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A Prospect and Retrospect – the Japanese Case

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The early through recent history of synchrotron radiation research in Japan, since the initial efforts in 1962, is reviewed. Following a period of parasitic use of an electron synchrotron, Japanese users attempted to build a storage ring as a dedicated soft X-ray source, which was completed in 1974. It opened up a new era of second-generation synchrotron radiation research. The Photon Factory, a dedicated X-ray source commissioned in 1982, provided a much wider research area as well as a number of technical innovations, among which insertion devices brought the further prospect of significant improvements in the properties of sources. As a consequence, the new concept of a light source oriented towards full exploitation of insertion devices, or the idea of a third-generation source, was created. The motivations and developments which led to Spring-8, a third-generation Japanese X-ray source that is currently being commissioned, will be reviewed briefly.

Keywords: dedicated sources; undulators.

1. Prehistory: 1947–1962

In the year 1947, which we are commemorating, Japan had no synchrotrons, or even accelerators. Before World War II came to an end, Japanese physicists had constructed four cyclotrons, in Tokyo, Kyoto and Osaka. However, as nuclear power became a highly political and military issue during World War II, the Supreme Commander of the occupation forces in Japan had ordered a ban over any attempts to operate and construct particle accelerators in Japan. Accordingly, all the then existing cyclotrons were destroyed and abandoned deep in the waters of Tokyo Bay and Osaka Bay in the winter of 1945–1946.

This policy of the occupation forces came to the attention of a few American physicists, such as Arthur Compton and Ernest Lawrence. Compton proposed compensation for the destroyed cyclotrons, and his idea of donating a helium liquefier, which was totally missing in Japan at that time, resulted in the founding of the Institute for Solid State Physics (ISSP), University of Tokyo, which was later strongly involved in synchrotron radiation activities. On the other hand, Lawrence came to Japan in 1951 with a clear intention to advise the occupation forces that pure scientific activities should not be prohibited. As a result of his intensive efforts, the ban on accelerators was lifted even before the peace treaty, which became effective in 1952.

Accordingly, a comprehensive program in particle and nuclear physics, including the construction of accelerators, started in 1952 under the leadership of Tomonaga and Kikuchi. The Institute for Nuclear Study, INS, was thus established in 1955 as the first Collaborative Research Institute affiliated to the University of Tokyo, with Seishi Kikuchi as the founding director general. These two laboratories jointly supported the initial phase of synchrotron radiation research in Japan until the Photon Factory was established. Kikuchi took with him the majority of his laboratory staff from Osaka University, including Hiroo Kumagai and Seitaro Yamaguchi who took the initiative in constructing an electron synchrotron. Construction of the 1.3 GeV electron synchrotron started in 1956, and was completed in 1961 with an initial energy of 750 MeV.

2. Parasitic use of synchrotron radiation: 1962–1974

It should be noted that many Japanese physicists had already paid serious attention to the potential use of synchrotron radiation as early as the 1950's, being stimulated by the intensive activities at Cornell University under the strong leadership of Dean Tomboulian and Paul Hartman. Two leading physicists from Osaka University were staying in Ithaca at this time and witnessed the developments with synchrotron radiation. They were Seishi Kikuchi, a nuclear physicist, well known for the term 'Kikuchi patterns' in electron diffraction, and Masao Sawada, an X-ray spectroscopist, who was aware of the potential use of X-ray spectroscopy, in particular, in the structure studies nowadays known as EXAFS. Sawada advised Yamaguchi to attach a beam port for extracting radiation from the synchrotron orbit.

The organization of a user group 'INS-SOR' was started soon after the commissioning of the electron synchrotron, in April 1962, under the initiative of Takanori Oshio and the present author. Oshio (Oshio & Sasanuma, 1968) was engaged in soft X-ray astronomy, and made evaluations of the practically available soft X-ray output at the front end. At that moment my major concern was the determination of dielectric functions in the extreme ultraviolet beyond 10 eV. My light source was a spark discharge of mixed vapors under low pressure which produced a number of discrete ionic emission lines beyond 10 eV. The photon flux available at the sample for reflectance measurements from this source was typically below $10^9\ s^{-1}$. On the other hand, the predicted photon flux from the INS electron synchrotron was two orders of magnitude higher than this value, and furthermore it would provide a broad continuum, or unlimited tunability beyond 25 eV, which was the practical upper limit of any discharge sources. We submitted our proposal for a full exploitation of this tremendous potential capability as soon as the use of the synchrotron was made open. We were admitted as one of the authorized user groups of the synchrotron with a two-week beam time every half year. The rest of the operational time was additionally made available although only in parasitic mode.

The first successful users' experiments were performed in March 1965 by a single collaborating team, INS-SOR. The synchrotron in that initial phase concerned us greatly with its noise levels, and we started with photographic detection. As the INS synchrotron was operated at a much higher electron energy than the NBS, it allowed us to explore deeper inner shells, for instance, Cl $L_{2,3}$ absorption spectra of alkali chlorides (Sagawa, Iguchi, Sasanuma, Nasu *et al.*, 1966) and also the K and $L_{2,3}$ spectra of a few metals (Sagawa, Iguchi, Sasanuma, Ejiri *et al.*, 1966). The first observations of the Rydberg series of argon $L_{2,3}$ (Fig. 1) and nitrogen K-absorption spectra are also examples of these initial results (Nakamura *et al.*, 1968).

We were aware that Bob Madden's group at the NBS had already published beautiful spectra of rare gases. We followed them in observing several rare gases to confirm their results, and also to check the resolving power of our spectrograph by comparing our spectra with the aforementioned NBS spectra (Sasaki, 1987). Everybody who took part in this experiment was strongly impressed in its intensity, to notice the extremely short exposure time required for recording any one absorption spectrum, and also in the beauty of the continuum, which nobody had ever seen with other light sources. Its unique and extraordinary character was fully envisioned at this first moment.

3. Users attempts towards a dedicated source: 1965–1975

Exciting achievements by the NBS group, followed by the initial efforts of INS-SOR, envisaged that a huge previously unexplored regime of interactions of radiation with matter was emerging beyond the horizon. These initial findings opened up a new era of inner-shell spectroscopy of atoms, molecules and solids. At the same time, they had also demonstrated the unique and extraordinary power of synchrotron radiation. As a natural consequence, this new field of science attracted the attention of a number of physicists, chemists and researchers from other disciplines, who anticipated a variety of new opportunities provided by this novel scientific tool.

It resulted in Japan in a sudden increase in the number of demanding users, who had to share the beam time allotted by high-energy researchers, and consequently the beam time available to each user group was now divided into a smaller and smaller fraction of a definite total. Another beamline was implemented and second and third monochromators were set up, but all these efforts did not meet the growing demands. Furthermore, a majority of the beam time available to users was in parasitic modes, which were awfully inefficient, or sometimes simply a waste of time. Our early experiences convinced us that we needed our own machine as a light source.

On the other hand, it came to our attention that the storage ring of MURA, Wisconsin, primarily designed as a test machine for colliding beam experiments, was going to be converted to light sources. The idea of hunting any operating or abandoned machines to be converted to a light source was being discussed here and there in Europe and the United States, as Paul Hartman (1982) of Cornell recollected later that: 'We, synchrotron radiation people, were pirates'. The technical feasibility of a small storage ring had already been demonstrated with the small ring 'AdA', which was an Italian–French collaboration (Bernardini, 1963). We learned from their initial tests that the lifetime of a beam in a small storage ring is primarily density-limited, or governed by the 'Touschek effect', as far as it is operated under ultra-high vacuum, and if we compromise with a reasonable emittance, a fairly stable operation will be guaranteed. These results encouraged INS-SOR users, because a small ring of 300 MeV could easily be run by feeding electrons from the INS synchrotron which was already in operation, and it would cover the soft X-rays in which they were interested. Furthermore, it was obvious that a storage ring had a great advantage over electron synchrotrons. In electron synchrotrons, electron energy varies repeatedly from injection to the final stage of acceleration. Accordingly, the spectral character of the radiation, as well as the source size, shape and position, may also vary, whereas a storage ring is much more stable in all these respects and will allow a higher current, or a higher brightness. If there had been any attempts to construct a high-energy collider among the Japanese high-energy physicists, we might have tried to be a parasite on it, or even a 'pirate'. But at that moment the high-energy community in Japan were not interested in colliders and a proton machine was given priority.

Therefore, INS-SOR had no other option than to build a storage ring as a dedicated light source by their own efforts. They submitted a proposal in 1965, and finally the budget was approved in the fiscal year 1971–1973 and the first beam was stored in December 1974 (Fig. 2). SOR-RING (Miyahara *et al.*, 1976), the first purpose-built accelerator as a light source, was unique in some respects. The designers and constructors, including the present author, were all synchrotron radiation users without any previous experience or knowledge of accelerator physics, or engineering.

Although initially funded by the INS, this newly built storage ring was transferred soon after its completion to the ISSP, which took over the responsibility for operation in 1975. It ran without interruption for more than 20 years, serving more than a hundred users every year, and terminated its operation for users in March 1997.

4. Extension to X-rays: Photon Factory 1978–

Shortly before the SOR-RING was completed, a study group of crystallographers was organized to investigate the means of achieving high-brightness X-rays under the initiative of Kazutake Kohra. Two options were considered, a high-current rotary anode or a synchrotron source. They soon reached the conclusion that a storage ring with an energy higher than 2 GeV should be preferred to meet their demands. Kohra soon expanded the study group to a large user group for promoting the project, and asked Kazuo Fuke, an accelerator physicist at the INS, for collaboration. The plan was authorized by the Science Council of Japan in the fall of 1974. Under an agreement with the Laboratory of High Energy Physics (KEK) in Tsukuba, the facility, the Photon Factory, was placed within the site of KEK. Construction was financially supported for the fiscal years from 1978 through 1981.

Meanwhile there were rising demands from X-ray users worldwide. It was indeed a second crest of synchrotron radiation fever, which was stimulated by the advent of big colliders, in Stanford, Hamburg and Novosibirsk, in the mid-1970's. All these newly established facilities associated with colliders allowed parasitic use as light sources, and sooner or later they were converted to dedicated sources. This again came as benefits provided by developments in high-energy physics, but in this case the actions taken by the synchrotron radiation users were prompt. Even though they were admitted only as parasites, it soon turned out that the X-ray part of synchrotron radiation opened up tremendous novel opportunities in science and technology. It should be noted that such a novel experimental technique as EXAFS had already been developed during this period of parasitic use of storage rings.

The novel opportunities envisaged by high-energy storage rings encouraged users to propose dedicated X-ray sources worldwide and they were completed one after another during the early 1980's. The Photon Factory was one such source completed in 1982, and since then has been serving more than 2000 active users, academic and industrial, domestic and overseas, at more than 50 experimental stations (Kohra & Sasaki, 1983).

The impact of these new X-ray facilities was explosive. In addition to the previously employed techniques of spectroscopy, a number of new opportunities came into the menu, such as EXAFS, protein crystallography, fluorescence analysis, surface and interface studies with diffraction and interference, diffraction topography, microscopy with absorption or phase contrast, photoelectron diffraction, small-angle scattering of living substances such as muscles and membranes, real-time studies of phase transitions of solids under high-pressure/high-temperature regimes, and medical applications such as coronary angiography and computed tomography, to mention only a few examples at random.

Also it should be emphasized that the dedicated sources allowed rapid progress in the techniques of design and operation of accelerators as light sources. They were optimized to produce a stable, high photon flux to a small, limited sample area as far as possible. Undertaking such

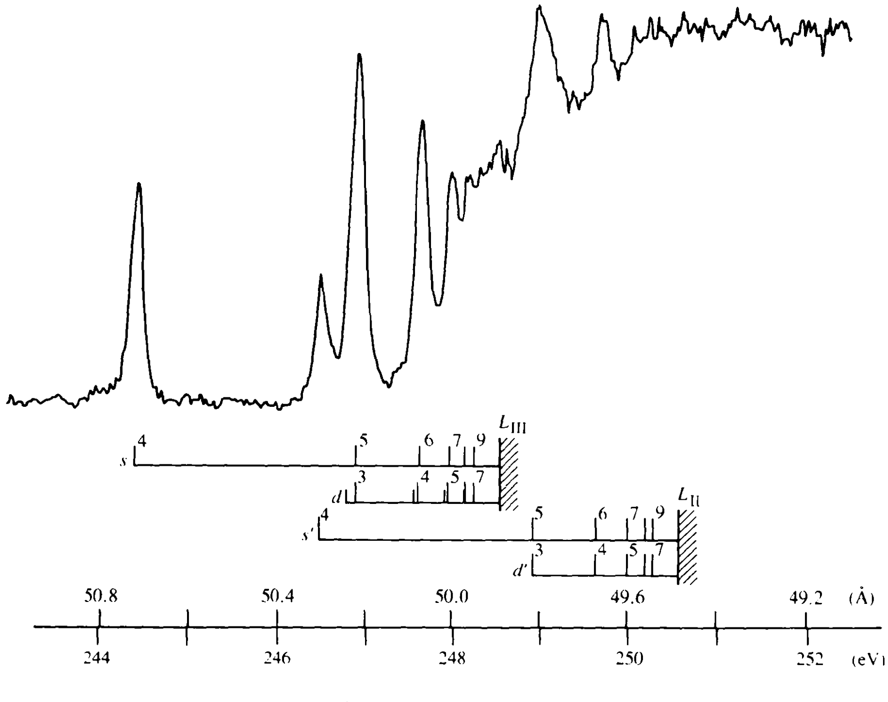

Figure 1
Rydberg series of the argon $L_{2,3}$ absorption spectrum (Nakamura *et al.*, 1968).

efforts in optimizing a source optically would never have been made when the machine was dominantly used for high-energy experiments. This was the most significant effect of dedicated sources. The initial design goal of dedicated sources was typically a stored current of 100 mA with a lifetime of 10 h, but this landmark was soon overtaken, and a current of 300 mA with 50 h lifetime is no longer exceptional. Reducing the emittance of the source for obtaining higher brightness was also seriously attempted.

Figure 2
First light from SOR-RING, December 1974.

Monitoring the profile and position of the beam is essential in attaining high stability of the source, which is in turn essential in any precision measurements. Now such monitoring is made to an accuracy of 10^{-6} m, or even better. Small beam-position drifts due to daily and seasonal temperature variations *via* expansion of the accelerator housing were so clearly observed that the corresponding correction measures have easily been taken. It turned out during the initial operation of Spring-8 that the tidal motion of the earth due to both the positions of the sun and the moon may also cause orbit dilation by 10^{-5} m in diameter.

5. Advent of insertion devices: 1980–

The concept of an undulator was first presented 50 years ago in a Russian journal (Ginsburg, 1947). Therefore, 1997 is the joint 50th anniversary for the first observation of synchrotron radiation to be described by Dr Blewett and for the first proposal for an undulator by a Russian physicist.

The principle and character of undulator radiation were already confirmed in the 1950's through pioneering experiments by Motz (1951, 1953) at Stanford. He even anticipated that it could be used as a vacuum ultraviolet source, if an electron accelerator of GeV class became available. It should be noted that the undulator became a practical device, compatible with the stable operation of a storage ring, only when Klaus Halbach (Halbach *et al.*, 1981) and Herman Winick invented a permanent-magnet design to be inserted from outside to the orbit (Brown *et al.*, 1983). Being stimulated by this idea, a joint Photon Factory and University of Tokyo team built a prototype ten-period undulator and tested it at SOR-RING in 1981 (Kitamura *et al.*, 1982; Maezawa *et al.*, 1983) (Fig. 3).

The results were very encouraging, such that the spectral brightness, angular divergence and polarization of an undulator predicted by the theory were fully confirmed in the absolute scale, and it turned out that the perturbation to the electron beam caused by insertion was negligible, or easily corrected.

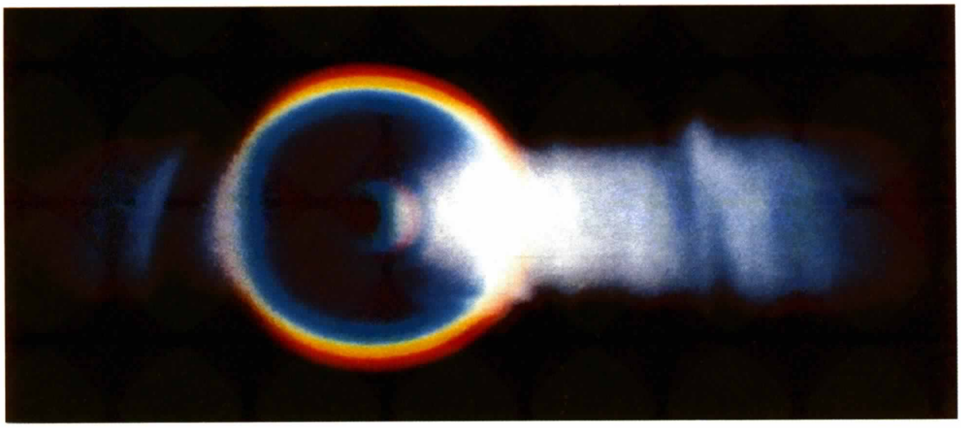

Figure 3
A rainbow of undulator radiation at SOR-RING operated at 200 MeV, December 1981. Two faint extra rainbows at the center are produced by fringing fields of dipoles, and stay even if the undulator magnets are removed (Kitamura *et al.*, 1982).

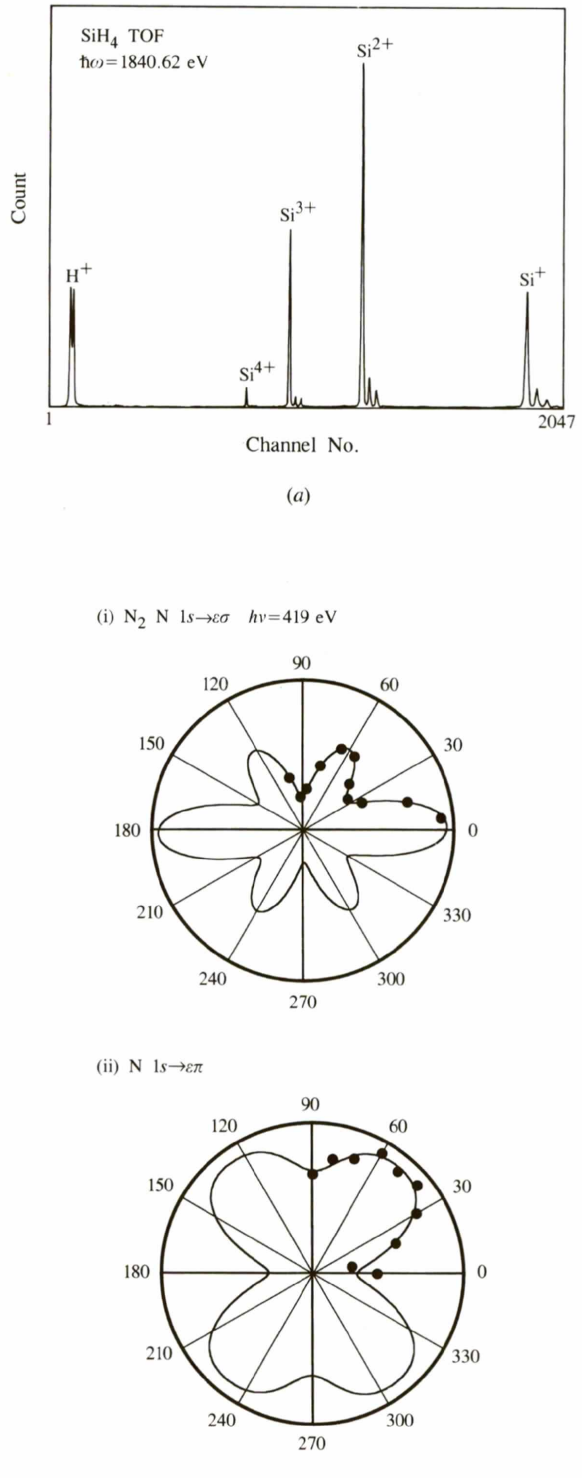

Figure 4

(*a*) Time-of-flight mass spectra of molecular-ion fragments produced by Si *K*-photoionization of SiH$_4$. Ions are dominantly bare silicon without traces of hydrogen bonded to Si. Satellites observed at higher mass sides are due to Si isotopes (Shigemasa *et al.*, 1990). (*b*) Angular distributions of *K*-shell photoelectrons against the molecular axis (angle 0°) of nitrogen at the shape resonance, 419 eV, with the final-state symmetry (i) parallel and (ii) perpendicular to the axis. The final state (i) shows dominantly *f*-symmetry, whereas (ii) has $p + d$ character, supporting the theory of shape resonance (Shigemasa *et al.*, 1996).

Undulators were not included in the initial plans for the Photon Factory, and a formal budget proposal for undulators was presented only after the success of the prototype device. Nevertheless, the design of the light source was already modified to accommodate the possible insertion of wigglers and undulators, based on the studies of the orbit design team of the Photon Factory headed by Kihara with respect to the compatibility of the reasonable orbit with these insertion devices. The full circle of the initial design orbit of the Photon Factory was thus modified into a quasi-ellipse, which included two long straight sections. One of them, BL-16, was occupied by a superconducting vertical wiggler designed by Yamakawa, and another line, BL-2, was reserved for an undulator with a fundamental magnet period of 6 cm, designed by Kitamura.

The spectral brightness of undulators at their fundamental peak, which could be three orders of magnitude higher compared with radiation emitted from the normal dipole at the same photon energy, and the excellent collimation and high degree of polarization, are great advantages of undulator radiation, suggesting to users that new frontiers are now open. The results of two experiments performed so far at BL-2 will be given below as examples.

The first example is a time-of-flight mass–charge spectrum of molecular-ion fragments produced by the Auger-cascade decay of the *K*-shell hole of silicon in silane, or SiH$_4$ (Fig. 4*a*). Chemical instability induced by *K*-shell photoionization is so destructive that the molecule is fully decomposed to yield dominantly bare silicon ions without

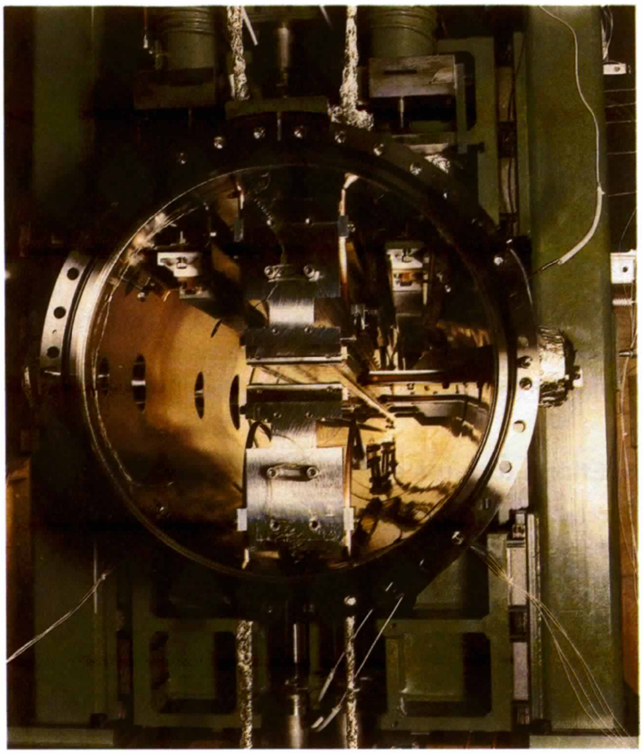

Figure 5

In-vacuum undulator of the 6 GeV TRISTAN AR-ring at the Photon Factory (Yamamoto *et al.*, 1992).

traces of hydrogen (Shigemasa *et al.*, 1990). It was shown that undulators are not only useful in their fundamental peak, but the higher harmonics are also useful. This experiment was performed with the fourth-order component of the 410 eV fundamental.

The second example (Fig. 4*b*) shows the angular distributions of photoelectrons associated with the specific molecular excitations of spatially oriented nitrogen molecules by Shigemasa and Yagishita (Shigemasa *et al.*, 1996). The virtual orientation of molecules is realized by specifying the final-state symmetry of photoions in relation to the plane of linear polarization of the undulator photons. It is necessary to use the coincidence technique of photoions and photoelectrons, which is feasible only with such intense and highly polarized undulator radiation.

The activities of the Photon Factory were later extended to include a part of the 6 GeV accumulator ring (AR), an electron/positron injector to Tristan, a 30 GeV collider. Two further novel types of undulator were introduced to the AR beamlines: one is an elliptical multipole wiggler (EMPW) with crossed-field magnets for producing circularly polarized X-rays, and the other is tunable small-gap magnets sealed in a vacuum (see Fig. 5), both designed by Yamamoto and Kitamura (Yamamoto & Kitamura, 1987; Yamamoto *et al.*, 1992). The former proved to be extremely powerful in magnetic Compton-scattering experiments, the latter in nuclear resonant scattering experiments at 14.4 keV.

After some time, no room remained for mounting more undulators at the 2.5 GeV ring of the Photon Factory, and also the running undulator beamlines were always over-occupied. People began to feel its potential had now been almost exhausted.

On the other hand, an intensive effort towards characterizing the undulator of BL-2 on an absolute scale (Maezawa *et al.*, 1986) demonstrated that the full strength of an undulator cannot be achieved unless the emittance of the beam is reduced to 10 nm rad, or less. It was concluded that we have been losing nearly two orders of magnitude of peak brightness (Sasaki, 1986) (Fig. 6) due to the high emittance of the Photon Factory, which was 400 nm rad in 1985. It was clearly shown that the next target of a synchrotron radiation source was to achieve a low-emittance lattice-design architecture so as to exploit the full capability of undulators.

6. Efforts towards a new generation source: 1985–

Successful commissioning of undulators stimulated a new hope that we may be able to improve the capability of synchrotron radiation dramatically by the intensive and systematic use of such powerful devices in the improved design of future light sources. However, the existing second-generation machines are obviously all too small to produce hard X-rays with undulators, and if one wants to produce 10 keV, or 20 keV X-rays at the fundamental peak of the undulator spectrum, one has to go to 5–10 GeV machines.

In 1985, the Japanese user community, mainly composed of the users of the Photon Factory, started feasibility studies for a high-brightness X-ray source of 6–8 GeV as well as the program for its utilization, as a joint effort of two independent groups, one centered in Kanto, or eastern Japan, with Kikuta as its representative, another from Kansai, or western Japan, represented by Kakudo and Mitsui (Mitsui, 1986). The Kansai group expressed the hope of inviting the new facility to Harima, Hyogo Prefecture, with strong support being guaranteed by the local government of Hyogo Prefecture. This is exactly what we see now as the site of Spring-8.

RIKEN initially planned to have their own modest synchrotron radiation source independently, but they finally decided to take up users' demands for a third-generation X-ray source under the leadership of Kamitsubo, as a national facility supported by the Science and Technology Agency. Thus, Spring-8 has been authorized and funded by STA since 1988, and also it was decided that it should be built in Harima.

A serious problem arose as soon as the design calculations started: the low-emittance beam might be driven only within a very small region of space. It looked at first a tough problem. However, once the incentive for designing a low-emittance lattice was created, the progress was rapid, and a number of options for meeting these requirements were proposed. These concurrent efforts started in Europe, America and Japan around 1987, first independently, soon with close international collaboration.

We note that in the SRI conference in Stony Brook, Wrulich of Trieste reviewed the achievements of the few

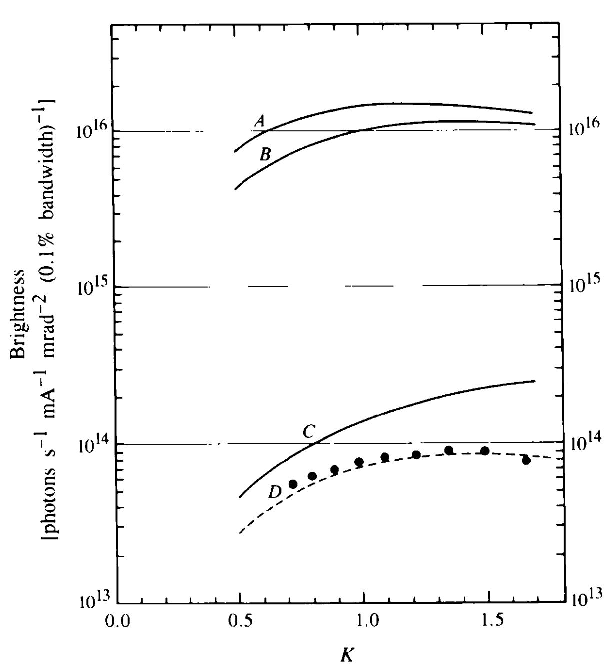

Figure 6

Calculated (*A*, *B*, *C*) and observed (*D*) peak brightness of 6 cm × 60 period undulator at BL-2, Photon Factory, against the field parameter *K*. Curve *A* is plotted by assuming a low-emittance limit (<10 nm rad^{-1}), *B* is corrected for a finite aperture, and *C* is the value for the actual emittance of PF-ring, 400 nm rad^{-1}. The absolute brightness (*D*) was deduced from the photoelectron count of helium as the target gas (Sasaki, 1986).

third-generation sources that were being commissioned then and concluded that all the new machines had achieved excellent performance in this respect and that low-emittance operation had been proven to be compatible with the high current and stability. Results were very encouraging, or even better than theoretical predictions. This is also likely to be true for the newly commissioned Spring-8.

7. Concluding remarks

We may draw a few key notes from the history of synchrotron radiation research over the last half century. It was provided in the first phase of its development as a benefit or a by-product of high-energy physics, and the pioneers, mostly spectroscopists, found in it what they had eagerly awaited for a long time. Synchrotron radiation filled a dark gap in the electromagnetic spectrum which had not been explored previously by anybody. Early users paid a formidable price to access this valuable source solely because they knew nothing else could replace it, and they found it deserved to do so. Soon these efforts were rewarded with many new findings in the atomic and electronic structures of matter and technical innovations to meet these objectives. The advent of dedicated sources was a natural consequence of these developments, and it in turn promoted a tremendous expansion of the research area and technical progress of synchrotron radiation sources and optics. The main driving force dominating these interactive processes has been the initiative of users in pursuit of a better understanding of nature, namely, the quest for previously unexplored features of the interaction of radiation with matter, and efforts towards higher spatial, temporal, momentum, or energy resolution in any optical or spectroscopic observations, which eventually led to the desire for higher brightness, specific polarization and coherence of the source. These ascending processes have not yet come to an end, but still go on ceaselessly.

Collaborative interactions between users and providers of light, namely, accelerator, optics and instrumentation workers, also contributed greatly to the rapid progress that yielded the present state of the art, and it should be emphasized that the role of such interactions will be more and more crucial in the forthcoming phase of synchrotron radiation developments.

This review is a rough sketch of the history of synchrotron radiation studies in Japan only along the main stream. A more comprehensive review will be found elsewhere (Sasaki, 1994)

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