente Lebech *et al.* [25] had determined the crystal structure, including the positions of the hydrogen atoms, at ambient conditions using neutron diffraction. On the basis of their results, they suggested a high-pressure structure with space group $P6_3/mmc$ and with Yb atoms in (2*c*) and hydrogen atoms in (2*a*) and (2*d*) positions. A careful analysis showed that this suggestion was indeed consistent with our X-ray data [26].

The investigation of $YbH_2$ became the prototype for a long and ongoing series of high-pressure structural

studies by our group. Also internationally there was now a growing interest for the energy-dispersive method. Buras travelled to workshops and symposia in the United States to tell about the method and its application to high-pressure studies [27, 28]. Personally, I had the pleasure of representing Buras at the 1979 Spring Meeting of the American Crystallographic Association (ACA) in Boston [29]. It was my first visit to the United States, and Buras took great care in instructing me about the local conditions.

For two decades, the energy-dispersive method was *the* method for high-pressure structural studies in conjunction with the diamond anvil cell and synchrotron radiation. It is only recently that the energy-dispersive method gradually is being replaced by high-resolution angle-dispersive techniques. Still, it is an excellent method for getting a quick overview of the high-pressure behaviour of a given material.

## 4. European Synchrotron Radiation Facility (ESRF)

At this point I lost my almost daily contact with Buras, who was now moving on to new and greater challenges at the European level. We published a summary of our common work in a review paper [30], and in a contribution to the *International Tables for Crystallography* [31].

Particle-physics accelerators had become inadequate to meet the growing demand for synchrotron radiation. Third generation sources, which from the beginning were constructed and optimised for synchrotron radiation, were being planned in Europe and elsewhere. In the case of a European source, it was recognized that an international collaboration was needed in view of the complexity and cost of a machine for radiation in the hard X-ray regime.

H. Maier-Leibnitz presided over a working group set up in 1975 by the European Science Foundation (ESF) to study the feasibility of a synchrotron radiation source to span the entire X-ray region down to wavelengths of the order of 0.1 Å. In 1977 the ESF issued a report "Synchrotron Radiation. A Perspective for Europe", the so-called Black Book.

An ad-hoc committee was set up in 1978–79 and chaired by Y. Farge. Two subgroups were established, one dedicated to the machine and chaired by D.J. Thompson; and one dedicated to instrumentation and chaired by Buras. This work resulted in the publication of a four-volume document "European Synchrotron Radiation Facility. The Feasibility Study" (the Blue Book). An updated document "A Case for a European Synchrotron Radiation Facility" (the Yellow Book), edited by Jens Als-Nielsen, was worked out in 1980–82, incorporating new technological developments, in particular the so-called magnetic insertion devices, wigglers and undulators.

In 1983–84, a European Synchrotron Radiation Project group (ESRP) was created under the leadership of Bronislaw Buras and Sergio Tazzari and located at CERN. Its conclusions were given in "European Synchrotron Radiation Facility – Report of the ESRP" (the Green Book), describing the project goals, the source, experimental equipment, time scale, cost and construction requirements.

The location of ESRF was still undecided, and Risoe National Laboratory made a serious bid for hosting the facility [32]. However, in the end France and the Federal Republic of Germany proposed the implementation of ESRF in France and invited other countries to join. A provisional ESRF Council was formed in December 1985 to set up a construction team in Grenoble, led by Ruprecht Haensel, another synchrotron radiation pioneer from DESY. Electrons were injected into the storage ring for the first time on 17 February 1992, and the first user beamlines were installed during the autumn of 1992. The European dream of a third generation synchrotron radiation source had materialised.

## 5. Some personal recollections

Buras had an indomitable optimism, also in hard times, as his story of life clearly demonstrates. In the beginning of our collaboration, the experimental means were minute. We managed to raise the money for a Si(Li) detector, but the multichannel analyser was borrowed from another group, and the X-ray generator was an antiquated unit that had been delivered by the Germans shortly after World War II. Breakdowns were of course plentiful. At one occasion we removed the front panel and looked into the dusty interior. A post doc that happened to pass by exclaimed: "Look, it's brand new!" However, the experimental conditions improved as we began publishing papers on energy-dispersive diffraction.

Although he understood Danish conversation, Buras seldom used that language himself. He always communicated in English with his students and colleagues. Alan Macintosh, former head of Risoe, addressed this issue in a dinner speech at Buras' 70$^{th}$ birthday. In Denmark, so he said, Buras had got another native language – broken English!

Buras was an inspiring and well-liked teacher and adviser for students and colleagues. Even after having settled in Denmark, he continued to care for his former students in Poland. Buras felt most at ease at his home base, the Risoe National Laboratory. In the experimental hall, he liked to show any available neutron spectrometer to students and visitors. He always impressed by his ability to scan reciprocal space as effortlessly as other people are moving in their own sitting room.

Buras was a prolific writer and a travelling ambassador for science. He was always on the move, on his way to conferences and other meetings. In particular, he cared for the neutron time-of-flight method and its X-ray counterpart, the energy-dispersive method. These interests led him to take a great responsibility for the development of synchrotron radiation as an X-ray source in studies of materials.

In conclusion, I am deeply grateful for having had Bronislaw Buras as a mentor and colleague. During our years of collaboration I learnt a lot from his inspiring leadership and enthusiasm for scientific work, combined with a good sense of humour.

**Acknowledgments**: I wish to thank Wojciech Paszkowicz for suggesting this contribution to the *Bulletin of the Polish Synchrotron Radiation Society*. I am grateful to Andrzej Buras, Bente Lebech, Asger Lindegaard-Andersen and Jens Als-Nielsen for helpful discussions. Finally, I wish to thank Janus Staun Olsen for a nice collaboration over more than 30 years.

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