History of Synchrotron Radiation Research

Taizo Sasaki

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Outline

- 1. Introduction --- Early History
- 2. Parasites on Electron Synchrotrons
  - The First Generation
- 3. Dedicated SR Sources
  - The Second Generation
- 4. Advent of Insertion Devices
- 5. Motivations for the 3rd Generation SR
- 6. Summary

1. Introduction

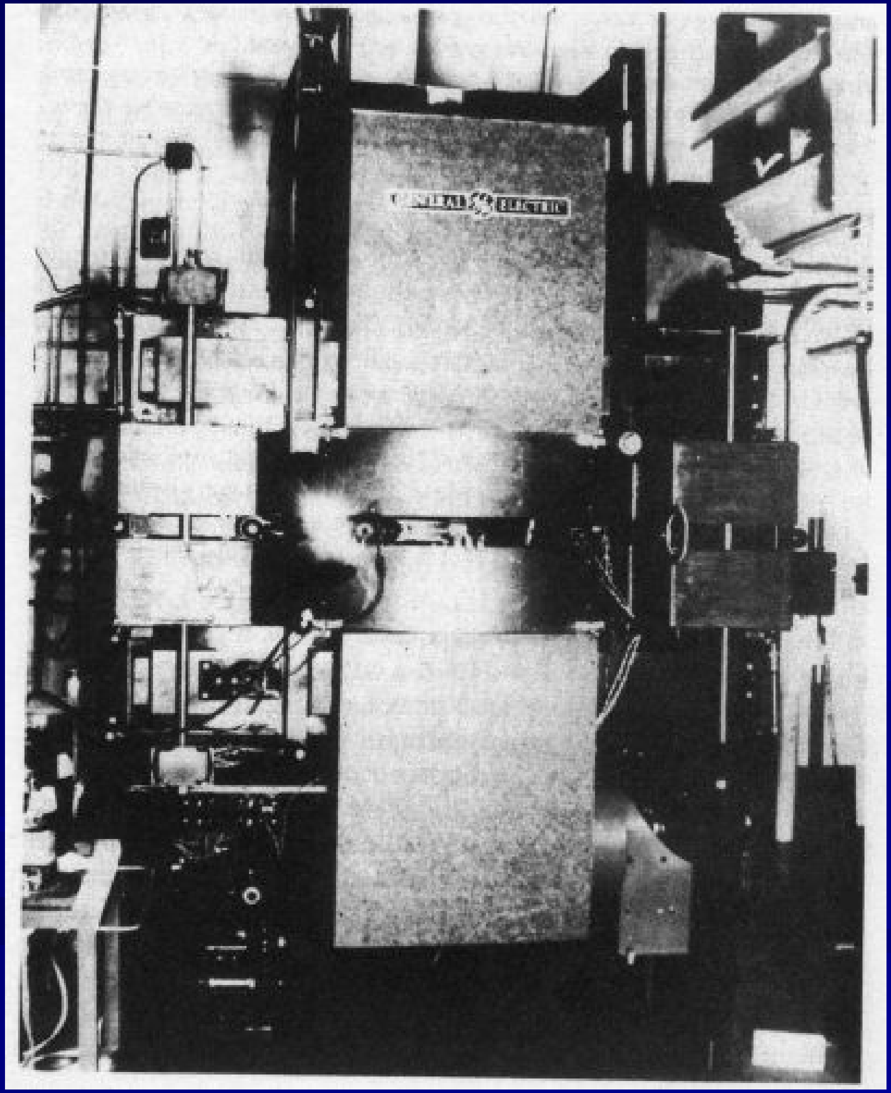


Fig.1 The first observation of SR, April 24, 1947

General Electric Co.
Schenectady, New York
70 MeV Electron Synchrotron
Pollock, Langmuir et.al.

SR as a Nuisance of Electron Accelerators.

Energy input for accelerating electrons is mostly lost to the radiation.

Radiation loss $\propto \gamma 4$

SR as Astronomical Phenomena

Fig.2 Crab Nebula, MS-1. Observation by Hubble Telescope

In 1928 John Hubble of Mt. Wilson Observatory noticed all the constituents of Crab Nebula were flying apart, and they started expanding at 1054 from a single point close to Taurus γ.

This point was just the location of the Super Nova recorded by ancient Japanese and Chinese astronomers. Thus the Crab Nebula became the first astronomical object identified as the source of SR.

R. Wielebinski, SRN 19(2007) 4.
T. Sasaki, SR Science and Technology Information ol.4,
No.6, 44; No.7, 31(1994)

Experimental Verifications of Schwinger's Theory

D. H. Tomboulian : Cornell University 300MeV ES

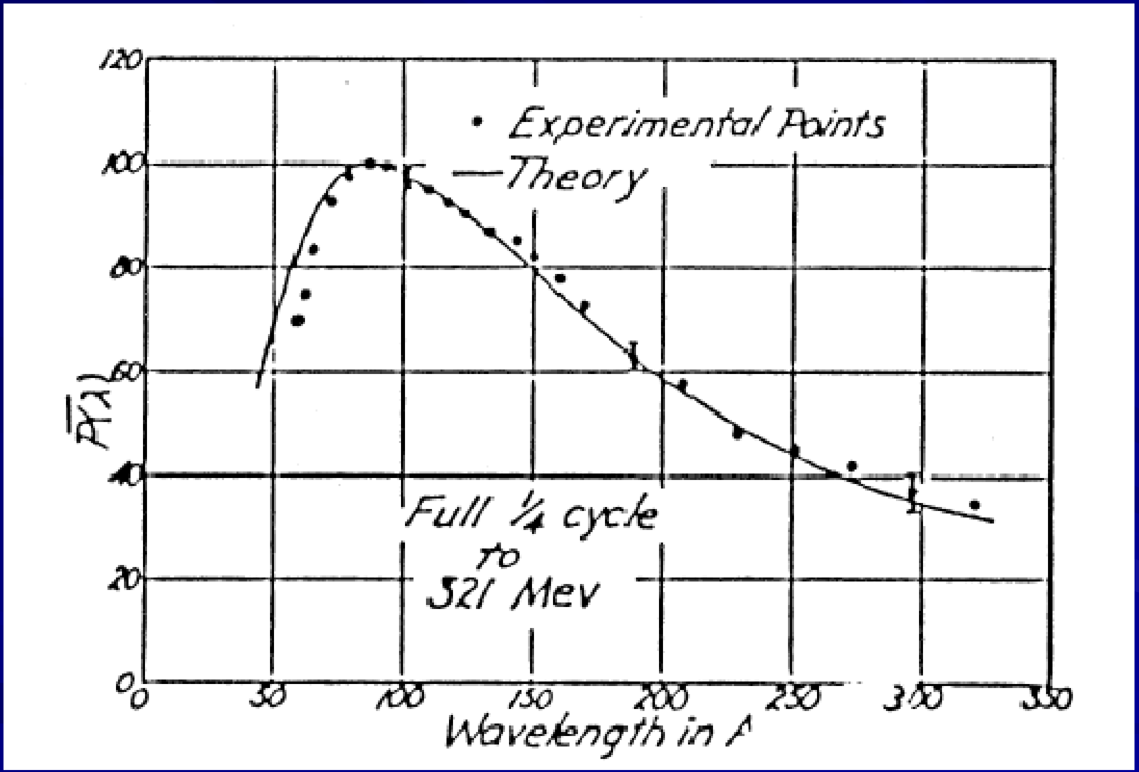


Fig.3 Characterization of SR in Soft X-ray

K.Codling and R. P. Madden: NBS 180MeV ES

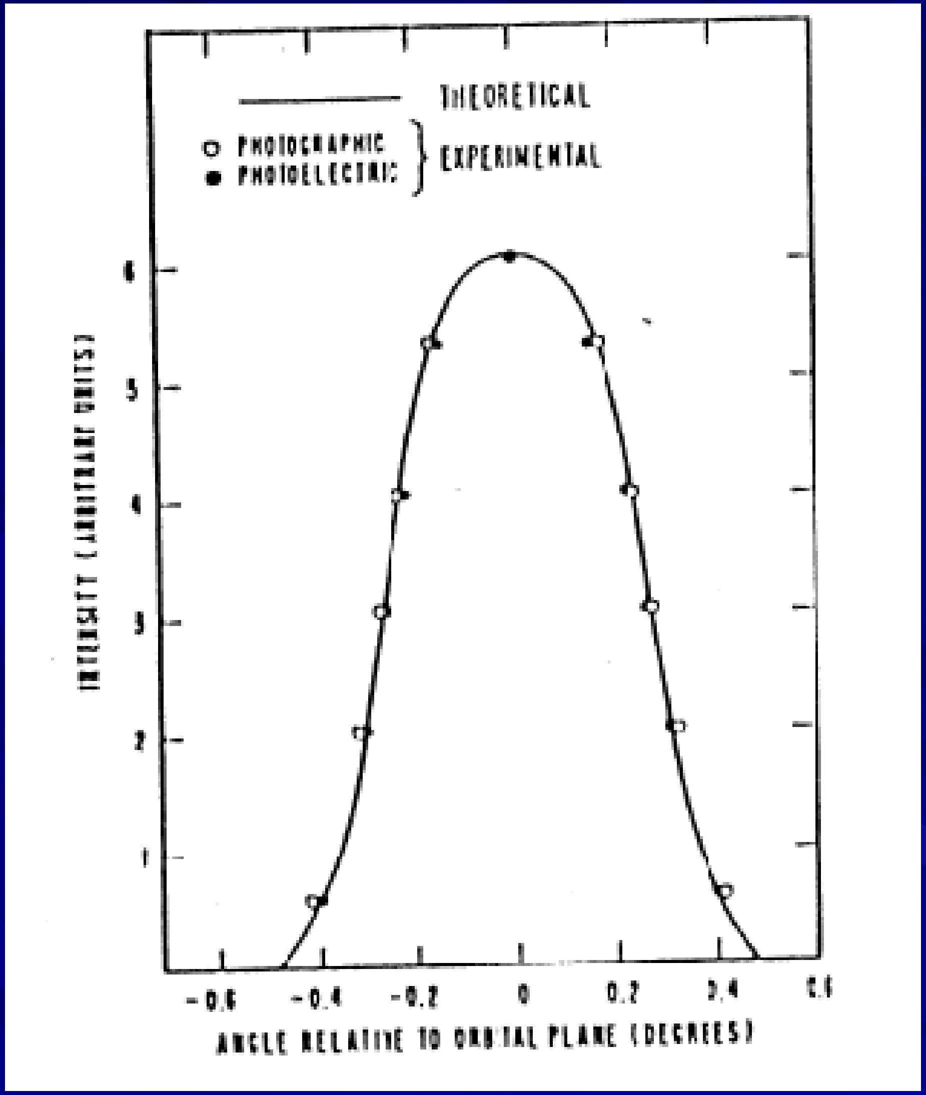


Fig.4 Polarization and Angular Distribution

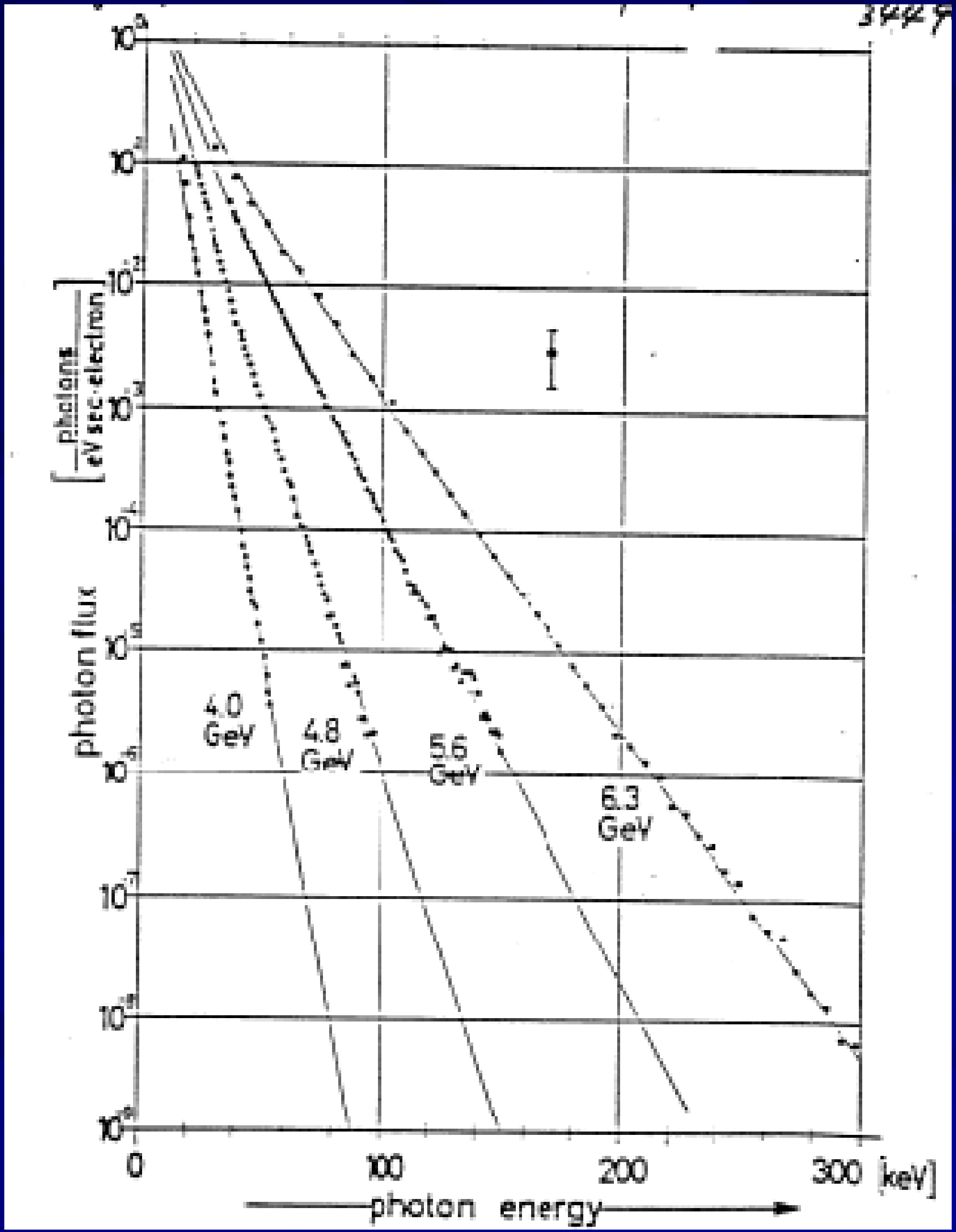


Fig.5 Spectra in X-rays

R.Haensel
DESY 6.6GeV ES

2. Parasites on Electron Synchrotron

Moscow University (Lebedev Institute),
  Moscow(0.56)
Deutsches Elektronen-synchrotron (DESY),
  Hamburg(6.6)
Roma University (INLF),
  Frascati (1.2)
Bonn University,
  Bonn (0.5)
National Bureau of Standards,
  Washington D.C. (0.18)
University of Tokyo(Institute for Nuclear Study),
  Tanashi(1.3)
Cornell University (CHESS),
  Ithaca(0.33)

Parasitic Access to SR——1960's

The First Generation of SR

I

Parasitic use of SR was subject to the following constraints

1. No initiatives allowed for parasites on

    1. Choice of energy
    2. Choice of current
    3. Choice of modes of operation
    4. Determination of operating schedules
    5. Access to the controlled area

    As a result, opportunities of performing any successful experiments are quite limited.

2. Electron synchrotrons are unstable in general. Why?

    1. Electrons captured on orbit vary every injection
    2. Electrons are injected in a low energy(e.g.10MeV), and then ramped up by increasing magnetic field. The orbits used to vary.

Fig.7 An example of the beam profile monitoring at INS

The energy dependence of the electron beam size.
The energy of electron is 100MeV (a),270MeV (b),520MeV (c),800MeV (d),900MeV (e),900MeV (f).

End of acceleration

← beam shrinking

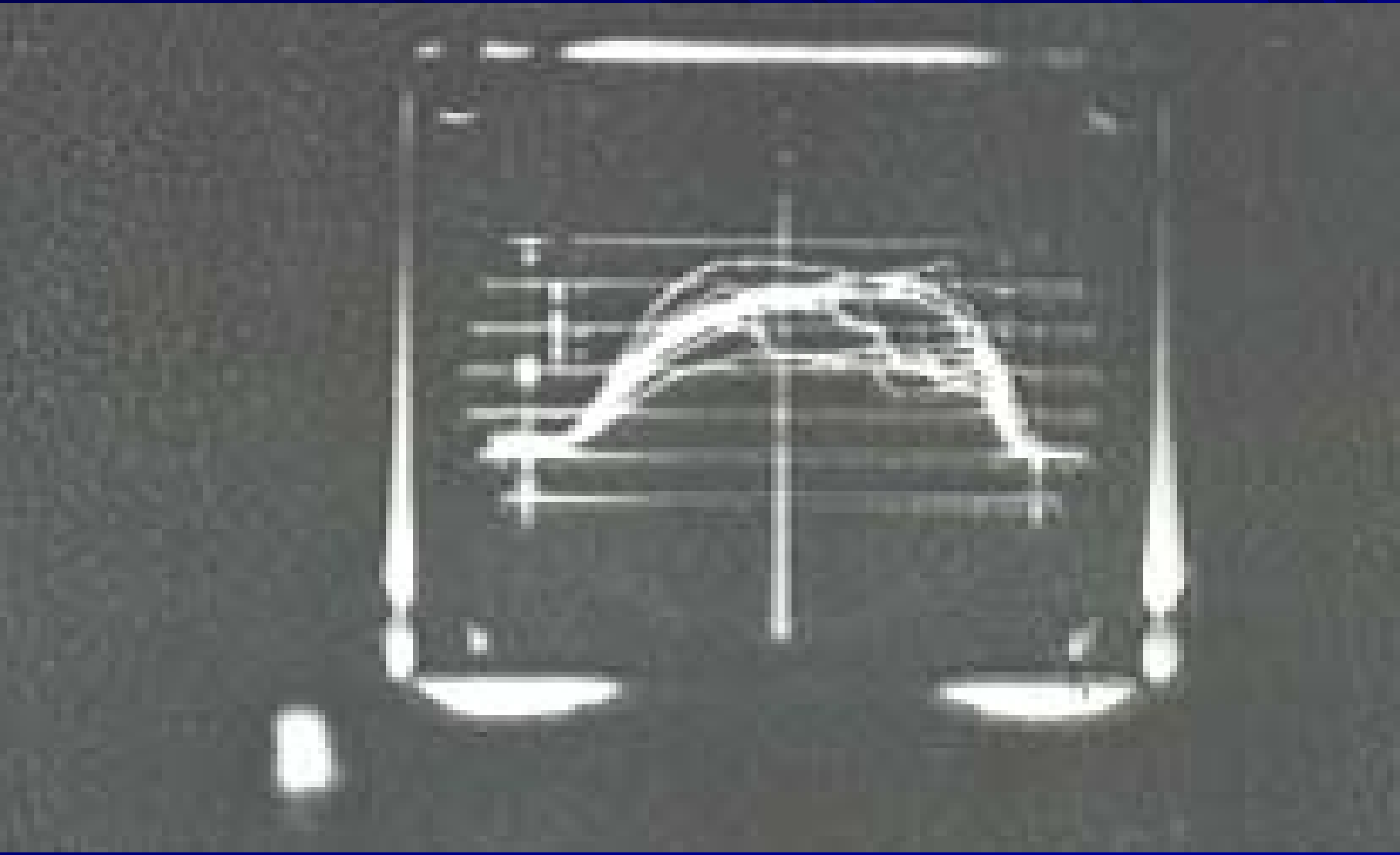


Fig.8 An example of the beam instability observation at INS

Parasitic Access to SR——1960's
The First Generation of SR
II

In spite of a number of difficulties, a few early spectroscopic experiments have achieved a lot of conspicuous success, and at the same time they demonstrated astonishing capabilities of SR both in its brightness and advantage of continuum. It was indeed commencement of the atomic inner-shell physics which led to the later dramatic development of atomic, molecular, and solid state physics.

It was definitely only one light source covering entire soft X-Ray region with a single source, then and now, perhaps forever. It met with the demands of spectroscopists perfectly.

The first exciting achievements came from NBS, the National Bureau of Standards, U.S.A. Robert Madden and his coworkers demonstrated with their small converted synchrotron a number of beautiful absorption spectra of rare gases, He, Ne, and Ar.

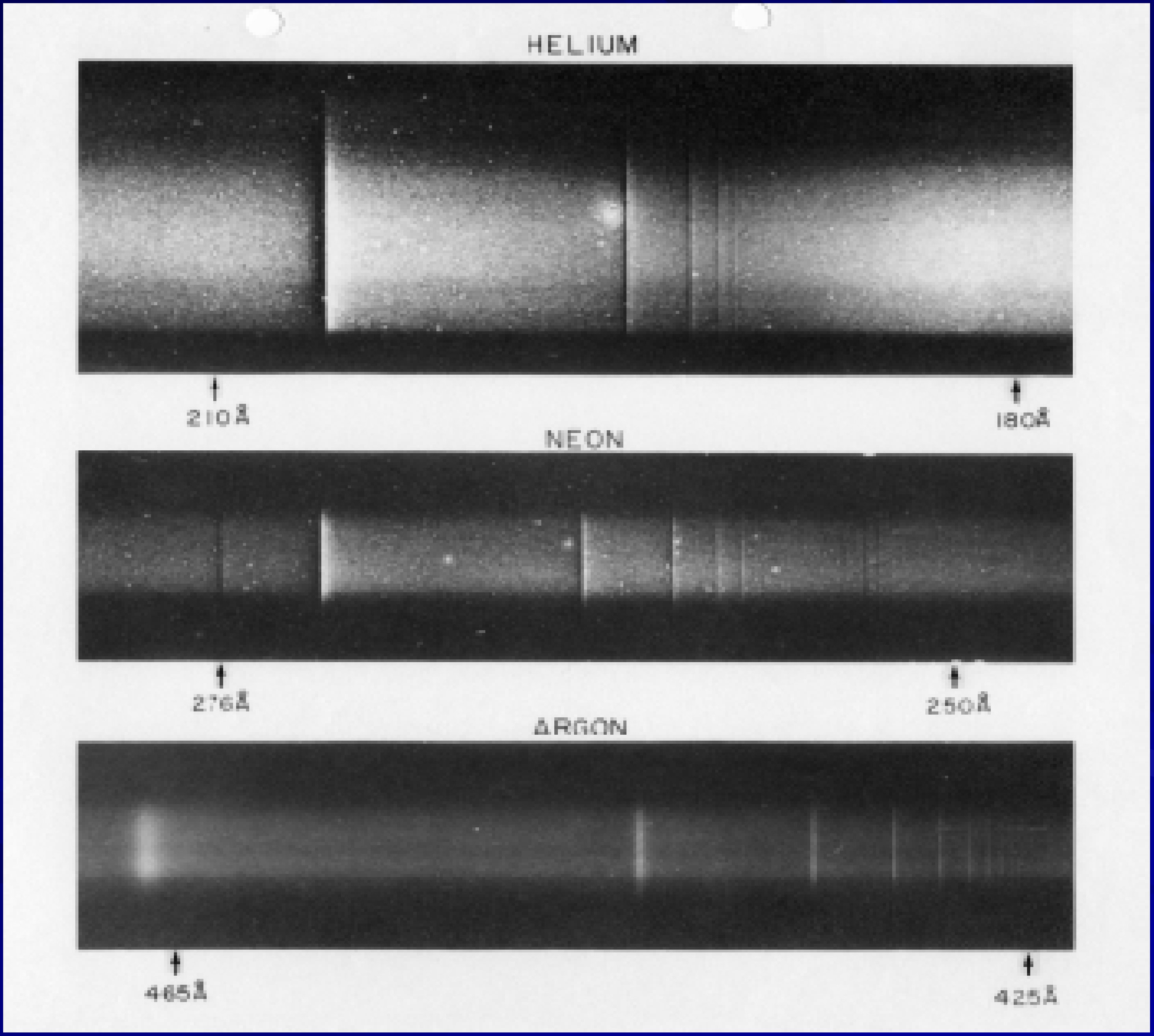
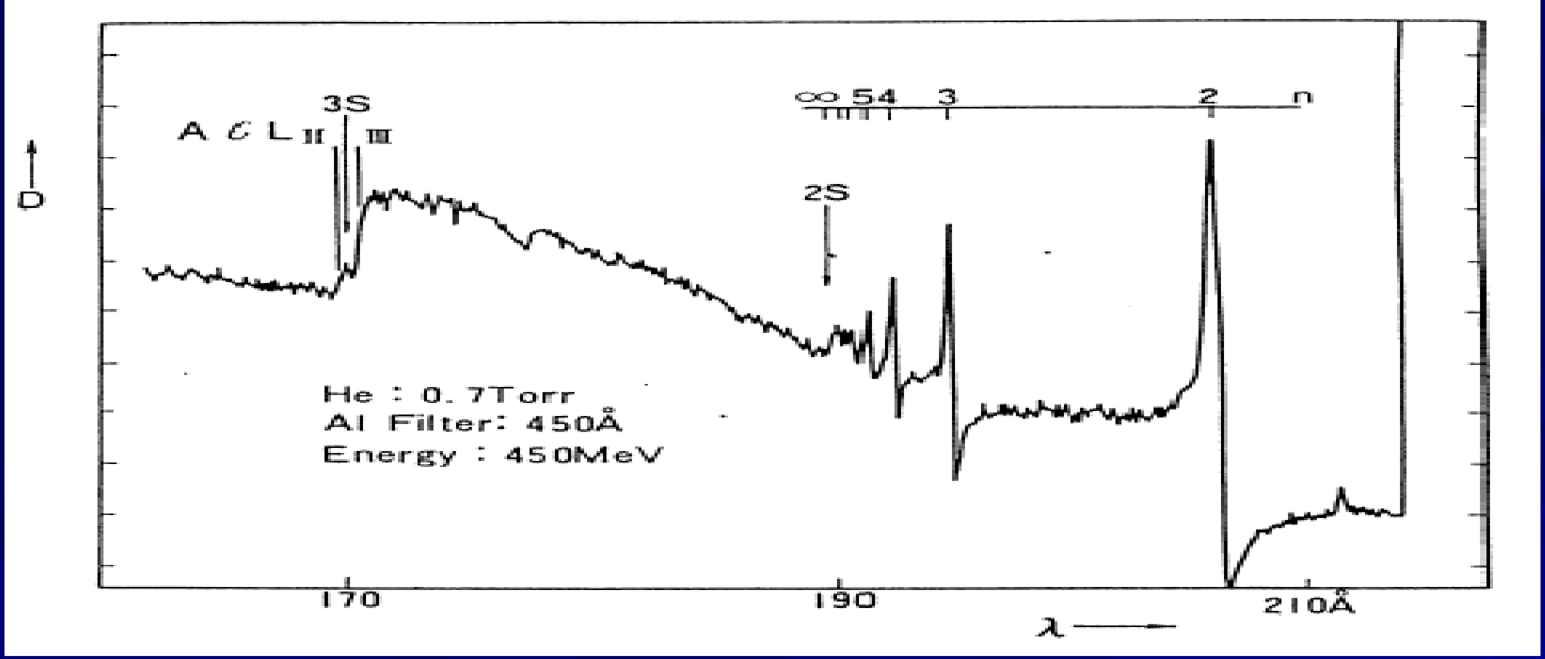


Fig.9 Absorption spectra of He, Ne, and Ar.



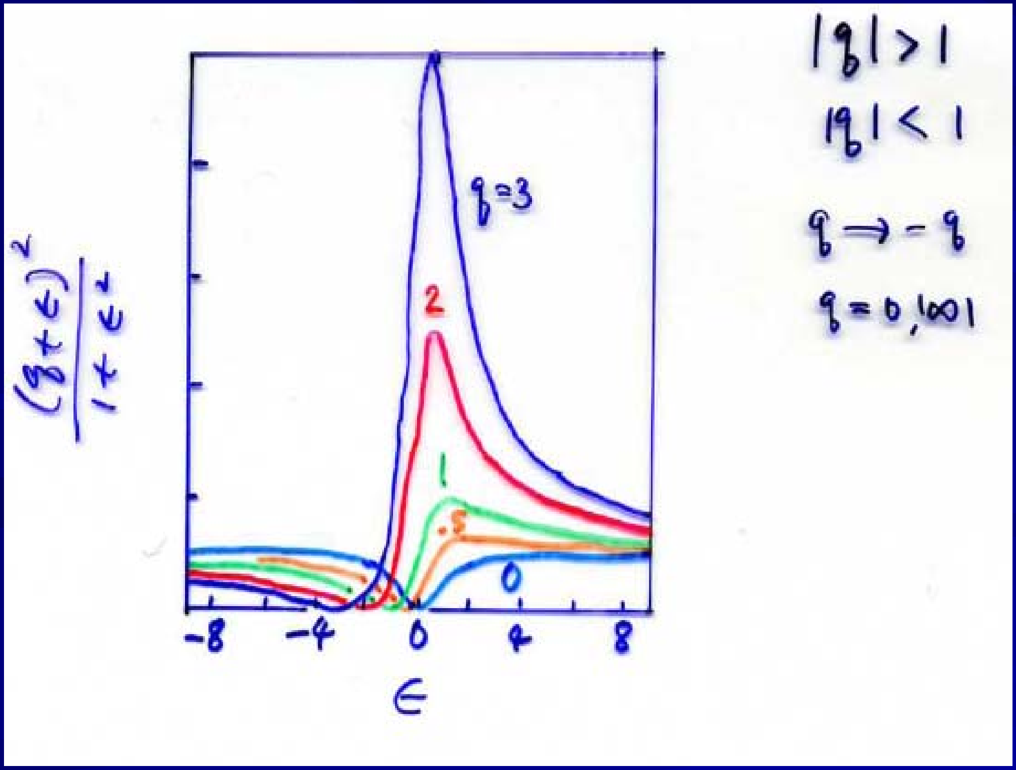


Fig.10 Configuration Interaction

Historical Background of Beutler-Fano Resonance

This strange resonance was reported earlier by H.Beutler in 1935 in absorption spectra of Ar. Ugo Fano, as an assistant of Enrico Fermi in University of Rome noticed this unusual profile of resonance, and was very much interested in looking for its origin. Fano discussed the problem with Fermi and reached to the common conclusion that this should be interpreted in terms of the configuration interaction between the bound states and the degenerate ionization continuum. The relevant theory was completed by 1961, when Fano was with NBS, and he wanted to verify this theory with experiment, first with electron spectroscopy, then finally with SR spectroscopy.

Fermi, the mentor of Fano escaped from Fascist Italy in 1938 on his way back from Nobel Prize awarding ceremony in Sweden, and became a refugee in Chicago. Fano followed as an immigrant to the United States next year.

Beutler, as a student of Professor Paschen in Berlin, had also to escape from Germany as he was Jewish, but Paschen tried hard to defend his excellent student, but the result was disastrous.

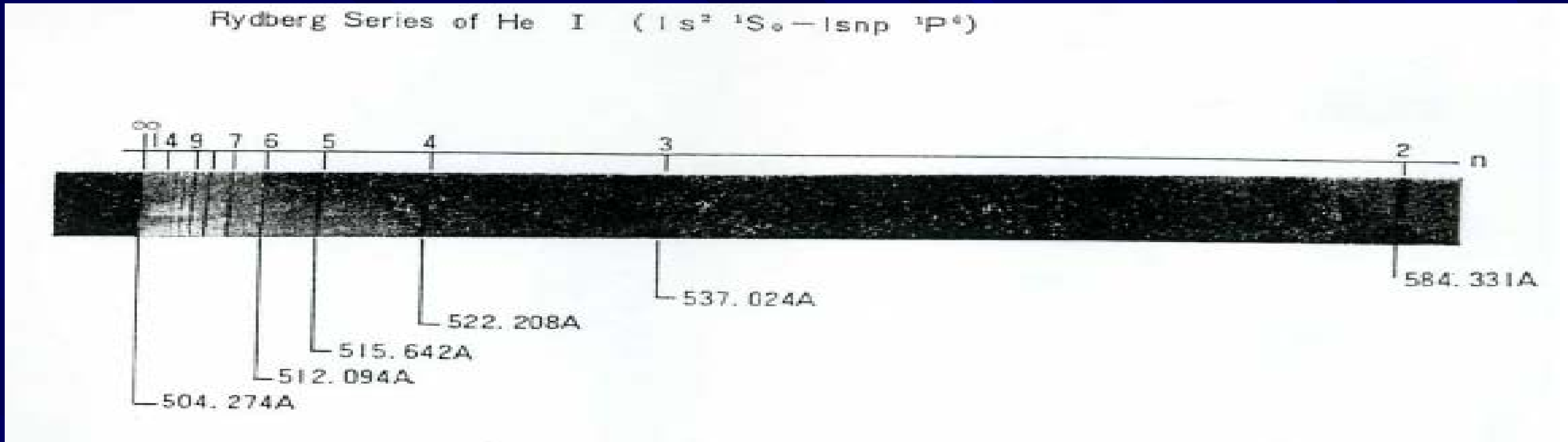
Beutler was able to escape from his urgent danger, but his courageous teacher was purged from the University and also from the position of the president of Physikalisch-technische Reichsanstalt(PTR), a German counter-part of NBS.

Beutler came to Chicago and died in poverty during the war. He left a manuscript of his comprehensive calculations on the theory of concave grating, which was published after his death in JOSA, and later regarded as the fundamental theory of optics of concave gratings. The theory of Seya-Namioka monochromator was also based upon Beutler's paper as the starting point. It should be remembered that the monumental experiment by Madden was intended to demonstrate the accurate profiles of "Beutler-Fano" resonances.

INS-SOR and DESY in 1960's

A group started preparation of parasitic use of INS Synchrotron since 1962, and set up a beam-line in 1963. The first successful measurement was made in March 1965, when NBS synchrotron was closed for the big move-out plan of entire Bureau from Washington D.C, to Maryland. Accordingly INS-SOR was only one active facility for SR for a while, until F41, a parasitic user group at DESY started in 1966.

Experiments were mostly photographic recordings of absorption spectra of atoms, molecules, and solids for studying inner-shell excitations. The energy of INS Synchrotron was higher than NBS, obtained spectra represented excitations from deeper inner-shells compared with NBS.



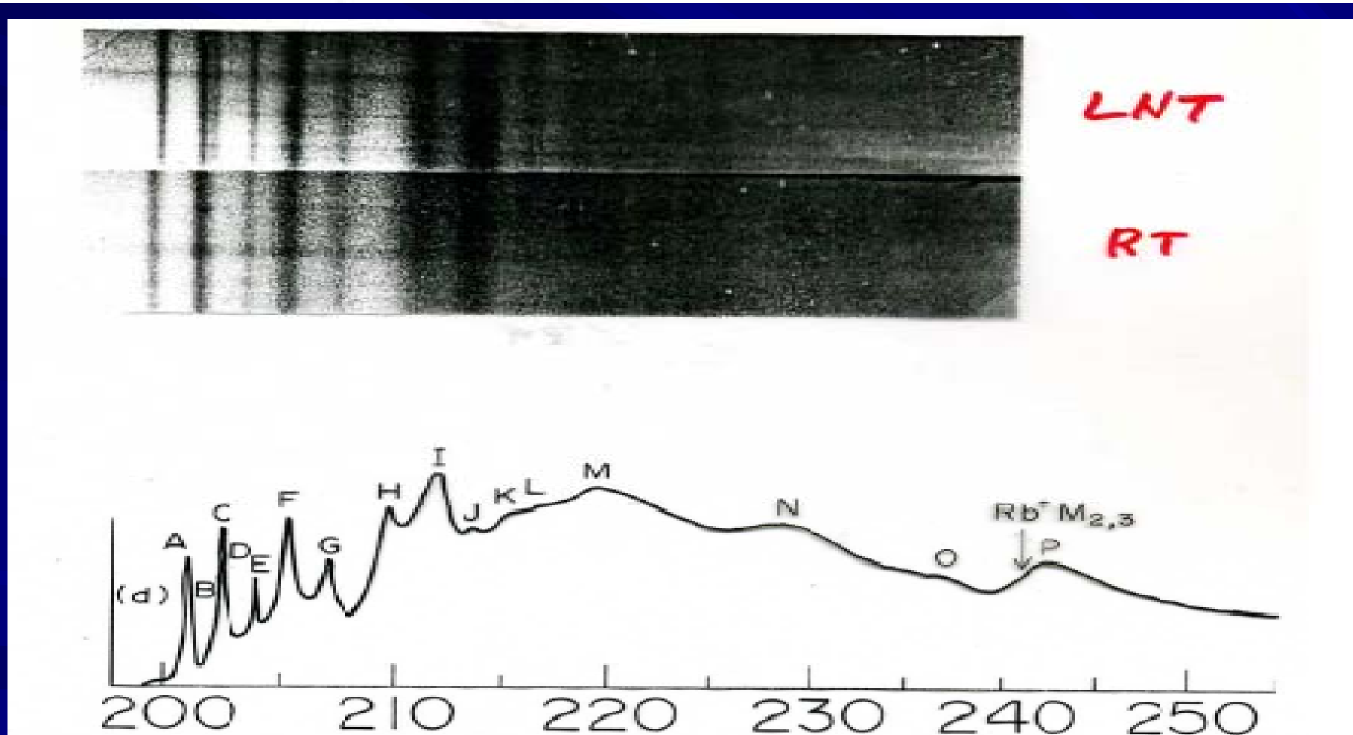


Fig.11 A few examples of early INS-SOR data.

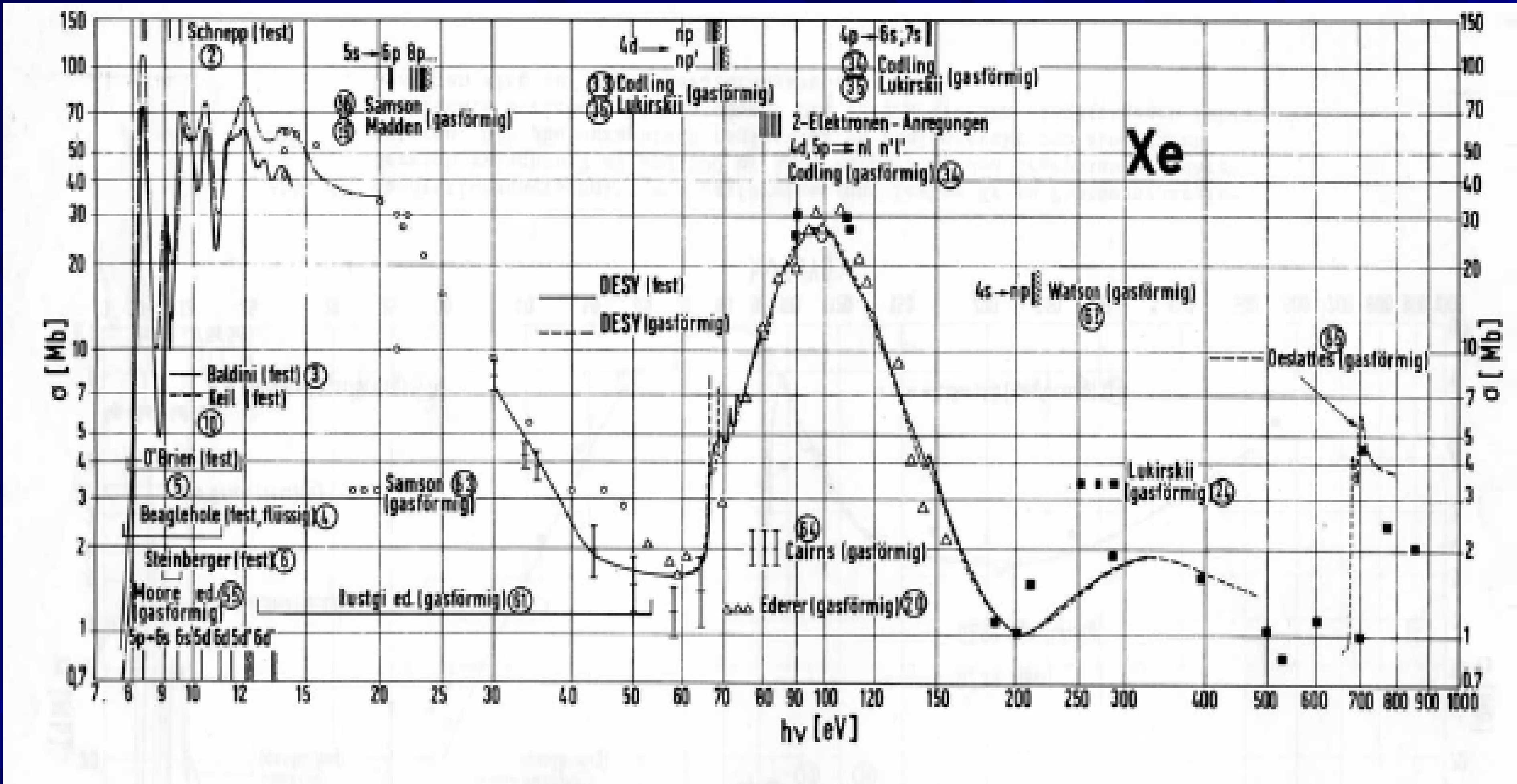


Fig.12 An example of early F41 DESY data

3. Dedicated SR Sources

The 2nd Generation of SR

Initial success of NBS, INS, and DESY, in spite of various handicaps of the parasitic status as machine users, stimulated strong demands for independent experimental facilities with electron accelerators as the light sources specifically designed for spectroscopy. Users circumstances soon deteriorated as the number of demanding users dramatically increased, and it also pushed requirements for dedicated sources.

INS-SOR proposed construction of a dedicated SR source by supplying electrons from the already running ES of INS in the fall 1965.The proposal had been approved in 1970 and a 300MeV electron storage ring was completed in late 1974. This was a unique machine designed and constructed not by accelerator experts but substantially by SR users only. After commissioning it was transferred to Institute for Solid State Physics, University of Tokyo, and made open for all the Japanese SR users since 1976, and ceased operation in 1997 when SPring-8 had just come into operation. The main body was transferred to SPring-8 and now exhibited in the next room here.

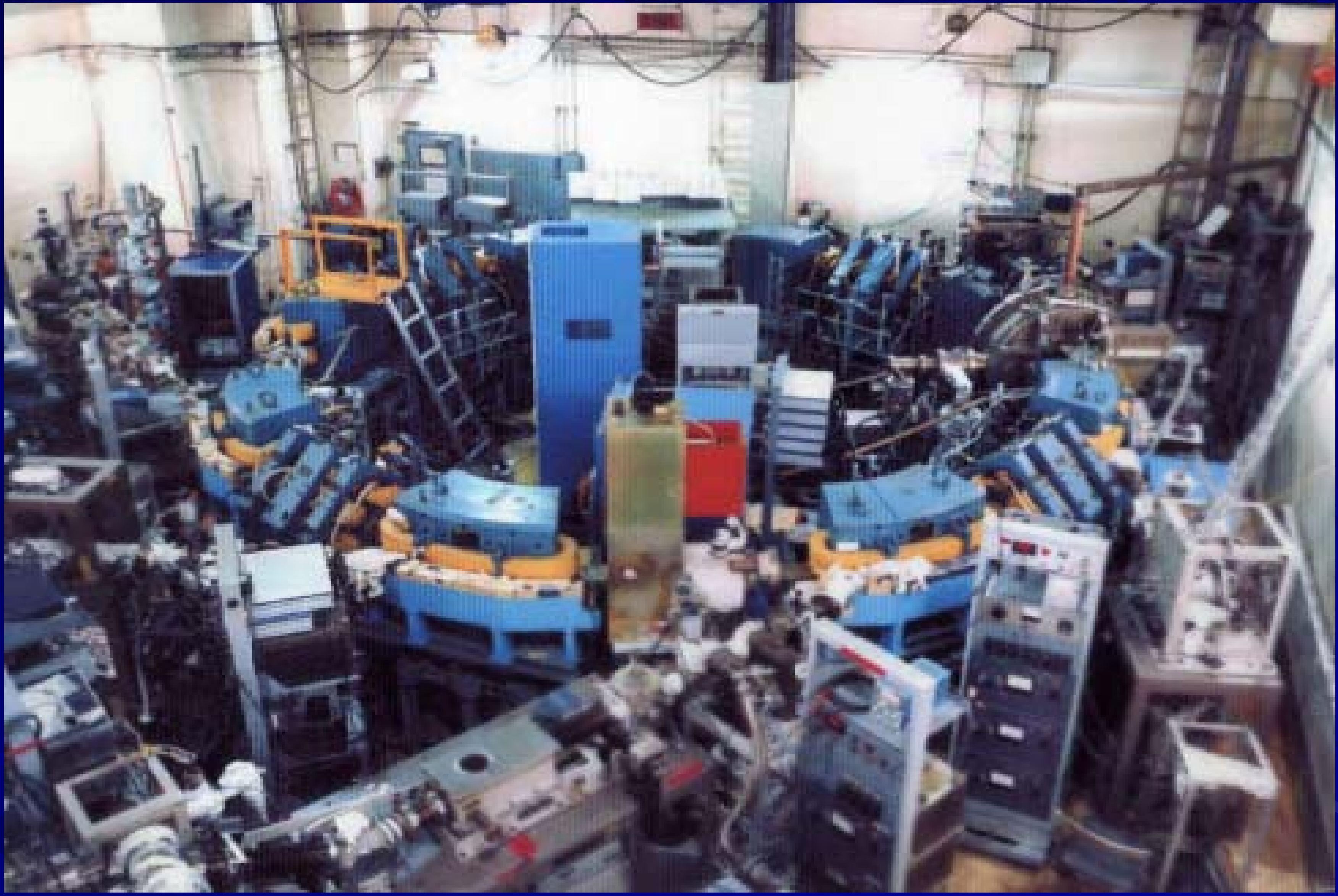


Fig.13 SOR-RING, ISSP

An electron storage ring for dedicated use was already running and made open to public at the Wisconsin University since 1968 prior to the commissioning of SOR-RING. But its design was not intended for SR use, as it was not an initial idea. For instance, it captured electrons by single injection only and never accumulated. By 1970's dedicated sources were below 1GeV, mainly for spectroscopic uses in soft X-Ray.

Storage Ring as the X-Ray Source

The idea of storage ring was originally proposed by Richter of Stanford in 1962 for achieving high energy by colliding electrons with positrons, whereas it took 12 years until he had finally built the machine SPEAR and discovered the charm-quark. The reason why it took so long was that there were persistent arguments against the idea. They were suspicious on that the useful cross sections would not be achieved with this technique.

We also recollect that there were arguments when the low-emittance storage rings as high brightness SR sources of the third generation were proposed. Some model calculations indicated that the low-emittance ring could not be operated stably, but finally it turned out that it was not the case.

Grau, treuer Freund, ist alle Theorie, Theories are all gray

The word of Mephistopheles is still true in contemporary physics.

After this rewarding success in Stanford, a competition in constructing colliders went on to the higher energy machines so that 5GeV class rings were sooner or later given less importance. SR users in the US and Europe began to attempt taking over these obsolete machines and converting them to the light sources.
SPEAR of Stanford, DORIS of Hamburg, ACO and DCI of Orsay, CESR of Cornell, and VEPP III of Novosibirsk were such cases. P. Hartman of Cornell recollects of these transactions as
“We, SR researchers, were the pirates”.
Only in Japan there were no treasure boat to take over, so that we had to build a ring from the beginning. Along with those converted sources, proposals for constructing dedicated SR X-ray sources were presented by SR users in Europe, US, and Japan.
These new sources were completed consecutively in years 1982 through 1986, as SRS of Daresbury, NSLS of Brookhaven, and Photon Factory of Tsukuba.

List of 2nd Generation Sources

1. Converted Source

SURF-I	USA	NBS	Gaithersburg
ACO	France	LURE	Orsay
DCI	France	LURE	Orsay
ADONE	Italy	INLF	Frascati
VEPP-II, III	USSR		Novosibirsk
NINA	UK	DL	Daresbury
TANTALUS-I	USA	Wisconsin Univ.	Stoughton
BONN-ES	W. Germany	Bonn Univ.	Bonn

2. Dedicated Source

SOR-RING	Japan	ISSP	1974	Tanashi
SRS	UK	DL	1981	Daresbury
TERAS	Japan	ETL	1981	Tsukuba
PF	Japan	KEK	1983	Tsukuba
NSLS-UV	USA	BNL	1983	Upton
UV-SOR	Japan	IMS	1984	Okazaki
NSLS-X	USA	BNL	1986	Upton
Aladdin	USA	PSL, UW	1986	Stoughton
Super-ACO	France	LURE	1987	Orsay

Triumph of Dedicated SR Sources

What resulted from the advent of dedicated light sources?

1. Independent control of the light source, experimental setup, and the entire facility free from the parasitic constraints.
   FREEDOM was the ultimate merit we achieved!!
2. Optimization of an accelerator as the light source

   Stability — temporal stability
   position stability
   effect of housing distortion
   instability hunting

   Brightness — higher current
   lower emittance
3. Novel opportunities brought about by higher source qualities.

Advent of the dedicated sources caused two great jumps. One is that this development stimulated a lot of novel research techniques and methods in broad fields of science, on the other hands it urged many innovations in accelerator technology. For instance, the control and measuring techniques, which are basis of the high stability of the source, and the theory of electron orbit which allowed achieving higher brightness. These efforts were all beyond undertaking if we were the parasitic visitors of a machine.

Emergence of SR X-Ray Sources not only created so many novel and promising research fields, but X-Ray crystallography was totally renewed in Its objectives and techniques.

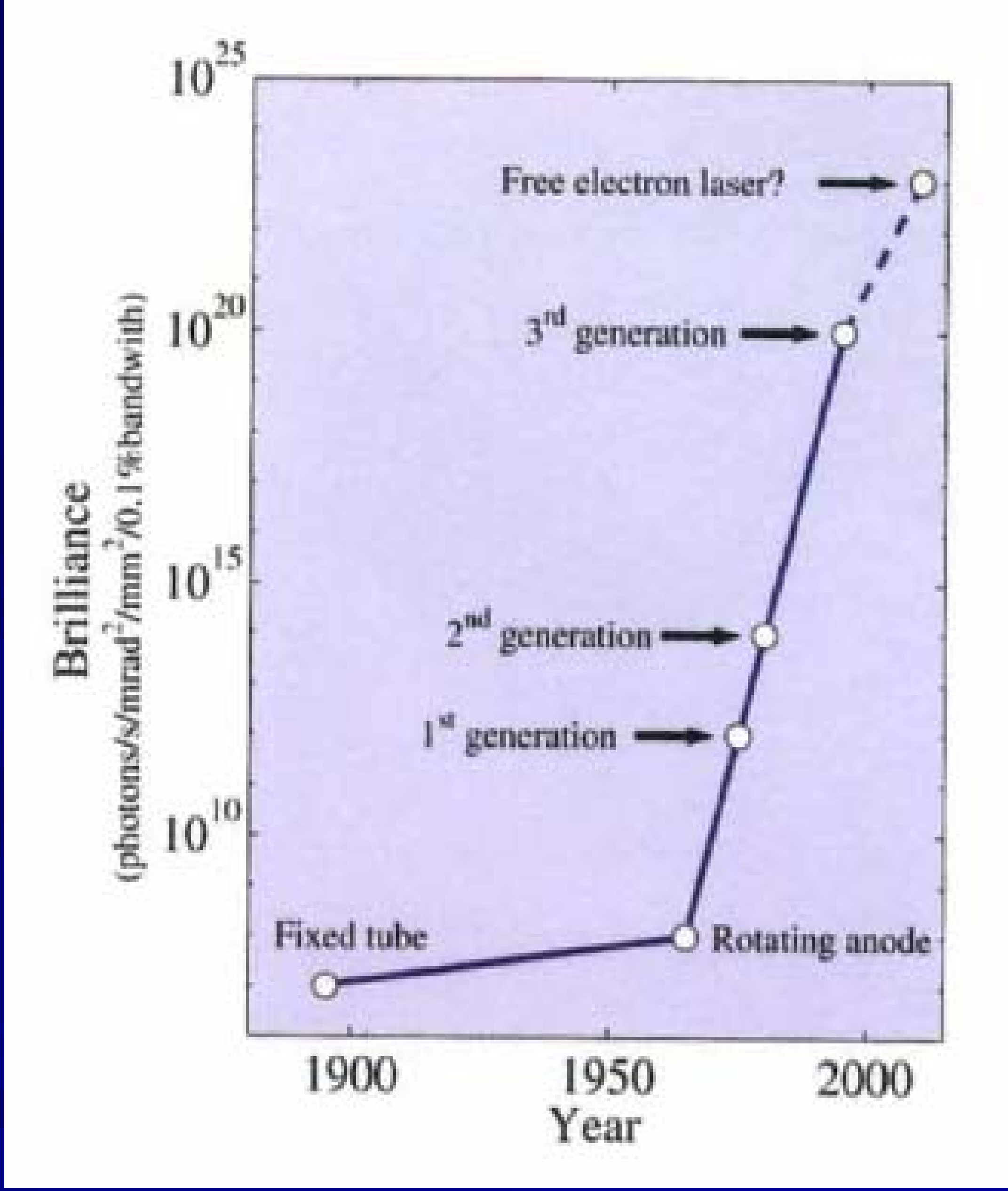


Fig.16 Progress in the Brightness of X-ray Sources(1895～2005)

Protein Crystallography

Protein is a category of very large, and extremely complex molecule with multifold steric structures.   Protein crystal structure analysis previously was a formidable time-consuming task, and it was regarded fortunate if he could determine two or three structures a life. Advent of SR changed the situation dramatically and knowledge of mankind on this extremely important substance is being accumulated with very high speed. A textbook published by Carl Branden, Swedish chemist in 1992 , "Introduction to protein Structure" proudly declared that the structure data of protein registered at the International Protein Data Base(PDB) reached 906.   However, it exceeded $2x10^4$ during next ten years according to the establishment of the 2nd and 3rd generation SR facilities and now it has reached to 45744@ 070911. Nevertheless the number of protein crystals of which structure determination is seriously demanded is estimated to 100 million or even more, so that our current knowledge is seriously insufficient.

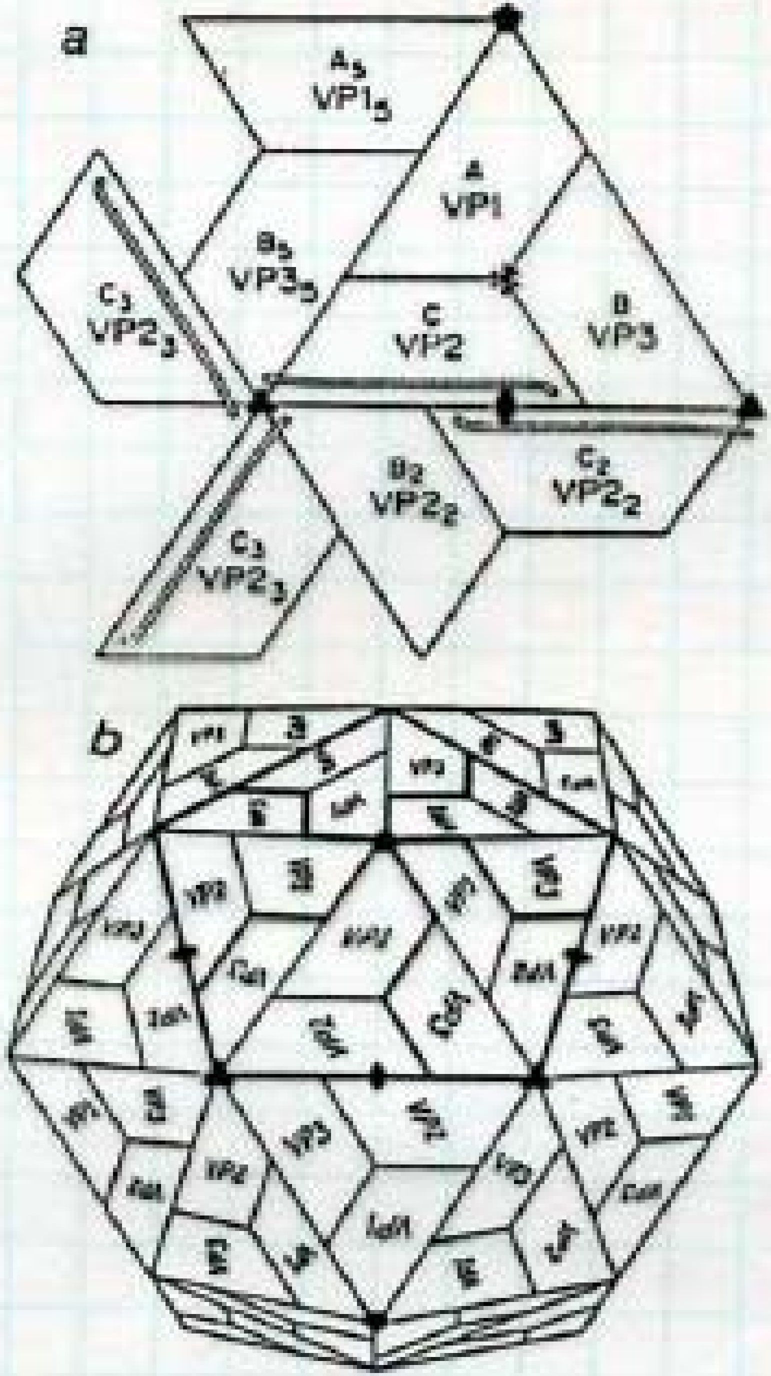


Fig.17 Structure of HRV14 Virus

EXAFS

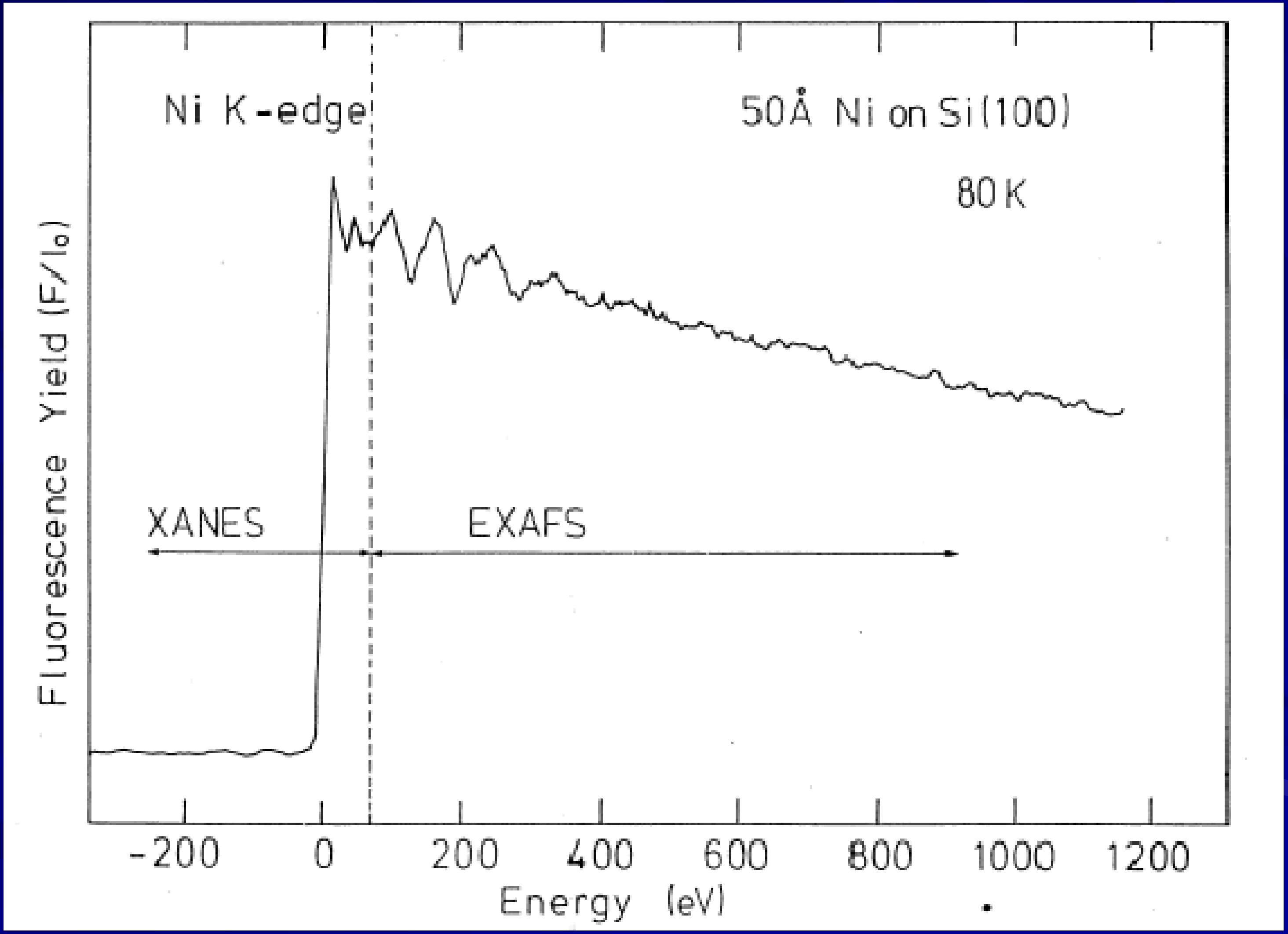


Fig.18 An example of EXAFS data analysis

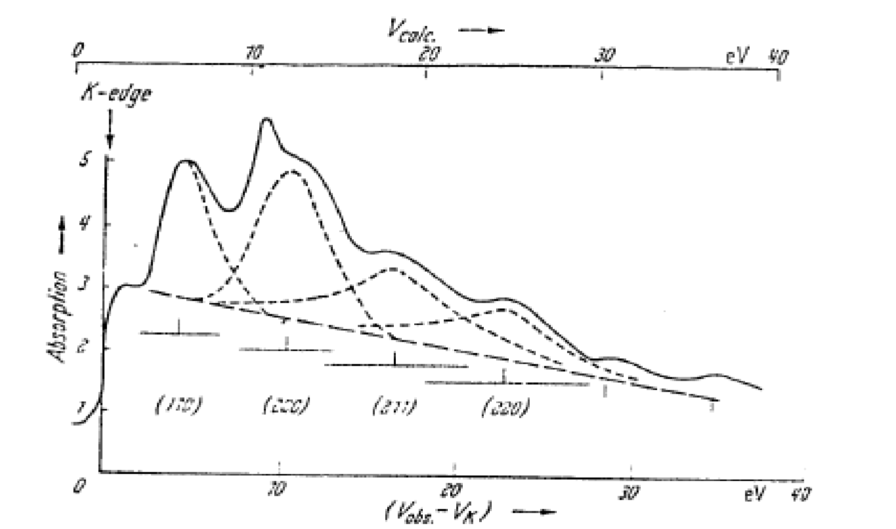


Fig. 34. Secondary structure on the high energy side of the Li K absorption edge (after HAYASI, see reference on page 298). The dotted curves show the individual absorption bands whose peak energies are calculated from HAYASI theory. Solid horizontal lines correspond to the half-widths of the quasi-stationary states.

Fig.19a Tomboulian's data on Li

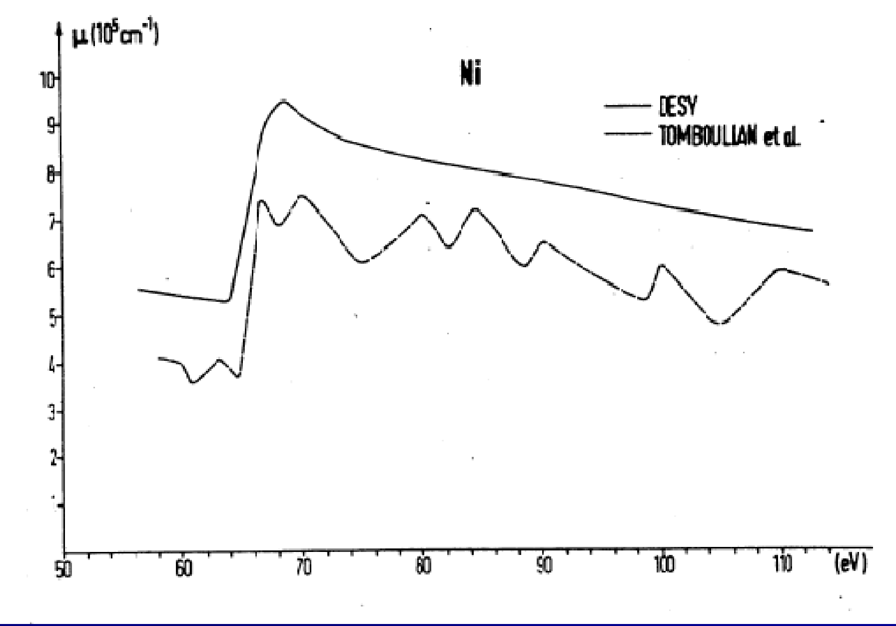


Fig.19b

Sonntag's data on Ni

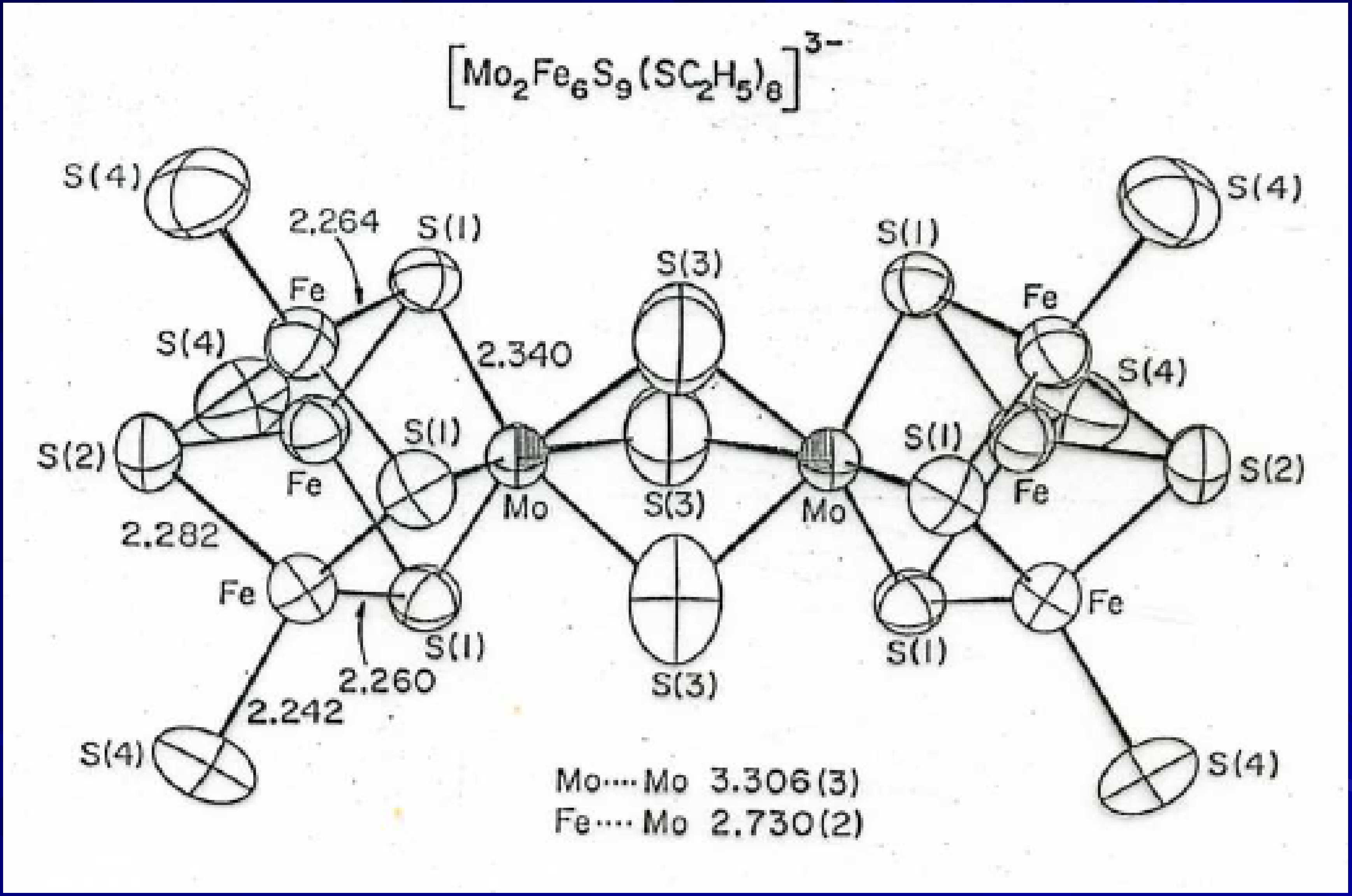


Fig.20 Structure of the active center of Nitrogenase

This is the active center of the enzyme Nitrogenase, with complicated arrangements of Mo, Fe, and S atoms. Many crystallographic determinations of the structure were attempted earlier, but no reliable conclusions were reached, while EXAFS led to this complex structure unequivocally. They analyzed the absorption spectra above K-edges of S, Fe, and Mo, respectively, then inter-atomic distances to each other were finally assessed consistently.
EXAFS is well adapted to the structure studies of disordered arrangements, surface and interface structures, elements of low concentration, which previously out of reach with the diffraction crystallography, thus the both techniques are complementary to each other.

Structure Studies under Extreme Circumstances

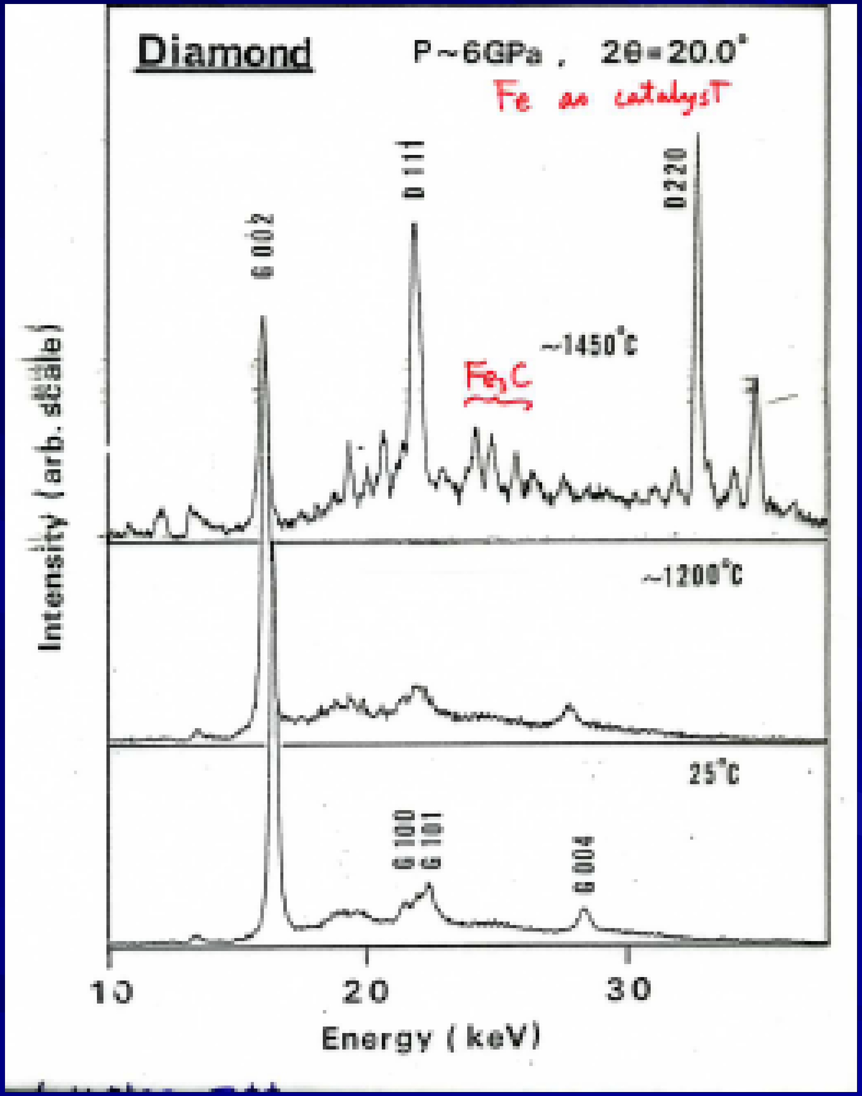


Fig.21 Phase transition of graphite to diamond. PF 1984

A few Unhappy Disappointments Lithography and Angiography

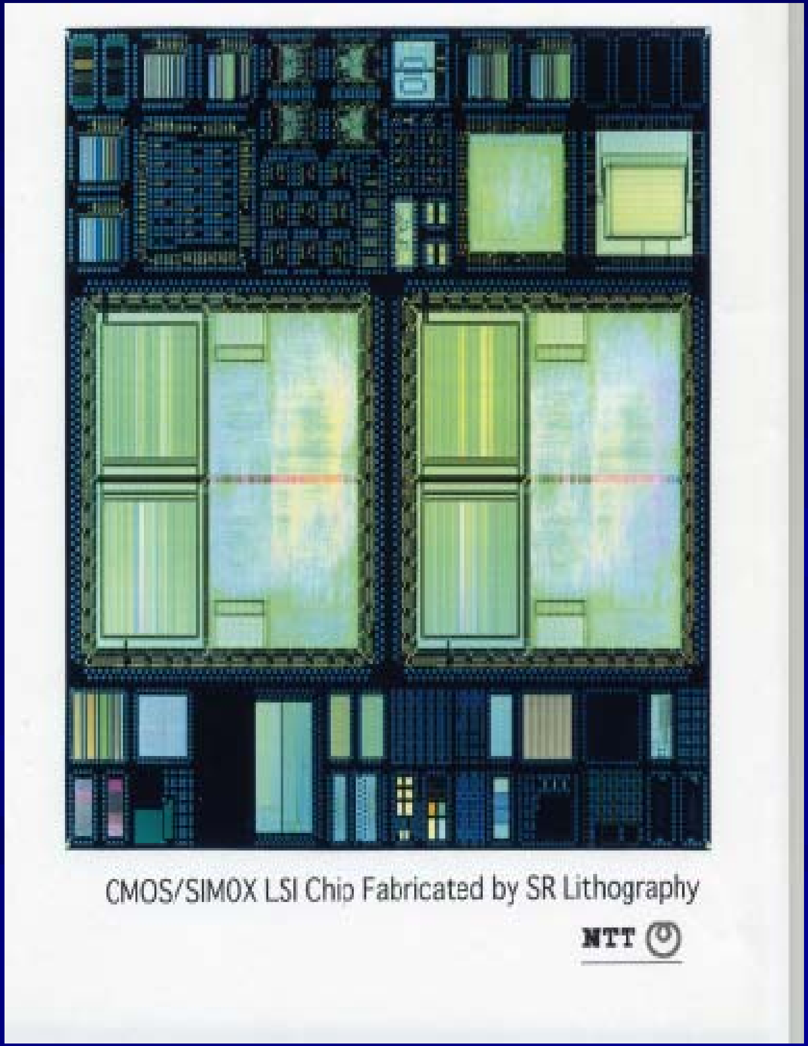


Fig.22 SR-based circuit

SR lithography for manufacturing the semiconductor microchips as the circuit components of computer hardware was attempted since the successful trials by DESY in 1974 and INS-SOR in 1976. It generated a big dream that some day whole semiconductor device industry may shift to the SR lithography. Big investments started in Germany, England, US and Japan for constructing compact storage rings to be incorporated within the clean room of microchip production. A number of rings of fancy design had been completed and commissioned successfully, but the techniques of optical lithography based on excimer lasers made a big progress and won the competition simply due to the superior cost-performance. Consequently nearly all these efforts of SR lithography were forced to retire. Thus the unused machines were finally converted to meet the other purposes, for instance, Helios II constructed by Oxford Instruments was finally transferred to Singapore for the academic use, SORTEC, the ring of Japanese government-assisted consortium, went to Thailand as as a national facility, AURORA, the world-smallest ring of Sumitomo, went to Ritsumeikan University for the public use.

SR angiography was another effort ended up in failure after many year's intensive trials.   This was also considered a promising medical technique for diagnosis of the coronary artery, and in fact it was proved the quality of image was greatly improved compared to the conventional X-ray angiography, but once the practical applications were attempted, it turned out the entire operation including the medical staff and equipments of diagnosis and surgery cost amounts very high.   DESY had once established the ad-hoc facility of SR diagnosis combined with the university hospital team of surgery, and accepted several hundred patients.   Operation itself was regarded as success, but both the hospital and DESY authorities finally judged it didn't pay. They closed the facility and their partially successful operation was finished.

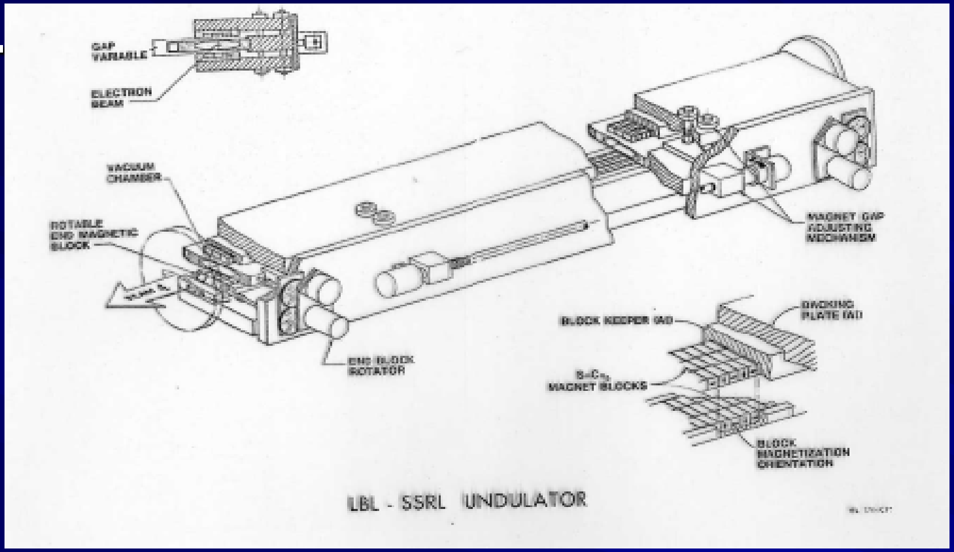
4. Advent of Insertion Devices

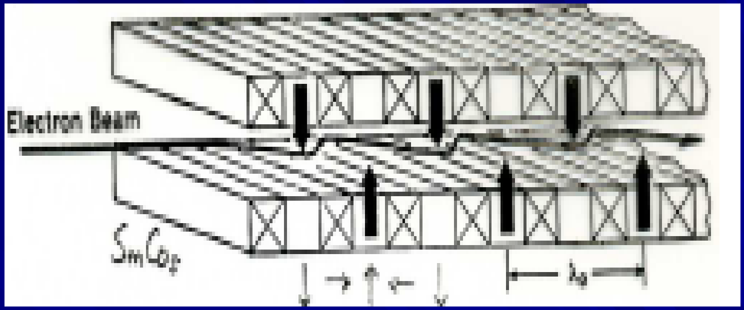
The first idea of undulator was proposed in 1947 by Vitaly Ginzburg, the Nobel Laureate of Physics in 2003, as pointed out by E. Koch.

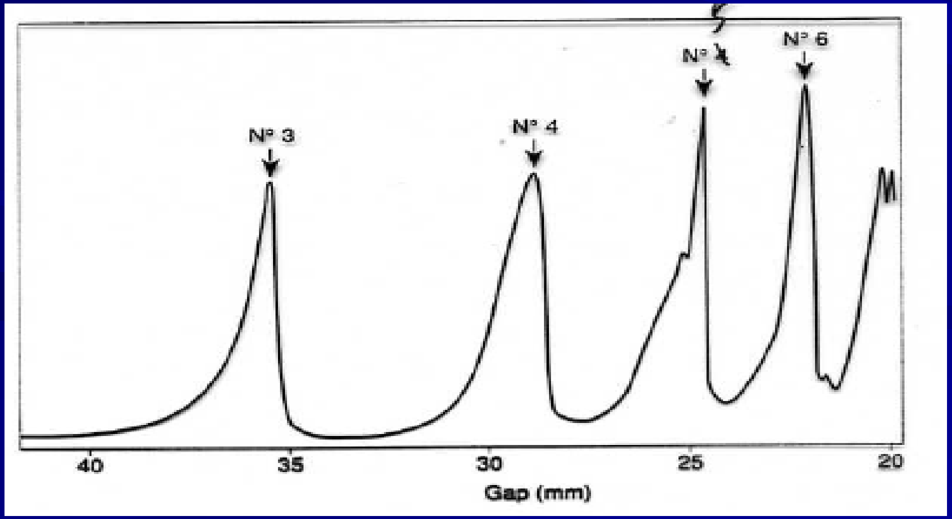
The first demonstration of undulator was made by Hans Motz at Stanford in 1953. He used a magnetron magnet array of 50 poles as the periodic undulator magnet and the electron beam was supplied from 3~5 MeV Linac. His intension was development of sub-millimeter wave generator with this scheme, and it generated 1W output successfully. However, this device was a little too big to be a practical emitter of sub-mm wave, and was soon overtaken by semiconductor diodes so that further efforts were abandoned.

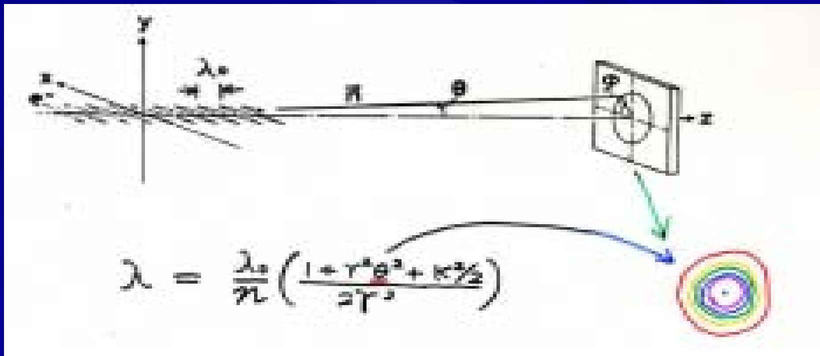
The revival of this idea came from a successful experiment of free electron laser based on the double helix of superconductor coil by a team of Berkeley in 1976. This approach was soon applied as an insertion device in the storage ring ACO by a joint French-American team, but it turned out that the liquid helium based device was not compatible with the storage ring.

A decisive progress in undulator development came by introduction of a permanent magnet design proposed by Klaus Halbach and Herman Winick in 1979.







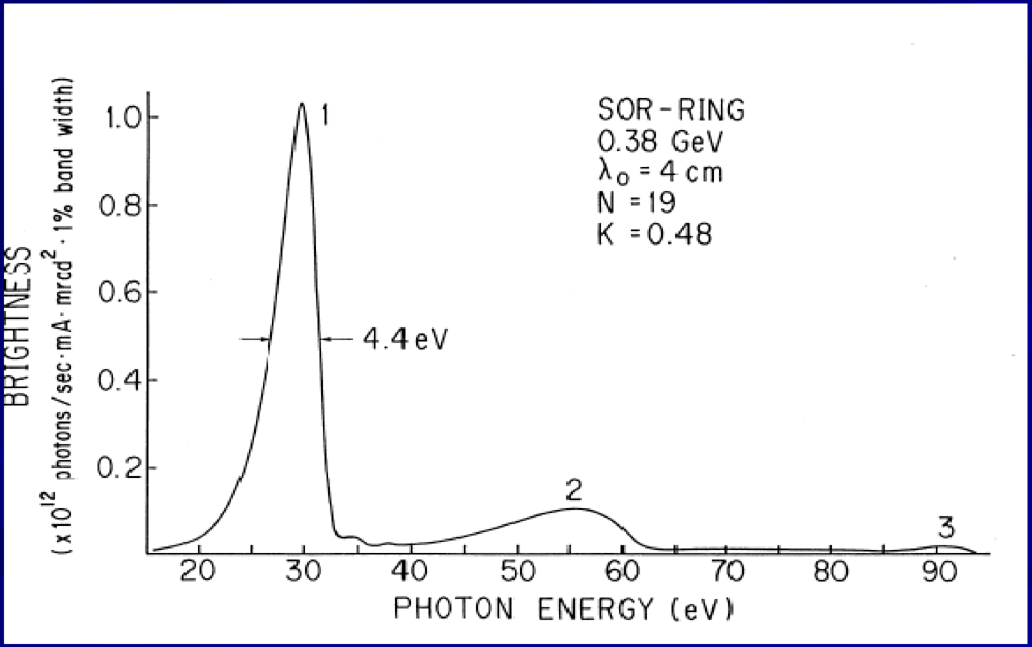


Characterization of UR

A joint team of PF and University of Tokyo followed the Halbach-Winick design and tried the verification of UR formula

$$\lambda n = (\lambda_0 /2n\gamma^2)(1 + K^2 /2 + \gamma^2\theta^2)$$

where $K = \psi/\gamma^{-1} = eB_0\lambda_0 /2\pi m_0\, c = 0.934\lambda_0\,(\mathrm{cm})\; B_0\,(\mathrm{Tesla})$ is the maximum deflection normalized by the characteristic angular spread of SR or γ^{-1}, and λ_0 is magnetic field period, n is the harmonic number, and θ is the angle of observation against the mean electron velocity.



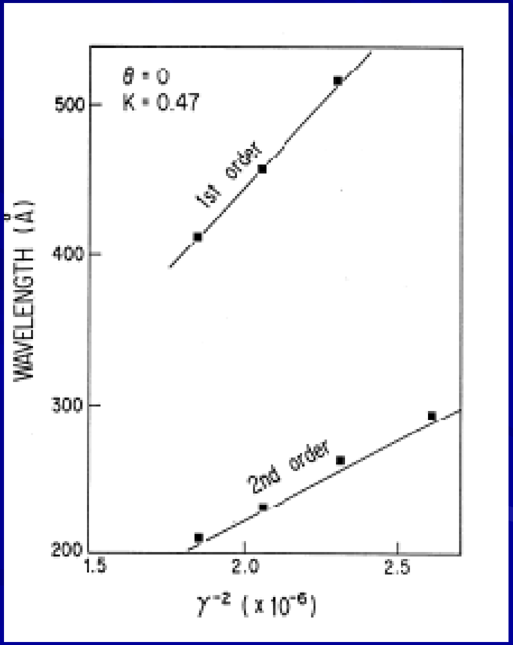
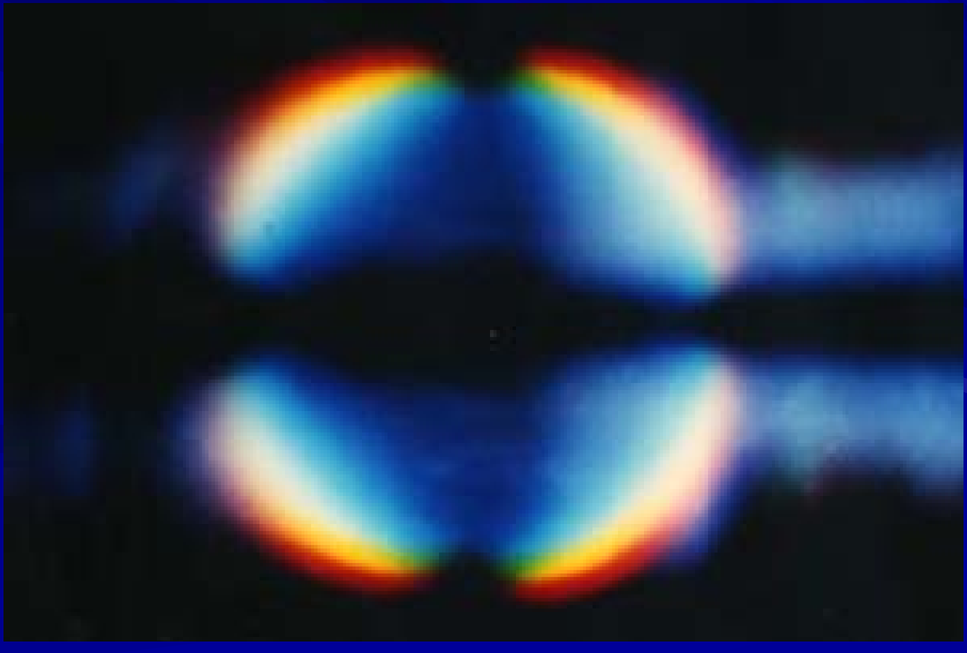


Fig.24 Characterization of UR, SOR-RING 1980



The test was performed December 1980 and the results are shown in Fig.24 It should be noted that apparent peaks in the observed spectra shifted to lower energies consistently. λn of the formula are not the apparent peak of the curves. This shift was caused by a relatively large emittance of the electron beam. The formula assumes the low emittance limit.

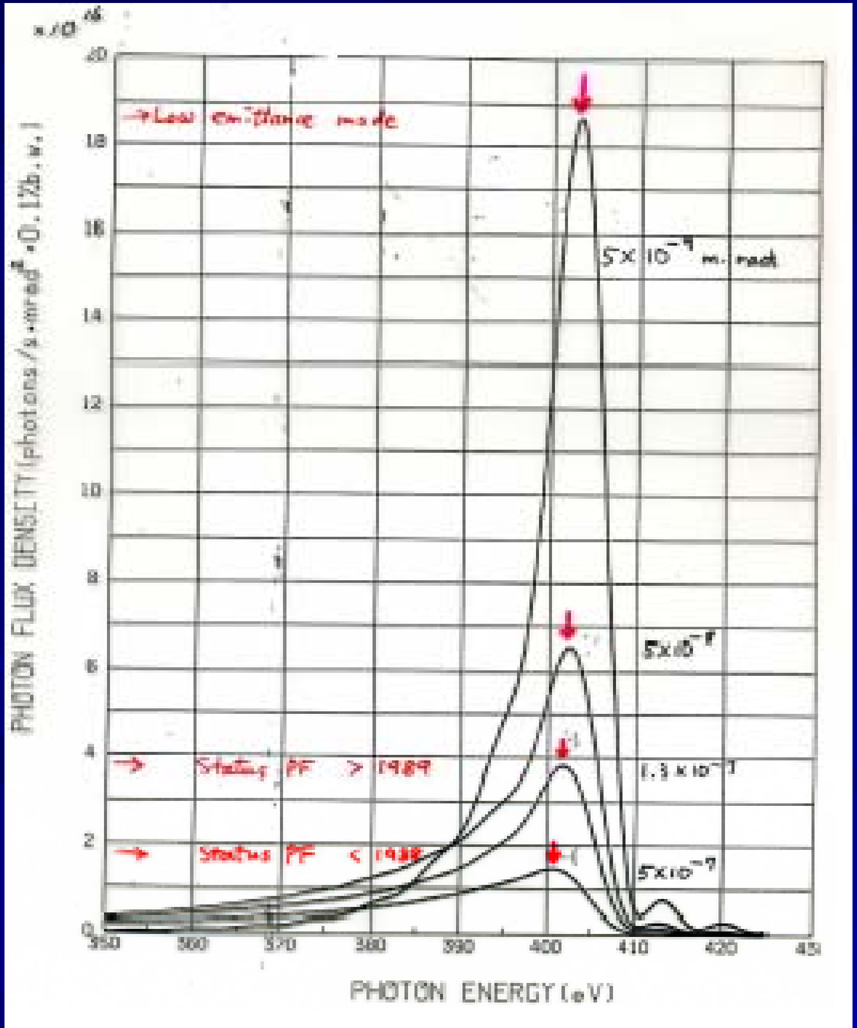
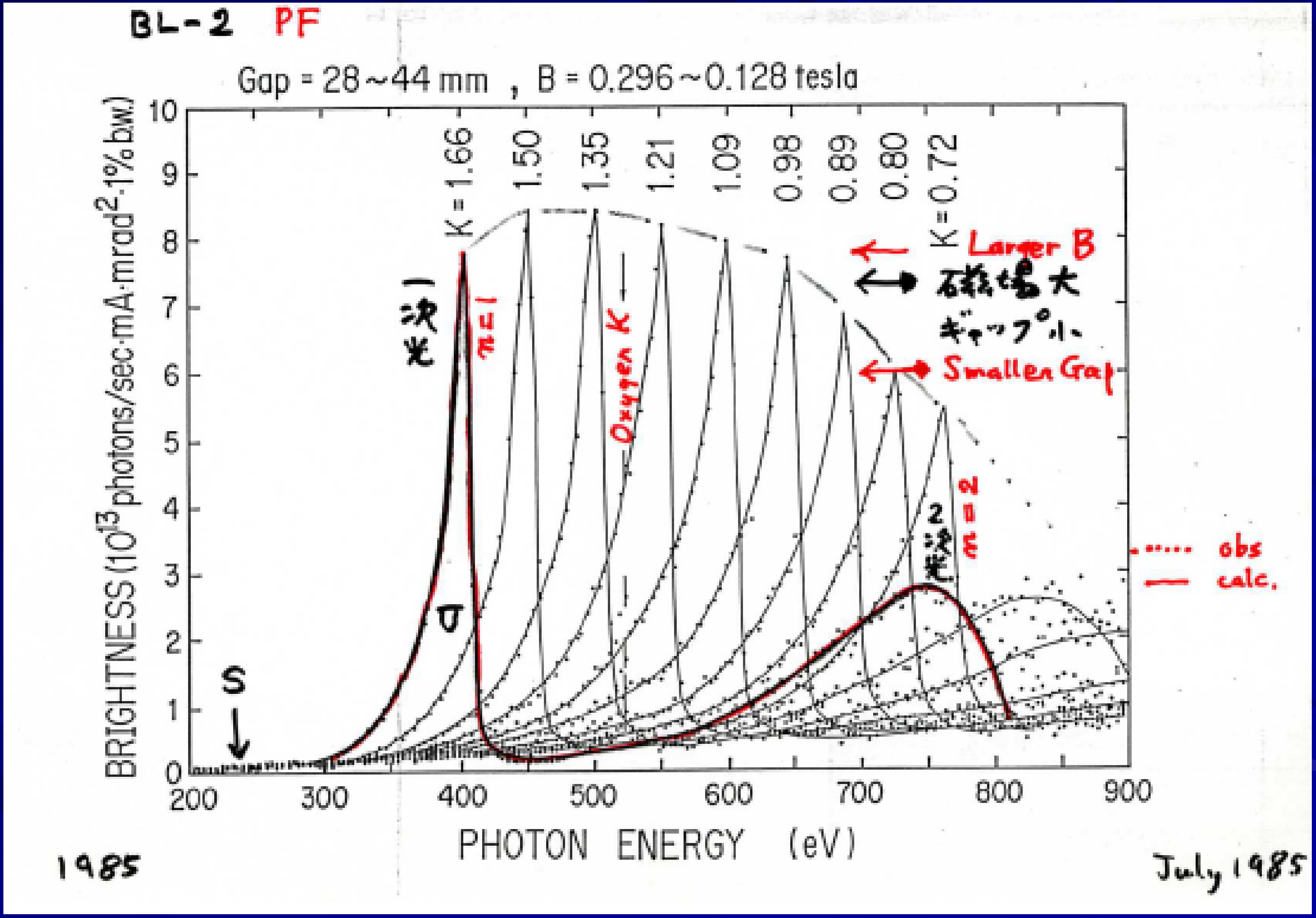


Fig.25 Effect of emittance on the profile of UR. H.Kitamura, 1980.

Effects of finite emittance are the peak shift to lower energy, reduced peak brightness, and increased width. It should be noted, however, that in spite of those negative effects of emittance, the brightness enhancement was dramatic. It was confirmed that two or three orders of magnitude enhancement was easily obtained even with such unfavorable condition.

The experiments for characterization of UR were made by measuring photoelectrons emitted by rare gas targets of known cross section of ionization. Therefore the intensity was given in the absolute scale so that the quantitative comparison with the theory was made possible.

A further efforts of characterizing UR was continued at PF at the first inclusion of a practical undulator into BL-2, PF. It was a 60- period array with 4cm period, and the critical wavelength of the first harmonic was 3nm. Similar measurements have been repeated and the degree of polarization was carefully studied by measuring the angular dependence of photoelectrons emitted from helium.



BL-2 PF
Gap = 28~44 mm , B = 0.296~0.128 tesla
BRIGHTNESS (10^13 photons/sec·mA·mrad^2·1% bw)
PHOTON ENERGY (eV)
K = 1.66
1.50
1.35
1.21
1.09
0.98
0.89
0.80
K = 0.72
1次光
n=1
Oxygen K
2次光
n=2
σ
S
Larger B
磁場大
ギャップ小
Smaller Gap
obs
calc.
1985
July 1985

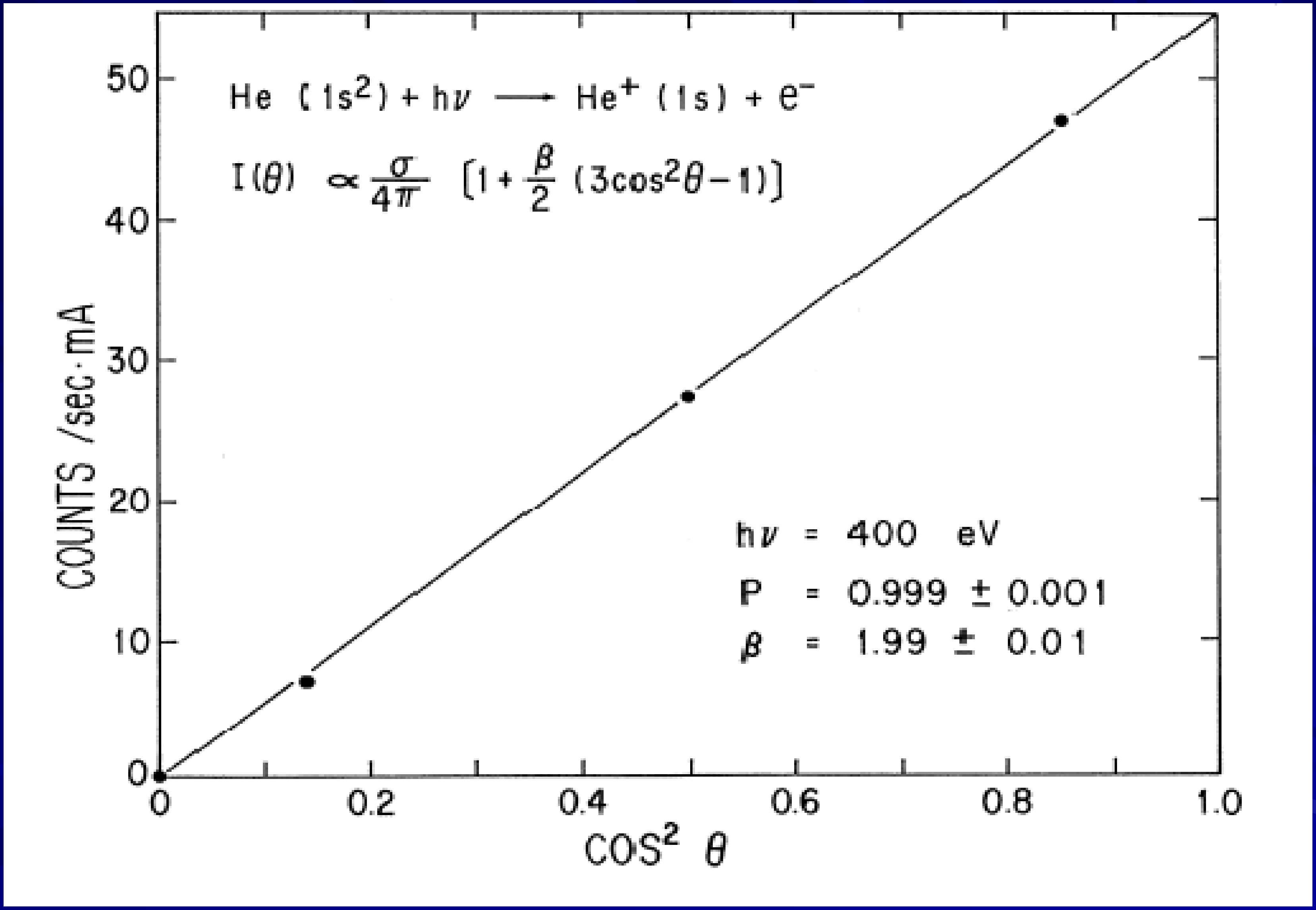


Fig.26 UR from BL-2 PF, Polarization measurement

The results of characterization was published in 1985 at SRI-2 in Stanford, and its scientific value was confirmed by the users community. There were some concerns about the validity of the theory in SRI-I in 1982, but it was no longer an issue of debate in 1985. Permanent magnet undulators of Halbach-Winick type have been quickly developed and insertion started everywhere, including US, Japan, Germany, and France, and Russia.

A big dream of getting the next generation light sources with dramatically improved brightness had grown immediately in Europe, America, and Japan. It was the start of a next competition for the high-brilliance X-ray source with undulators as the main feature of the machine.

Undulators for generating circularly polarized radiation have been developed worldwide, and many promising fields of application such as the magnetic circular dichroism, etc. have been intensively pursued since the 2nd generation SR, but the devices are full of variety and complex so that it would take a lot of space and time to describe them in full account. The subject is accordingly omitted in the present lecture due to the limited time.

5. Motivations for the 3rd Generation SR

The 3rd generation SR is not yet the history, but the current reality. Therefore the present lecture may have to finish with the previous chapters. However a few additional comments would deserve to follow for understanding what the motivations in planning the 3rd generation SR was.

The motivation for the 3rd generation SR was obvious. In spite of the strong impact of undulators as the several orders of magnitude brighter new source, 2~4GeV storage rings of the 2nd generation could not meet the demand. Those rings could provide UR in soft X-rays but not in hard X-rays. Furthermore, the most of the 2nd generation storage rings were not designed specifically with low emittance to meet the full exploitation of the potential capability of undulators. It was obvious that the undulator-based SR X-Ray sources have to be 5~10 GeV class Machines with emittance of less than 5nmrad. Difficulties in designing such low emittance storage rings were anticipated at the initial phase of planning, but eventually the problem had been solved after a few years of international collaboration between US, Europe and Japan, and all these 3rd generation sources are now in full operation with even better brightness and stability than initially anticipated.

6. Summary

SR sources were found to be an extremely valuable continuum source of spectroscopy which filled the previously missing wide gap of electromagnetic radiation. It corresponds to the energy region of atomic inner-shells, so far almost unexplored field of physics.

Physicists came first as pioneers to open this uncultivated field of science, then chemists, biologists, and engineers of various disciplines followed.

The first generation of SR research started as parasites on the accelerators made for the other purposes, namely, the high-energy particle physics. They tried very hard under extremely unfavorable circumstances. Nevertheless, the merit of SR was soon fully demonstrated and turned out rewarding. Users' community grew up very rapidly worldwide.

The second generation of SR soon followed by constructing SR sources as users' own facility. This phase of SR studies was extremely fruitful, in developing optimized light sources, and also in creating a number of novel scientific opportunities. An invention of insertion devices such as undulators was also one of the achievements of the 2nd generation. It allowed a tremendous progress in quality of the source, and opened up the 3rd generation SR, namely, the high-brightness SR X-rays.
The progress is still going on toward further promising future, in which accelerator physicists and advanced SR users are now collaborating for a further jump to open up the next generation SR source, i.e. a coherent and dynamic source of extremely high brightness and femto second pulse, the SASE FEL, or ERL which would allow the ultimate structure and dynamical studies for material and life sciences.

Let's see what will come out next!

The end.