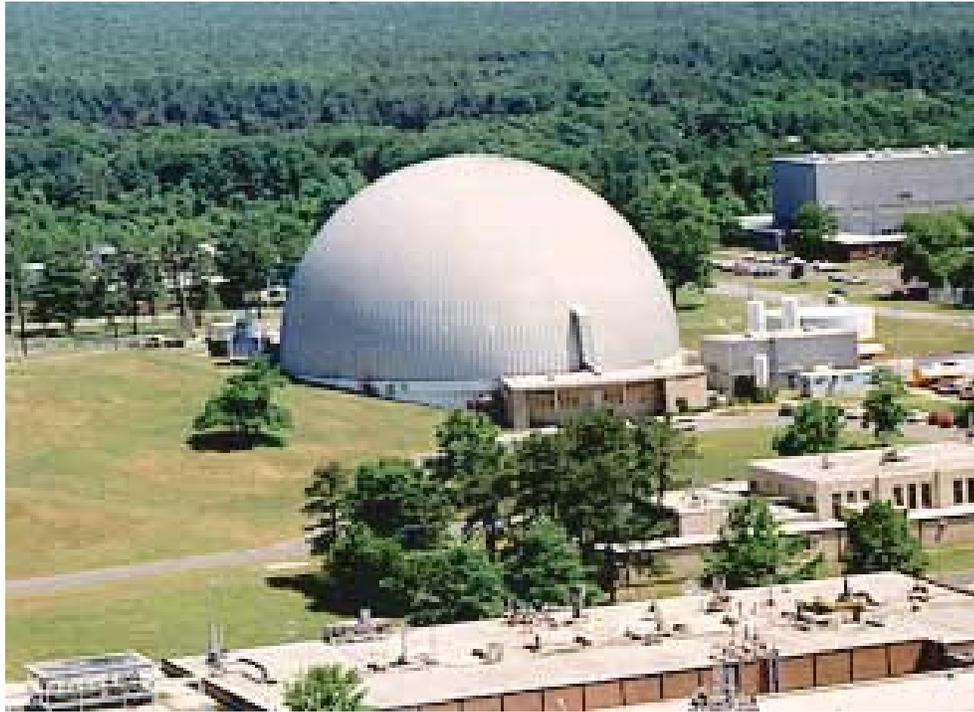


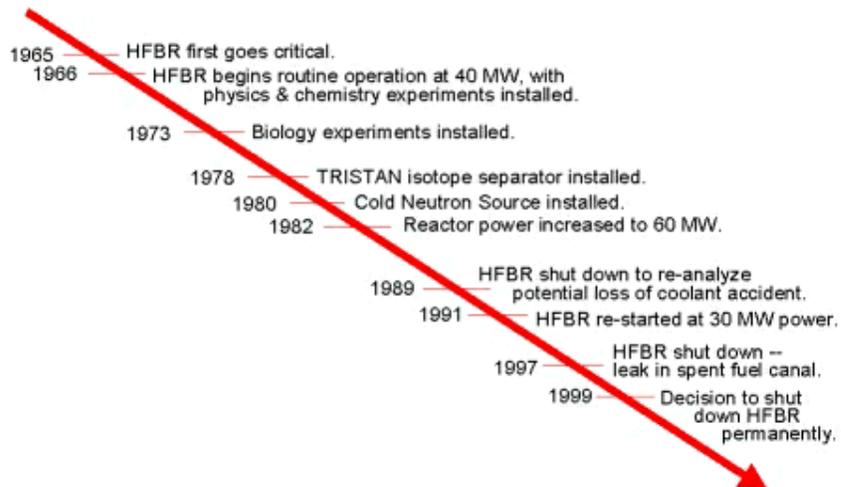
BROOKHAVEN NATIONAL LABORATORY'S HIGH FLUX BEAM REACTOR

Compiled by S. M. Shapiro

I. PICTORIAL HISTORY



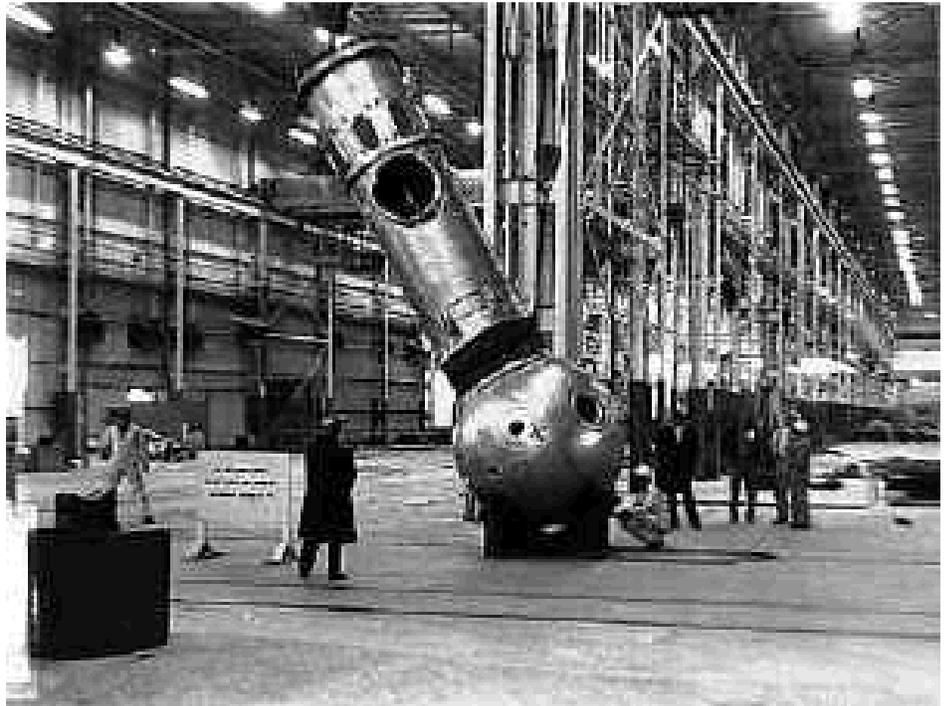
HFBR TIME LINE

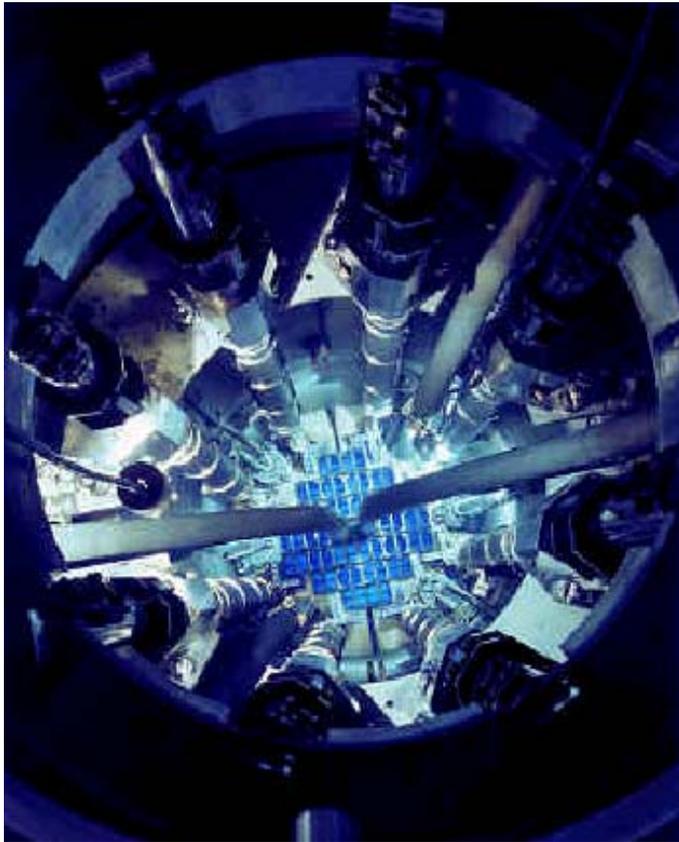




Erecting the dome in 1962. The building was designed as “confinement” to prevent leakage of radioactive materials into the environment. The interior of the building is maintained at a pressure slightly less than that of the atmosphere outside, so that air will tend to leak into, not out of the building. Entrance into the building is through double-door airlocks. The welded hemispherical steel dome is made of 1/4” thick plates and is 176 feet in diameter. It is insulated on the outside and covered with fiberglass insulation and a protective aluminum skin.

The reactor vessel is shaped somewhat like a very large light bulb. It is made of aluminum approximately 2” thick, stands 21 feet high and is 7 feet in diameter at the spherical end. It is located at the center of the reactor building and is surrounded by a lead and steel thermal shield (9” thick) and heavy concrete shielding nearly 8 feet thick.



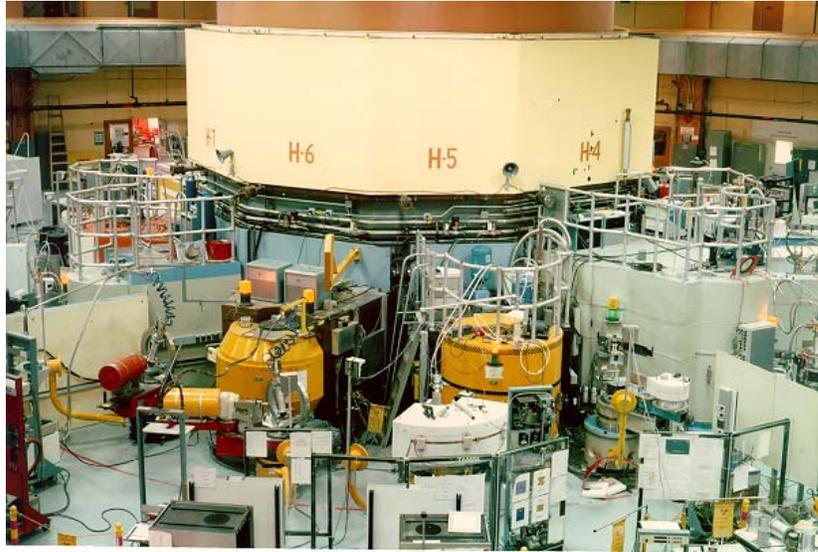


This is a view looking down into the reactor vessel into the core. The 28 fuel elements are seen at the bottom center of the picture. The twin tubes in the center of the core were used for irradiation of samples. Control rod drives are positioned around the periphery of the vessel, and are connected to right angle shaped control blades at the corners of the core. The bluish glow at the center is the Cerenkov radiation. All of the fuel elements were removed and shipped off site in 1997.

Cold source beam plug containing liquid hydrogen being prepared for insertion into the reactor in 1980



HFBR Experimental
Floor -1995



The spent fuel pool emptied of its water in January 1998. The pool contained 68,000 gallons of water up to 30 feet deep for cooling and shielding radioactive fuel and other components.

II. SCIENCE AT THE HFBR

The High Flux Beam Reactor (HFBR) at Brookhaven National Laboratory was built in the early 60's because of the constant needs of scientist to always want "more". In the mid-50's the Brookhaven Graphite reactor was producing a number of important new results when that generation of scientists realized the need for a high flux reactor and started down the political, scientific and engineering path that led to the HFBR. The

effort was joined by a number of engineers and scientist among them, Chernick, Hastings, Kouts, and Hendrie, who came up with the novel design of the HFBR. The two innovative features that have been incorporated in nearly all other research reactors built or planned since are: (i) an undermoderated core arrangement which enables the thermal flux to peak outside the core region where beam tubes can be placed, and (ii) beam tubes that are tangential to the core which decrease the fast neutron background without affecting the thermal beam intensity. In addition, the HFBR was designed with a number of vertical thimbles to accommodate material irradiation experiments with a variety of neutron energy spectra for neutron activation analysis, producing special radioisotopes for medical application and for studies of radiation damage in materials. Construction began in the fall of 1961 and four years later, at a cost of \$12 Million, criticality was achieved on Halloween Night in 1965. Thus began a nearly 40 year career of producing many seminal experiments in condensed matter sciences, nuclear physics, biology, chemistry and medical physics which came to an ignoble end when the Secretary of Energy ordered a permanent closure of the HFBR in November 16,1999. (For an historical account of the early days of the HFBR see Chapter 12 in *Making Physics: A Biography of Brookhaven National Laboratory, 1946-1972*, by Robert P. Crease (The University of Chicago Press, Chicago, 1999))

Nearly 3000 papers were published as a result of research performed at the HFBR. This was accomplished with a relatively small number of dedicated scientist hosting and training a large number of collaborators. There were originally the senior members of the solid state group of the BNL's physics department, Robert Nathans, Gen Shirane, and Chalmers Frazer, who were joined by Dave Cox and Larry Passell at about the same time that the HFBR started doing experiments. John Axe joined in the late 60's, Steve Shapiro followed in the 70's, and John Tranquada in the mid-80's. Julius Hastings and Lester Corliss were stalwarts at the graphite reactor and started doing their work on magnetism under the auspices of the Chemistry Department. Walter Hamilton, whose untimely death shortened a brilliant career, was the resident crystallographer in Chemistry and his protégé, Tom Koetzle, took over the chemical crystallography studies after Hamilton's death. Benno Schoenborn of the Biology department pioneered the use of neutrons for Biology and he recruited Dieter Schnieder to lead the instrument development. Vance Sailor and Bob Chrien originally headed the nuclear physics effort at the HFBR and were superceded by Rick Casten. Joining these more permanent members of the BNL staff were many talented scientists, some of whom started their scientific careers at BNL and went on to become leaders in the international scientific community. They include distinguished professors at leading universities, laboratory directors, government science advisors and prominent researchers throughout the world.

1960's

- **Structure and spin dynamics in ferromagnets and antiferromagnets:** [*Neutron scattering from K_2NiF_4 : a two-dimensional Heisenberg antiferromagnet*, R. J. Birgeneau, H. J. Guggenheim, G. Shirane, *Phys. Rev. Lett.* **22**, 720 (1969)]
- **Triple-Axis neutron techniques:** [*The resolution function in neutron diffractometry I. The resolution function of a neutron diffractometer and its*

application to phonon measurements, M. J. Cooper, R. Nathans, Acta Cryst., 23 (1967)]

The most important advance in inelastic neutron scattering techniques was the invention by B. N. Brockhouse (McMaster U., Canada), in the 1950's, of the triple axis spectrometer. The world's scientific community recognized this by awarding him the Nobel Prize in 1994 along with Cliff Shull of MIT. Perhaps, the second most important advance was the work of M. J. Cooper and R. N. Nathans at Brookhaven who wrote a series of papers in the mid-60's describing the resolution function of the neutron three axis crystal spectrometer. Their elegant formalism reduced the complicated resolution function to a four dimensional matrix (one energy and three momentum) that is used by every group in the world who does inelastic neutron scattering. The complete and accurate understanding of the resolution function makes the neutron triple axis spectrometer the most versatile scattering instrument in use today and allows current scientists to extract very subtle features of collective excitations and structures in materials from their data.

1970's

- **Soft mode description of structural phase transitions:** [*Critical neutron scattering in SrTiO₃ and KMnF₃, S. M. Shapiro, J. D. Axe, G. Shirane, T. Riste, Phys. Rev. B 6, 4332 (1972)*]
- **Phonons in He and rare gas solids.** [*Inelastic neutron scattering from solid Ar-36, Y. Fujii, N. A. Lurie, R. Pynn, G. Shirane, Phys. Rev. B 10, 3647 (1974)*]
- **Critical phenomena in magnetic transitions:** [*Quantitative Analysis of Inelastic Scattering in Two-Crystal and Three-Crystal Neutron Spectrometry; Critical Scattering from RbMnF₃, A. Tucciarone, H. Y. Lau, L. M. Corliss, A. Delapalme, and J. M. Hastings, Phys. Rev. B 4, 3206-3245 (1971)*]
- **Structure and dynamics of molecules adsorbed on surfaces,** [*A neutron scattering study of nitrogen adsorbed on basal-plane-oriented graphite, J. K. Kjems, L. Passell, H. Taub, J. G. Dash, and A. D. Novaco, Phys. Rev. B 13, 1446 (1976)*]

In the 1970's there was intense interest in physisorbed gases on various surfaces, both from a fundamental scientific value and the importance in the catalysis industry. Neutrons generally were not considered to be a good surface probe, because of the very weak neutron-atom interactions and, because of its penetrating power, the difficulty of distinguishing between the atoms on the surface and the substrate. However, the advantages of neutron diffraction over the more traditional low energy electron diffraction studies are significant: i) a wide range of surface thickness can be studied, ii) the neutron is a 'gentle' probe and will not cause significant film evaporation of the weakly bound atoms on the surface as the energetic electron beams do, and iii) inelastic studies can be performed to study the atomic motions of the films. Passell and his co-workers recognized these features and realized that Grafoil, a commonly employed

adsorber for macroscopic experiments, was an ideal substrate for neutron investigations. This was due to the facts that carbon has a small neutron capture cross-section and is a moderate scatter. Therefore the penetration is great and the scattering from the substrate would not totally mask the scattering from the film. Most important, however, was that Grafoil has a very large specific area with a preferred orientation which allows many atoms to be adsorbed onto the surface and thus enable detection by neutron diffraction. The first neutron work was on the structure of adsorbed nitrogen on Grafoil and then progressed to the studies of the dynamics of argon films. Afterwards more complicated molecules were studied such as methane on Grafoil and then studies on different substrates such as small cubes of MgO. These studies coincided with the arrival of synchrotron light sources, which, because of their brightness, were shown to be ideally suited for the structural studies. However, for the dynamical studies, the neutron beam is still the probe of choice.

- **Correlations in one- and two- dimensional magnets:** [*Magnetism in one dimension*, R. J. Birgeneau, G. Shirane, *Physic Today* **31**, no. 12, 32 (1978)]
- **Direct measure of electron-phonon interaction in “old” superconductors:** [*The influence of the superconducting enrgy gap on phonon line widths in Nb₃Sn*, J. D. Axe, G. Shirane, *Phys. Rev. Letters* **30**, 214 (1973)]
- **Magnetic superconductors:** [*Neutron scattering study of magnetic ordering in the re-entrant superconductor ErRh₄B₄*, D. E. Moncton, D. B. McWhan, J. Eckert, G. Shirane, W. Thomlinson, *Phys. Rev. Letters* **39**, 1164 (1977)]
- **Symmetry in nuclei:** [*Evidence for a new symmetry in nuclei: the structure of ¹⁹⁶Pt and the O(6) limit*, J. A. Cizewski, R. F. Casten, G. J. Smith, M. L. Stelts, W. R. Kane, H. G. Börner and W. F. Davidson, *Phys. Rev. Letters* **40**, 167 (1978)]
- **Structure determination of amino acids and sugars:** [*Precision Neutron Diffraction Structure Determination of Protein and Nucleic Acid Components. I*, *J. Am. Chem. Soc.* **94**, 2657 (1972), M. S. Lehmann, T. F. Koetzle, W. C. Hamilton]

Beginning in the early 1970's, the single-crystal diffractometers at the HFBR were exploited to study the structures of amino acids and sugars. This research was originated by Walter Hamilton (amino acids) and George Jeffrey (sugars). The studies carried out at the HFBR produced very precise molecular geometries for these biologically-important small molecules and, perhaps more importantly, made it possible to characterize their hydrogen bonding properties in exhaustive detail. For example, in their study of the amino acid L-alanine, Lehmann, Koetzle and Hamilton (1972) describe the three-dimensional network of N-H-O hydrogen bonds defined by the molecular packing arrangement in the solid state. The hydrogen bonding interactions in these systems are of great general interest, because they play a central role in molecular recognition phenomena in biology.

1980's

- **High Temperature Superconductors:** [*Antiferromagnetism in $\text{La}_2\text{CuO}_{4-y}$* , D. Vaknin, S. K. Sinha, D. E. Moncton, D. C. Johnston, J. M. Newsam, C. R. Safinya, and H. E. King, Jr., *Phys. Rev. Letters* **58**, 2802 (1987)];

[*Neutron diffraction determination of antiferromagnetic structure of Cu ions in $\text{YbBa}_2\text{Cu}_3\text{O}_{6+x}$ with $x=0.0$ and 0.15* , J. M. Tranquada, D. E. Cox, W. Kunnmann, H. Moudden, G. Shirane, M. Swenaga, P. Zolliker, D. Vaknin, S. K. Sinha, M. S. Alvarez, A. J. Jacobson, D. C. Johnston, *Phys. Rev. Letters* **60**, 156 (1988)]

The most exciting scientific event in the 1980's was the discovery of high temperature superconductivity by Alex Muller (a collaborator at BNL) and Georg Bednorz of IBM in the doped La_2CuO_4 , the so-called 214 compound, with a transition temperature near 40K. This set off a scientific frenzy to understand the properties of this material as well as a search for other oxides, which could be superconducting. The latter was achieved quickly with the discovery of superconductivity in $\text{YbBa}_2\text{Cu}_3\text{O}_7$, the 123 compound, with a $T_c \sim 90\text{K}$. After the discovery of superconductivity a major effort was made to understand the structural, electronic and magnetic properties of these materials. Foremost was the magnetic behavior of the undoped and lightly doped parent compounds. The first determination of the magnetic structures of these compounds was accomplished at the HFBR. A Participating Research Team including EXXON studied a polycrystalline sample of $\text{La}_2\text{CuO}_{4-y}$ and showed that it exhibited an antiferromagnetic order below $T_N = 220\text{K}$. From their data they were able to determine that the magnetic structure consisted of ferromagnetic sheets of copper spins alternating along the [100] orthorhombic axis with the spins aligned along the [001] orthorhombic axis. Shortly thereafter a group led by Tranquada determined the structure of the 123 compound to have a similar in plane structure but coupled antiferromagnetically along the c-axis. The Neel temperature was greater than 500K, twice as high as the 214 compound. These measurements were extremely important in aiding our understanding of the superconducting compounds in that they anchored the phase diagram and provided the crucial information that magnetic behavior has to be considered in trying to understand the origin of high temperature superconductivity, a quest that is still ongoing.

- **Impurity effects on phase transitions:** [*The effects of random fields on the critical and bicritical behavior of $\text{Mn}_x\text{Zn}_{1-x}\text{F}_2$* , R. A. Cowley, G. Shirane, H. Yoshizawa, Y. J. Uemura, R. J. Birgeneau, *Zeitschrift für Physik B-Condensed Matter* **75**, 303 (1989)]
- **Spin glasses:** [*Neutron scattering studies of the anomalous magnetic alloy $\text{Fe}_{0.7}\text{Al}_{0.3}$* , K. Motoya, S. M. Shapiro, Y. Muraoka, *Phys Rev. B* **28**, 6183, (1983)]

- **Magnetic correlations in heavy fermions:** [*Wave vector and magnetic field-dependent spin fluctuations in the heavy Fermion system CeCu₆, G. Aeppli, H. Yoshizawa, Y. Endoh, E. Bucher, J. Hufnagl, Y. Onuki, T. Komatusubara, Phys. Rev. Letters 57, 122 (1986)*]
- **Incommensurate modulations in metals and insulators:** [*Lattice dynamics of an incommensurate system: K₂SeO₄, J. D. Axe, M. Iizumi, G. Shirane, Phys. Rev. B 22, 3408 (1980)*].
- **Transition metal-hydrogen interactions:** [*An interstitial hydrogen atom in a hexanuclear metal cluster: x-ray and neutron diffraction analysis of [(Ph₃P)₂N]⁺[HCo₆(CO)₁₅], D. W. Hart, R. G. Teller, C-Y. Wei, R. Bau, G. Longoni, S. Campanella, P. Chini, T. F. Koetzle, J. Am. Chem. Soc, 23, 4012 (1984)]*]
- **The interacting boson model in nuclear physics:** [*Detailed test of the interacting boson approximation in a well-deformed nucleus: the positive parity states of ¹⁶⁸Er D.D. Warner, R.F. Casten and W.F. Davidson. Phys. Rev. Letters 45, 1761 (1980)*].

The Interacting Boson Model or IBM envisages the structure of nuclei, not in terms of the properties of single nucleons in orbit about the nuclear center of mass, but in terms of pairs of valence nucleons, bosons, in angular momentum zero and two (s and d) states, and their bosonic interactions. The model is analytic for its three dynamical symmetries. Most nuclei, however, are intermediate in structure and do not resemble any of the three limiting ideals. Yet the model Hamiltonian can describe such cases as a competition between terms representing two symmetries (similar to an Ising model). In the early 1980s, it was thought that the IBM would not be useful, or even correct, for deformed (ellipsoidal) nuclei. However, a series of experiments on ¹⁶⁸Er developed the most extreme and complete level scheme ever constructed for a deformed nucleus and showed that the IBM gave a simple and very accurate description with surprisingly few parameters. A key to this work was the development of the (n, γ)-based technique of Average Resonance Capture (ARC) that exploited 2 keV and 24 keV beams from the HFBR. This technique can give an *a priori* guarantee for finding all states in certain spin and energy ranges and is therefore ideal for testing comprehensive models of low energy nuclear collective modes. The ¹⁶⁸Er experiment ushered in a golden era for nuclear structure studies, many at the HFBR, designed to test and explore the evolution of nuclear structure in the IBM. It led directly to the development of a revised approach for the model, called the Consistent Q Formalism (CQF), developed by the same experimentalists at the HFBR who led the ¹⁶⁸Er study. The CQF is now the standard approach to the IBM. This work also indirectly led to the discovery (in the 1990's) of phase transitional behavior (both first and second order) in finite nuclei and to the very recent development of the concept of critical point symmetries [E(5) and X(5)] in nuclei.

- **Structure determination of small sub-unit of ribosomes:** [*A complete mapping of the proteins in the small ribosomal subunit of Escherichia coli, M. S. Capel, D. M. Engleman, B. R. Freeborn, M.*

Kjeldgaard, J. A. Langer, V. Ramakrishnan, D. G. Schindler, D. K. Schneider, B. P. Schoenborn, I. Y. Sillers, et al., Science, 238, 1403 (1987)]

The ribosome is a complex particle that makes the thousands of proteins that are required for the structure and function of each living cell. It is central to every cell because it provides the workshop and tools to synthesize all of the proteins. In order to understand the function of the ribosome it is first necessary to know the structure of the whole and its subunits. This has been a 30-plus year effort which led to the high resolution studies of the electron densities of 30S and 50S ribosome subunits reported in *Science* and *Nature* in 1999. One of the early studies of the 30S unit was accomplished at the HFBR by the biology department of BNL. Since neutrons interact with the nucleus, and not the electrons as do x-rays, the relative positions of the centers of mass of the 21 proteins in the 30S ribosomal subunit was determined using the neutron scattering data. This was a crucial step in eventually solving the final structure and thereby leading to a better understanding of the function of the ribosome and how it builds the proteins.

1990's

- **Exotic behavior on one-dimensional magnets:** [*Magnetic gap excitations in a one-dimensional mixed spin antiferromagnet Nd_2BaNiO_5 , A. Zheludev, J. M. Tranquada, T. Vogt, D. J. Buttrey, Phys. Rev B 54, 7210 (1996)*]
- **Shape memory materials:** [*Neutron scattering and electron microscopy studies of the premartensitic phenomena in Ni_xAl_{1-x} alloys, S. M. Shapiro, B. X. Yang, Y. Noda, L. E. Tanner, D. Schryvers, Phys. Rev. B 44, 9301 (1991)*]
- **Anomalous correlation lengths in phase transitions:** [*Origin of the second length scale above the magnetic spiral phase of Tb, P. M. Gehring, K. Hirota, C. F. Majkrzak, G. Shirane, Phys. Rev. Letters 71, 1087 (1993)*]
- **Structure of ceramics with negative thermal expansion:** [*Negative thermal expansion from 0.3 to 1050K in ZrW_2O_8 , T. A. Mary, J. S. O. Evans, T. Vogt, A. W. Sleight, Science 272, 90 (1996)*]

It is well known that materials expand on heating and contract on cooling. However, a discovery was made that the oxide compound ZrW_2O_8 behaves in just the opposite way; it expands on cooling. No other material is known to exhibit such unusual behavior over such a broad temperature range. The neutron diffraction study performed at the high-resolution powder diffractometer installed at the HFBR showed that the system remained cubic over the entire temperature range and thus the negative thermal expansion behavior is isotropic. The structure, determined by a combined x-ray and neutron diffraction experiment, is composed of rigid ZrO_6 octahedra and WO_4 tetrahedra in a relatively open framework with a high degree of flexibility. Modeling shows that any change in pressure can be accommodated by a coupled rotation of its constituent polyhedra that requires only changes in the relatively flexible Zr-O-W bond. These studies suggest that the negative thermal expansion of ZrW_2O_8 involves thermal librations of these rigid polyhedra. These materials are finding applications as components in composites in order to reduce the overall thermal expansion to near zero. Such

applications are very important in the electronics industry, as substrates for optical mirrors, components of high-precision thermometers, and catalyst supports.

- **Stripes in High T_c :** [*Stripe correlations of spins and holes in copper oxide superconductors, J. M. Tranquada, B. J. Sternlieb, J. D. Axe, Y. Nakamura, S. Uchida, Nature 375, 561 (1995)*]

The work on high temperature superconductors and related compounds continued to dominate the research at the HFBR. The most exciting result was the discovery of stripes in high T_c compounds by John Tranquada and his co-workers. This effort demonstrated the strong interplay between the theoretical and experimental groups at Brookhaven. Tranquada had previously observed an ordered striped phase in non-superconducting nickel oxide compounds. These stripes correspond to a modulation of the spin and charge in which antiferromagnetic ‘stripes’ of copper spins are separated by periodically spaced domain walls to which the holes segregate. Vic Emery had been working with Steve Kivelson (UCLA) in trying to understand the origins of high temperature superconductivity and since his office was merely 100 ft from Tranquada’s, the daily interplay between the two resulted in the discovery of the stripe correlations in copper oxide superconductors. The exact relationship between the stripes and the high temperature superconductivity has not been resolved and is still under intense investigation in the current decade by a large group of experimentalists and theorists.