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NEUTRON SCATTERING RESEARCH RESULTS FROM THE LOS ALAMOS NEUTRON SCATTERING CENTER

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ABSTRACT

The Los Alamos Neutron Scattering Center (LANSCE) is a new high intensity pulsed spallation neutron source for research in the structure and dynamics of condensed matter. With first operation in 1985, LANSCE is expected to produce the world's highest peak thermal flux of 1.7 X 10^{16} n/cm^2-sec by 1987. A description of the LANSCE facility and source characteristics is presented. Some experimental highlights of research at the low intensity (peak thermal flux of 6 X 10^{13} n/cm^2-sec) WNR predecessor to LANSCE are described which illustrate the scientific potential and instrumentation of LANSCE. Future plans for the LANSCE facility and research program are discussed.

I. INTRODUCTION:

The Los Alamos Neutron Scattering Center (LANSCE), which is just beginning operation at the Los Alamos National Laboratory, is a pulsed spallation neutron source for research in condensed matter science (solid state physics, chemistry, materials science, and biology).[1] The peak thermal flux of LANSCE is expected to reach 1.7 X 10^{16} n/cm^2-sec by 1987. This is a factor of forty increase beyond present pulsed source capabilities in the United States. It will be competitive with the new Spallation Neutron Source beginning operation at the Rutherford Appleton Laboratory in the United Kingdom. Based at the world's highest current proton accelerator, the Los Alamos Meson Physics Facility (LAMPF), LANSCE has the potential for significant further upgrades at a small fraction of the costs for constructing a new facility of comparable capability. This can keep the U.S. at the forefront of pulsed neutron scattering research through the remainder of this century. The accelerator, instrumentation, and research

experience developed at LANSCE should also lead to a proposal for a next-generation[2] neutron source (peak thermal flux of 10^{17}-10^{18} n/cm^2-sec) for condensed matter research in the 21st century.

This paper describes the characteristics of the LANSCE facility. Neutron scattering research highlights are presented from the past three years of operation of the low intensity (peak thermal flux of 6 X 10^{13} n/cm^2-sec) Weapons Neutron Research (WNR) predecessor to LANSCE, with a discussion of implications for LANSCE research. Long range plans are described for LANSCE facility development, instrumentation, and research programs.

II. LANSCE FACILITY DESCRIPTION:

Pulsed neutron scattering research began at Los Alamos in 1977 with the first operation of the Weapons Neutron Research (WNR) facility.[3] LAMPF produces protons at 120 Hz in "macropulses" of approximately 750 μsecs pulse width. The requirement for short pulse widths for time-of-flight neutron scattering allowed only a small fraction of each macropulse to be used. The characteristics of the WNR facility were 3.5 μA time average proton current, 120 Hz repetition rate, 5 μsecs proton pulse width, and 6 X 10^{13} n/cm^2-sec peak thermal neutron flux. Compared with an optimal pulsed neutron source, the proton current was too low, the repetition rate was too high, and the proton pulse width was too long. In addition, beam time had to be split between neutron scattering and the other competing uses of the WNR facility for nuclear physics, neutrinos, and defense programs. Nevertheless, the operation of the WNR facility from 1978 to 1984 provided valuable experience in pulsed neutron scattering research, to be described later in this report.

The WNR also led to the development of the world class LANSCE facility which is beginning operation in 1985. The Los Alamos Neutron Scattering Center, LANSCE, is the name given to the materials science research program at the facility created by the combination of the WNR facility with a new $22M Proton Storage Ring (PSR),[4] which acts as a pulse compressor of LAMPF macropulses. By compressing one in ten of every macropulse, a much improved pulsed neutron source is created with 100 μA time average proton current, 12 Hz repetition rate, 0.27 μsec proton pulse width and 1.7 x 10^{16} n/cm^2-sec peak thermal flux. The PSR first accepted proton beam on April 26, 1985, and the testing of this facility is proceeding on schedule with the design characteristics expected to be reached in October, 1986. The upgrade of the facility includes a proton beam multiplexing scheme and the construction of new target areas to allow simultaneous use by the four major research programs in neutron scattering, nuclear physics, neutrinos,

and defense. The neutron scattering target/moderator assembly is also being rebuilt to improve shielding to handle the much higher proton current, increase the number of possible flight paths to 17, and install a 20 K H_2 moderator for long wavelength neutron experiments. The new LANSCE facility is approximately a two orders of magnitude improvement over the WNR facility which preceeded it.

III. NEUTRON SCATTERING RESEARCH RESULTS FROM THE WNR:

This section reviews some highlights of condensed matter research and instrument development during the operation of the low intensity WNR facility. This will illustrate the characteristics of time of flight instrumentation and some of the possible research areas with the new LANSCE facility. Prototype neutron scattering instruments first became operational at the WNR in 1981, so the results discussed below were obtained in the three-year period from 1981 to 1984. Unfortunately, for many of the experiments in the 1984 run cycle, the data analysis is not yet complete or the papers have not yet been accepted for publication. Therefore, many interesting results from the WNR could not be included in this review.

Of course, the increase of 300 in peak neutron flux made possible by the PSR should open up qualitatively new research areas, such as the study of dispersive elementary excitations (e.g., phonons and magnons) at high energy transfers. Similarly, the factor of ten lower repetition rate and the installation of a cold H_2 moderator should make possible experiments with long wavelength neutrons. So the following represents only a glimpse into the future of LANSCE.

Single Crystal Diffraction

Fig. 1 shows a schematic drawing of the Single Crystal Diffractometer (SCD) at the WNR facility. A white beam of neutrons travels 7.5 m from the moderator to the sample mounted on a displex refrigerator, and then Bragg scatters into a ^{3}He multiwire position sensitive neutron detector. Data are recorded in a three dimensional histogramming memory corresponding to X and Y on the detector and the total time-of-flight. This allows the scattering angles and wavelengths to be reconstructed. The instrument may be thought of as a wavelength resolved Laue camera. The sample is mounted on a two-axis (ω and ϕ drive) goniometer, which is sufficient to cover the majority of the reciprocal space sphere.

The SCD was used to determine the hydrogen positions in the first example[5] of molecular hydrogen reversibly bound to the central metal of an organometallic transition metal complex. Fig. 2 shows the core atoms of $W(CO)_3(P\text{-}Pr^i_3)_2(\eta^2\text{-}H_2)$, where the circled P represents a phosphine ligand.

The hydrogen-hydrogen distance (0.82 Å) is significantly larger than that obtained in free hydrogen (0.74 Å). This type of binding had been proposed to explain some types of catalytic reactions and these structural results clearly demonstrate the existence of such a bonding mode.

A new technique[6] for performing room temperature high-pressure single crystal neutron diffraction (up to 20 kbar) has been developed for the SCD. This uses a cylindrically symmetric pressure cell mounted on the incident collimation with rotation axis coincident with the neutron beam and a specific crystal orientation chosen with beam passing midway between major lattice vectors. For a crystal of orthorhombic or higher symmetry, it is possible to refine a three-dimensional structure with only one pressure loading. Presence of an area detector allows easy identification of superlattice reflections which occur as a result of phase transitions, and the multiwavelength neutron source provides considerably more information for a single setting of the crystal than conventional single wavelength sources. This technique was used[7] to study the structural phase transition above 5.2 kbar in ReO_3, where the high pressure structure (Fig. 3) has ten times the compressibility of the normal phase (termed a "compressibility collapse" transition). At the transition, the Re-O_6 octahedron begin to rotate causing an apparent hinging of the linear Re-O-Re chains rather than a shortening of the Re-O distances, which doubles the unit cell giving rise to superlattice reflections easily visible with an area detector.

Powder and Liquids Diffraction

A large number of powder diffraction studies have been conducted at the WNR using the Rietveld powder profile refinement method. One should note that good resolution at high d-spacing is a major advantage of time-of-flight diffraction. Most of the research has involved actinide compounds which are important to the Laboratory's applied programs or to basic research on f-electron phenomena.

Fig. 4 shows the room temperature structure of $BaPuO_3$ determined[8] using the Neutron Powder Diffractometer on a 10 m flight path at the WNR. $BaPuO_3$ is an important material in the technology of nuclear waste recovery. This experiment required the use of special sample environments to handle extremely radioactive materials. The structure is found to be a distorted perovskite structure ($GdFeO_3$ type), with nearly regular PuO_6 octahedra but 1.46 degree hinging of the O-Pu-O angles, in excellent agreement with expectations based on crystal chemical models.

PGa_3 has the cubic Cu_3Au - type structure and exhibits a very small glitch in its magnetic susceptibility curve at ~ 65 K. Previous studies of this compound suggested that it was paramagnetic down to 4.2 K. Reactor neutron powder diffraction studies later suggested an antiferromagnetic

structure, though harmonic contamination of the incident beam obscured the superlattice reflections. A reinvestigation of this material by neutron time-of-flight techniques[9] eliminated the contamination problem and allowed unambiguous confirmation of the existence of a simple antiferromagnetic structure with a Neel temperature of 70 K. Also studied was UGa_2, a low-temperature ferromagnet with the hexagonal AlB_2 structure. Values of the moments obtained as a function of temperature are shown in Fig. 5. The value of the moment in UGa_2 is 3.0 μ_B indicating some delocalization of the moment. The value for UGa_3 is 0.95 μ_B indicating considerable delocalization. These observations are consistent with the observed U-U distances of 4.02 Å and 3.01 Å for UGa_2 and UGa_3 respectively.

Pulsed neutron sources are particularly advantageous for studies of the structures of liquids and amorphous materials because of the very high epithermal neutron flux. Using high incident energies at low scattering angles to achieve a desired momentum transfer Q greatly reduces the inelasticity, or Placzek, corrections required to convert a cross section to a structure factor. In practice, such corrections introduce unwanted model dependent assumptions into the data analysis. The high energies also allow one to reach very large Q values (50 $Å^{-1}$ is easily attained) which is especially important for molecular liquids. This allows fourier transform of the data to obtain radial distribution functions without introducing model dependent assumptions about the large Q scattering. Fig. 6 shows the hydrogen-hydrogen partial structure factor for liquid water measured[10] in a hydrogen/deuterium isotope substitution experiment at the WNR, as compared with two molecular dynamics simulations based on differing intermolecular water potentials. While agreement with the simulations is very good, the data reveal a water structure with stronger short range order (closer to the structure of ice) than either simulation.

Inelastic Scattering

The Be-BeO Filter Difference Spectrometer[11] (FDS) at the WNR facility is used for vibrational spectroscopy on non-dispersive excitations up to energies of several hundred meV. The FDS uses the low pass Bragg cutoffs of Be at 5.2 meV and BeO at 3.6 meV to fix a final neutron energy bandpass, with the incident energy determined by the total time-of-flight.

Fig. 7 shows measurements[12] using the FDS of the vibrational spectra of hydrogen chemisorbed on Raney nickel (top) and coprecipitated nickel (bottom), both materials that are closely related to commercial catalysts. The spectra give information about the strength of the bonding of hydrogen to the substrate (namely, the energy of the transition) as well as the geometry of the adsorption site. By a straightforward calculation the

peaks at 120 and 140 meV can be associated with the vibrations parallel and perpendicular (respectively) to a hydrogen atom adsorbed over the center of a triangular, or (111)-like, site. The pair of peaks at 80 and 200 meV would then correspond to hydrogen over the center of a fourfold, or (100)-like, site. The relative number of (100) sites in the coprecipitated nickel appears to be larger than in Raney nickel. The next step toward understanding catalysis of these systems would be to coadsorb oxygen and hydrogen to see which sites are more reactive.

The normal modes of H in metals cannot be studied by any other technique but neutron scattering, and they are of high enough energy to often require the unique spectral characteristics of a pulsed neutron source. In a bcc hydride such as TaH_x and NbH_x, the hydrogen vibrations can be described in terms of a localized, slightly anharmonic potential in a tetragonal ($\bar{4}2m$) site. Fig. 8 shows the vibrational spectra of $TaH_{0.46}$ measured[13] with the FDS at very high energy transfers up to 400 meV. These measurements of the positions of the higher harmonics of the singlet (out-of-plane) and doublet (in-plane) fundamentals have been used to determine the independent terms in the third and fourth order anharmonic perturbation expansion of the hydrogen potential, to compare with theoretical calculations and with other measurements such as the activation energy for hydrogen diffusion.

The Constant-$\underline{Q}$ Spectrometer[14] (CQS) shown in Fig. 9 is a pulsed neutron source instrument for studies of $\vec{Q}$-dependent collective excitations such as phonons and magnons in single crystal samples, which are studied with triple axis spectrometers (TAS) at reactor neutron sources. The CQS uses Bragg reflection of scattered neutrons from an array of analyzer crystals to fix the perpendicular momentum transfer and to determine the final neutron energy. Then, the total time-of-flight to the detectors fixes the remaining parameters ($\vec{Q}$, ω). Compared with a TAS, the CQS will allow a broad region of $\vec{Q}$,ω space to be measured simultaneously, will reach higher energy transfers, and will have complementary focusing conditions. This was a development instrument at the WNR, poised to exploit the increased flux provided by the PSR. WNR research demonstrated the viability of the CQS method. Background levels are comparable to current reactor practice, both dispersive and non-dispersive phonon branches have been measured in aluminum in good agreement with the literature, and the exploitation of reciprocal space focusing has been demonstrated.

The prospect of using high energy neutrons has also stimulated some theoretical work. In the high Q limit, the scattering is thought to approach the impulse approximation limit in which the scattering function $S(\vec{Q},\omega)$ becomes related to the ground state momentum distribution $n(\vec{p})$. Los Alamos work has shown[15] that the relationship between these two functions

is mathematically equivalent to a Radon transform, in which the matrix relating the polynomial expansions of both functions is diagonal. Further, in the case of ions in solids, these polynomial expansions will be rapidly converging for realistic potentials, even in the case of strong anharmonicity as in double well hydrogen bonds. Thus, a polynomial fit to $S(\vec{Q},\omega)$ in the impulse approximation limit will yield the terms in the polynomial expansion of $n(\vec{p})$. Planning for experiments on hydrogen bonded systems to test these ideas are in progress at LANSCE.

IV. THE FUTURE OF LANSCE:

The approximately two orders of magnitude improvement in source characteristics to be provided by the Proton Storage Ring should open up many areas for neutron scattering research at LANSCE which were impossible or marginal with WNR. The kinds of neutron scattering experiments which would benefit from more intensity are well documented,[16] including higher resolution, smaller cross section, differential scattering, and polarization analysis experiments. The new field of neutron scattering experiments on high energy excitations[16] in condensed matter made possible by pulsed neutron sources requires the improvement in peak flux as well as the better time-of-flight resolution provided by the decreased proton pulse width of the PSR. Because of the much lower repetition rate which eliminates "frame overlap" and the installation of a 20 K liquid H_2 moderator, cold neutron science (such as small angle neutron scattering studies of structural biology, polymers, and metallurgy) will become possible for the first time at Los Alamos.

To take optimal advantage of LANSCE intensities to perform high resolution experiments and to provide the conveniences and services required to operate LANSCE as a national user facility to attract the condensed matter science community to pulsed neutron scattering, an expansion beyond the existing WNR experimental hall is essential.[17] The present hall has a maximum flight path length of 13 m, so that higher resolution experiments requiring longer flight paths must be carried out in sheds built in a time-of-flight yard, and there are no ancillary support facilities for users. An $18M proposal[18,19] to the Department of Energy would provide for a new experimental hall with up to 50 m flight paths, as well as sample preparation laboratories, computers, office space and additional instrumentation. Provided initial funding in FY86, this expansion should by completed by the end of 1988.

Current instrument construction projects at LANSCE include two instruments scheduled for initial operation in 1986: a Low Q Diffractometer for small angle neutron scattering experiments and a Chopper Spectrometer for studies of high energy excitations. Three additional

instruments are in the early conceptual design stage to be constructed as part of the new experimental halls project. a High Resolution, High Throughput Powder Diffractometer (HRPD); a 10 μeV resolution Quasielastic Neutron Spectrometer (QENS) for studies of chemical spectroscopy, diffusion, tunneling, etc.; and a Polarization Analysis Instrument[20] (PAN) using a supermirror guide, neutron chopper, and the neutron spin echo technique to achieve very high resolution for the study of high energy excitations.

These projects require advances in neutron scattering technology. Los Alamos has active development efforts in very high rate (10 MHz) data acquisition systems (FASTBUS μVAX), target/moderator optimization including fission boosted targets and cold moderators, high speed position sensitive scintillator detectors, new plastic neutron scintillator materials, 600 Hz magnetic bearing neutron choppers, pressed Ge and Cu crystal monochromators, etc. Other technologies needed for instrument development (such as neutron guides and supermirrors) will be acquired through collaborations or purchase.

Provided Congressional approval for FY86 funding of the Neutron Scattering Experimental Halls project and adequate operations and research support from the Department of Energy, one can speculate on a possible timetable for LANSCE and U.S. pulsed neutron scattering development:

1985 - PSR reaches 20 μA at 24 Hz, W target installed (peak thermal flux of 1 X 10^{15} n/cm^2-sec)

1986 - Begin architectural design for new halls, PSR reaches 100 μA at 12 Hz (1 X 10^{16} n/cm^2-sec), LQD and CS begin operation

1987 - Install depleted uranium target (1.7 x 10^{16} n/cm^2-sec), construction of experimental halls

1988 - Occupy new halls, initiate formal National User Program, HRPD and QENS begin operation, bring PSR operation up to 200 μA at 24 Hz

1989 - Occupy remainder of construction project, PAN begins operation, install fission boosted target (peak thermal flux of 5 X 10^{16} n/cm^2-sec)

1990-2000 - Routine operation of LANSCE with continuing instrument development, begin to develop proposal for an advanced pulsed neutron source based on LANSCE experience (peak thermal flux 10^{17} - 10^{18} n/cm^2-sec)

1996 - Begin construction of an advanced U. S. pulsed neutron source for the 21st century.

One can see that the beginning of operation of the Los Alamos Neutron Scattering Center is an exciting and challenging opportunity to advance the state of the art in condensed matter research.

V. ACKNOWLEDGEMENTS

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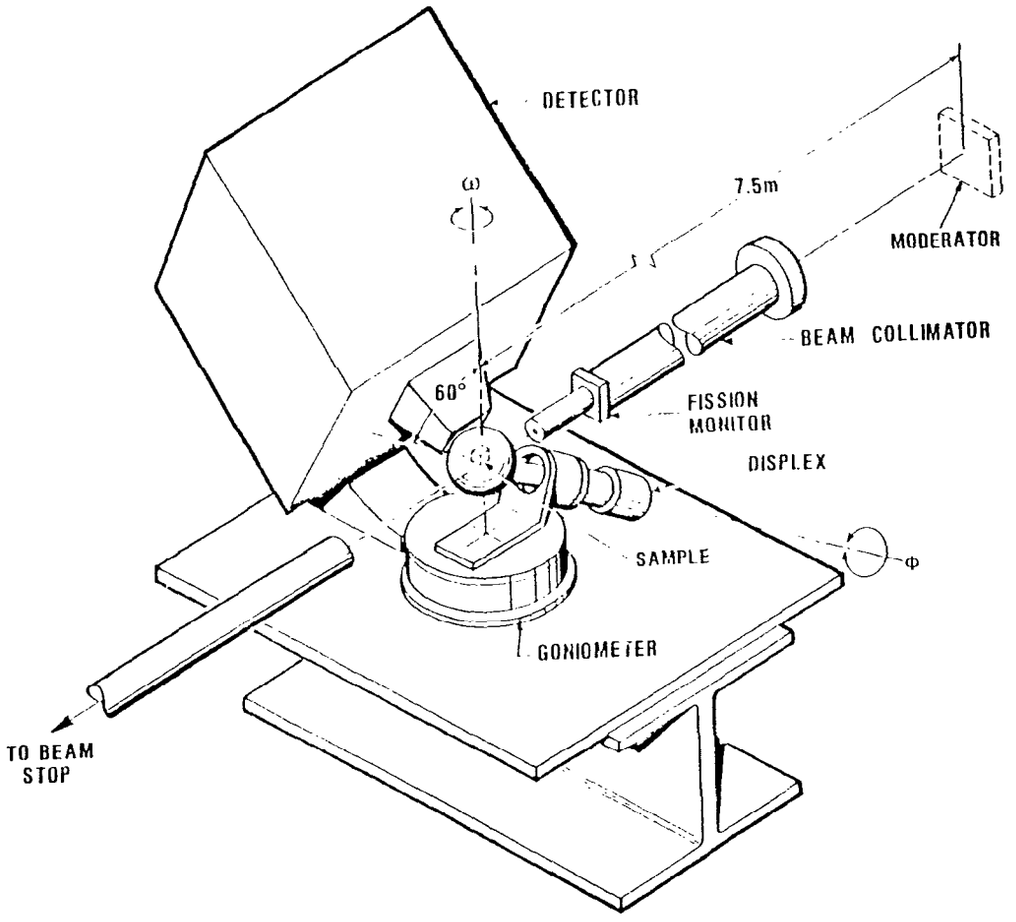

Fig. 1 Schematic drawing of the Single Crystal Diffractometer at LANSCE.

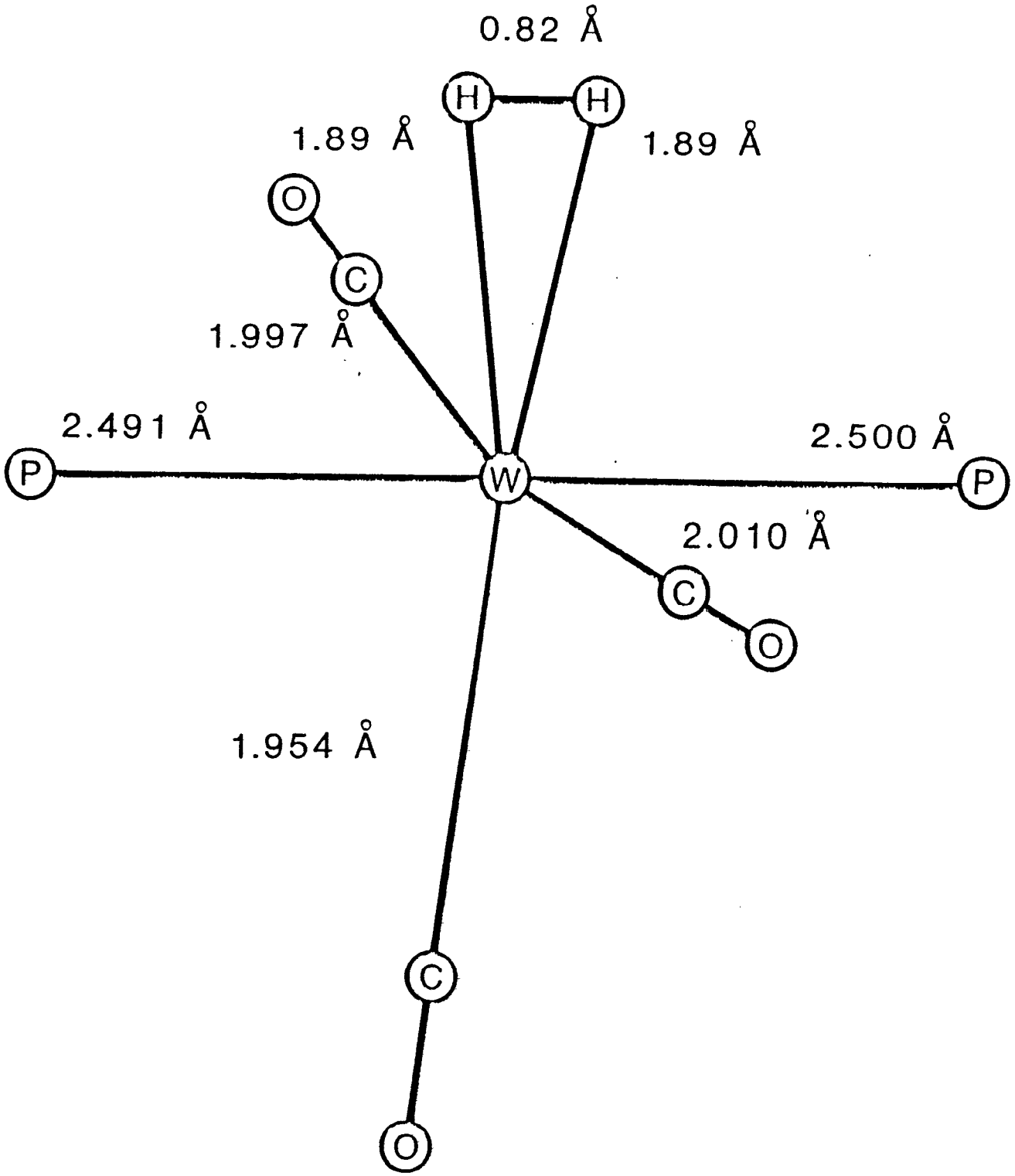

Fig. 2 Core atoms of $W(CO)_3(P\text{-}Pr^i_3)_2(\eta^2\text{-}H_2)$. This first example of an organometallic complex which reversibly binds molecular hydrogen was initially observed at the WNR facility. The circled P's represent phosphine ligands.

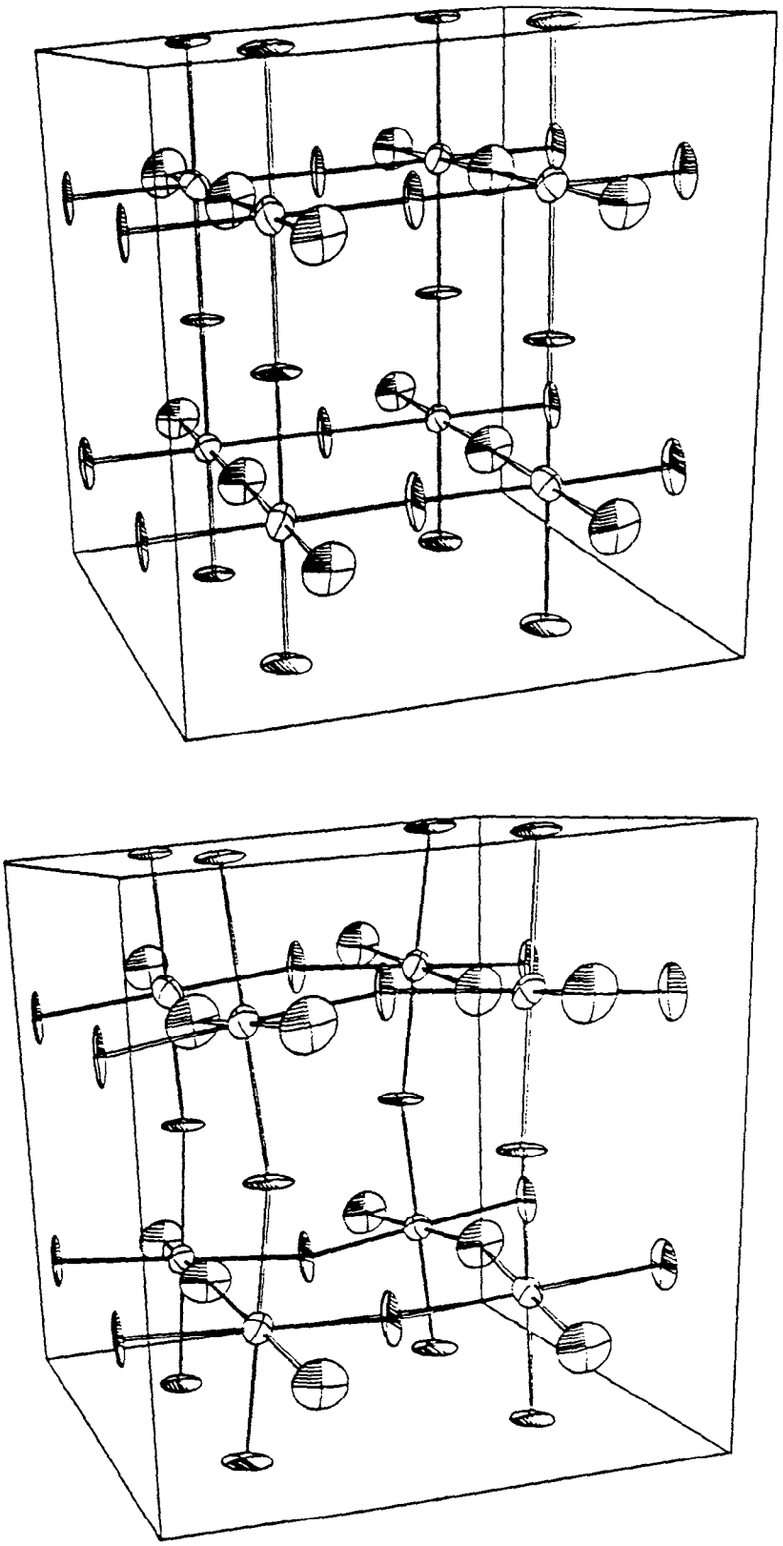

Fig. 3 The structure of ReO_3 at zero pressure (top - eight unit cells) and at 15 kbar (bottom - single unit cell) as measured on the Single Crystal Diffractometer at the WNR.

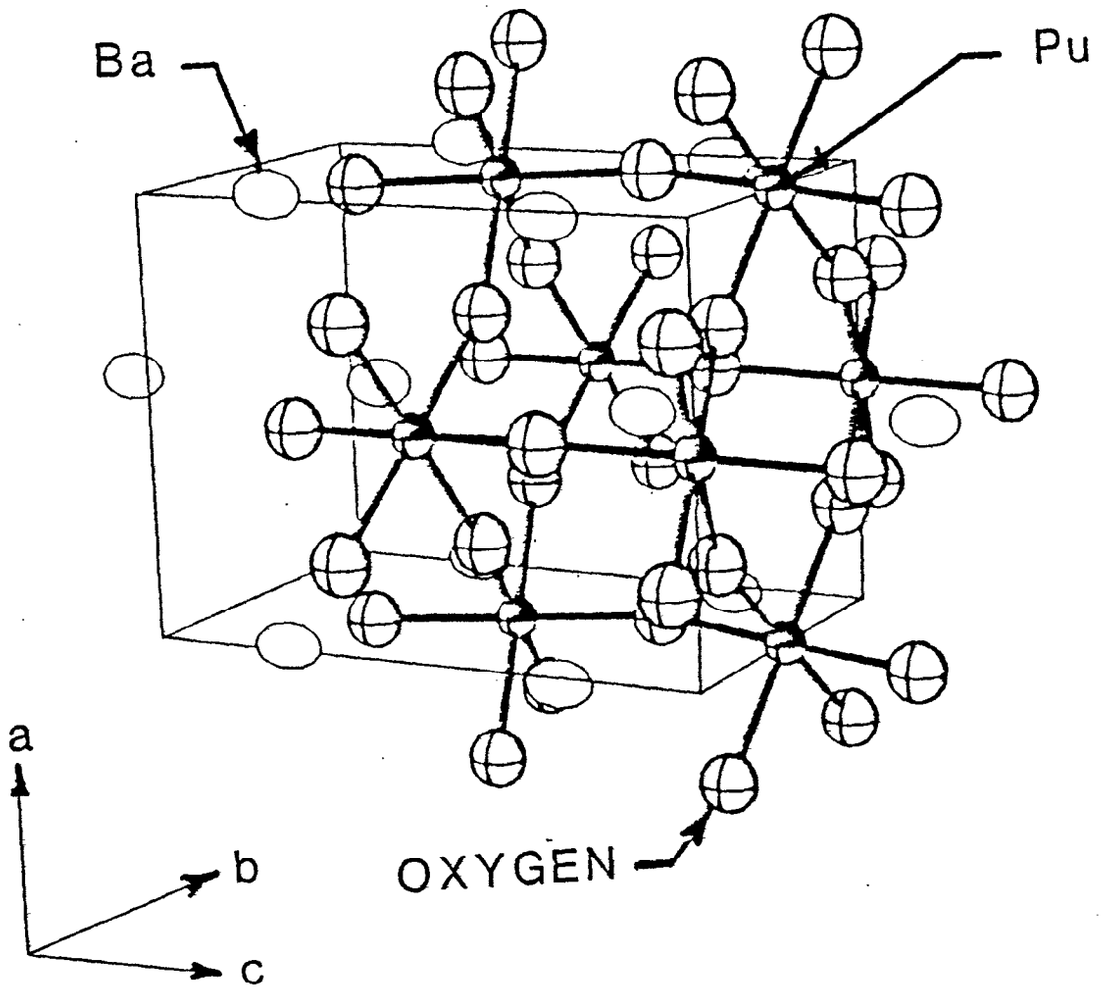

Fig. 4 The structure of $BaPuO_3$ as measured by time of flight powder diffraction at the WNR.

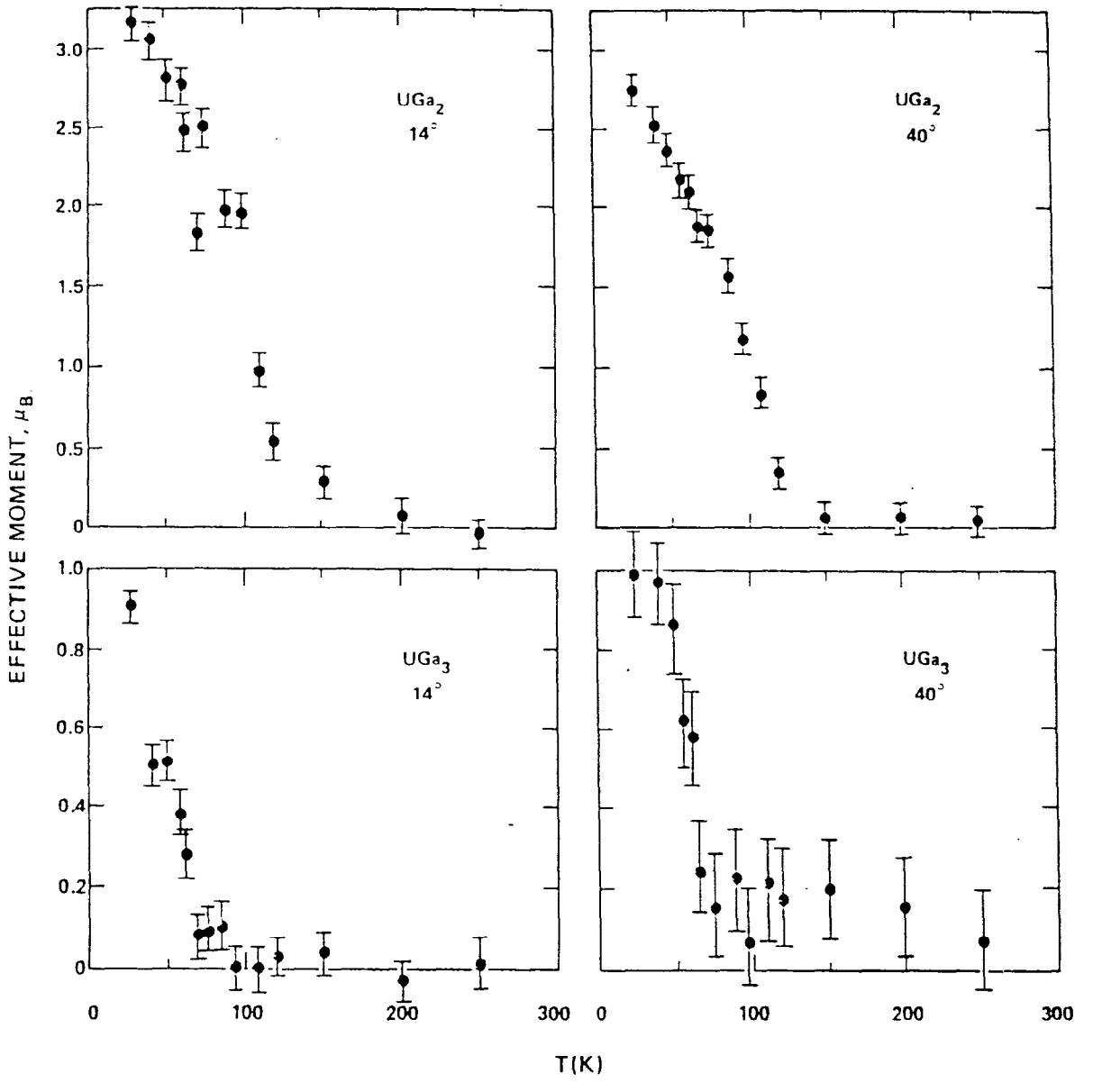

Fig. 5 Uranium magnetic moments as a function of temperature measured by time-of-flight neutron powder diffraction in the ferromagnet UGa_2 amd the antiferromagnet UGa_3.

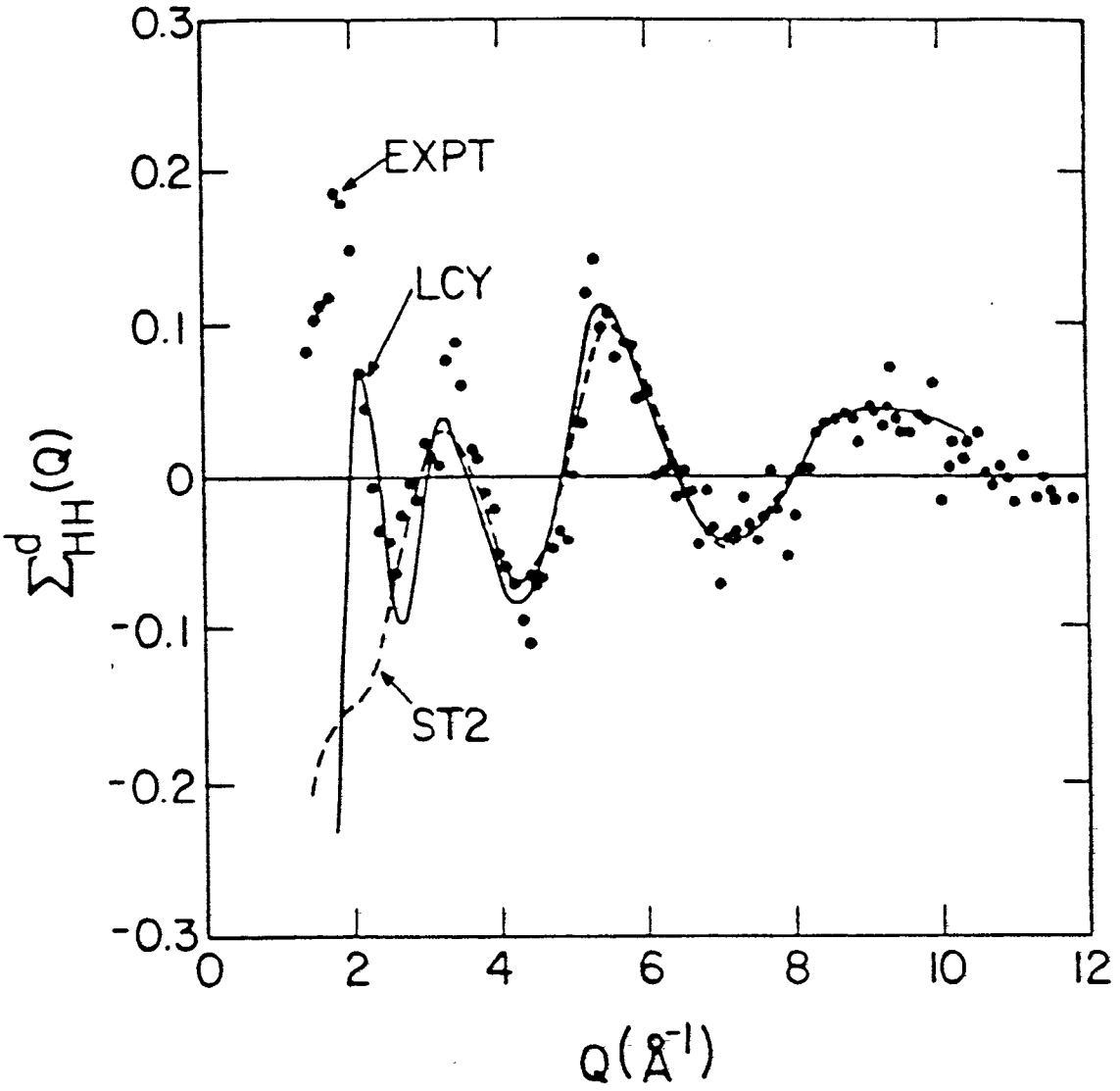

Fig. 6 Hydrogen-hydrogen partial structure factor for liquid water (dots) as measured in an hydrogen/deuterium isotope substitution experiment at the WNR, compared with two theoretical simulations LCY (Lee, Clementi, and Yoshimini) and ST2 (Stillinger and Raman) based on model intermolecular water potentials. Water has stronger short range order (more ice-like) than either simulation.

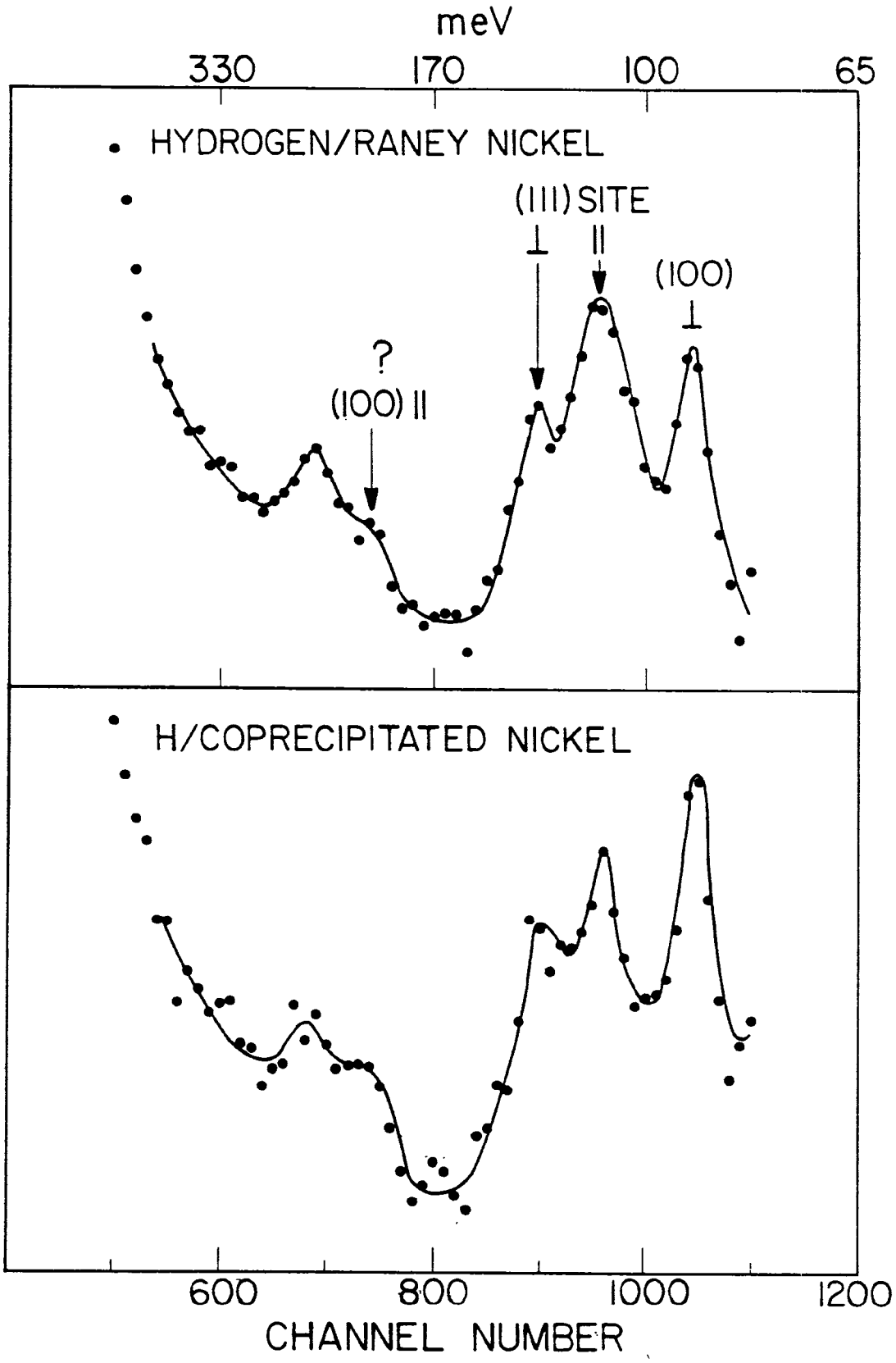

Fig. 7 Inelastic neutron scattering spectra of hydrogen chemisorbed on Raney nickel and coprecipitated nickel, as measured on the Be-BeO Filter Difference Spectrometer at the WNR facility.

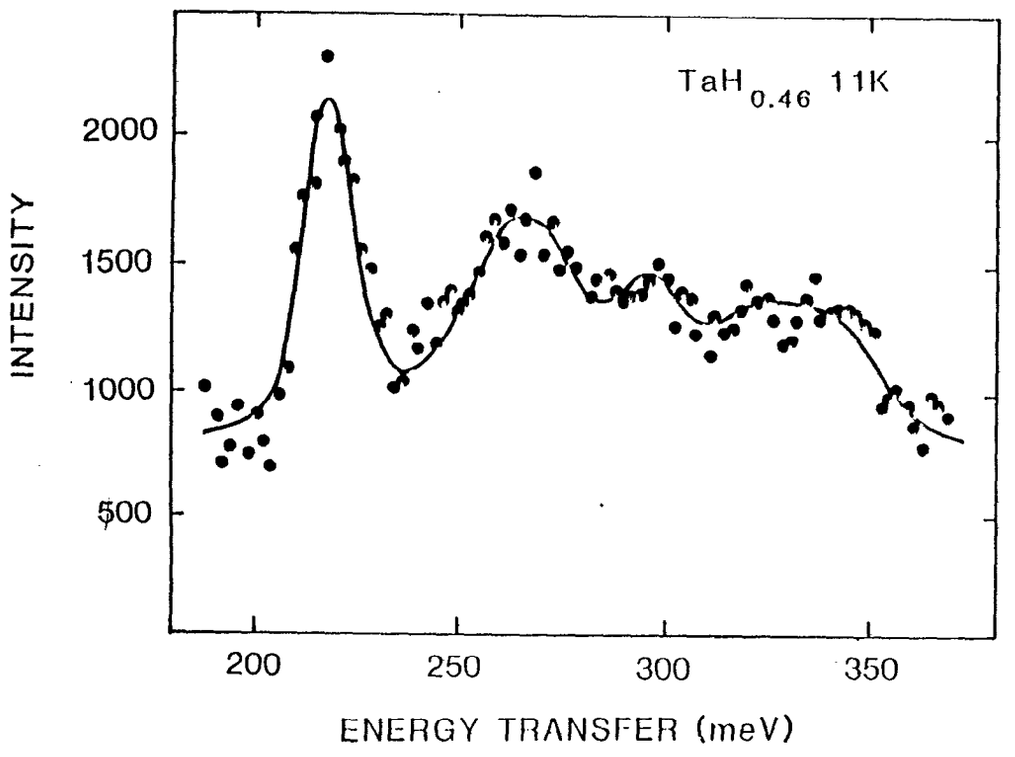

Fig. 8 High energy transfer part of the vibrational spectrum of $TaH_{0.46}$ as measured on the Be-BeO Filter Difference Spectrometer at the WNR facility.

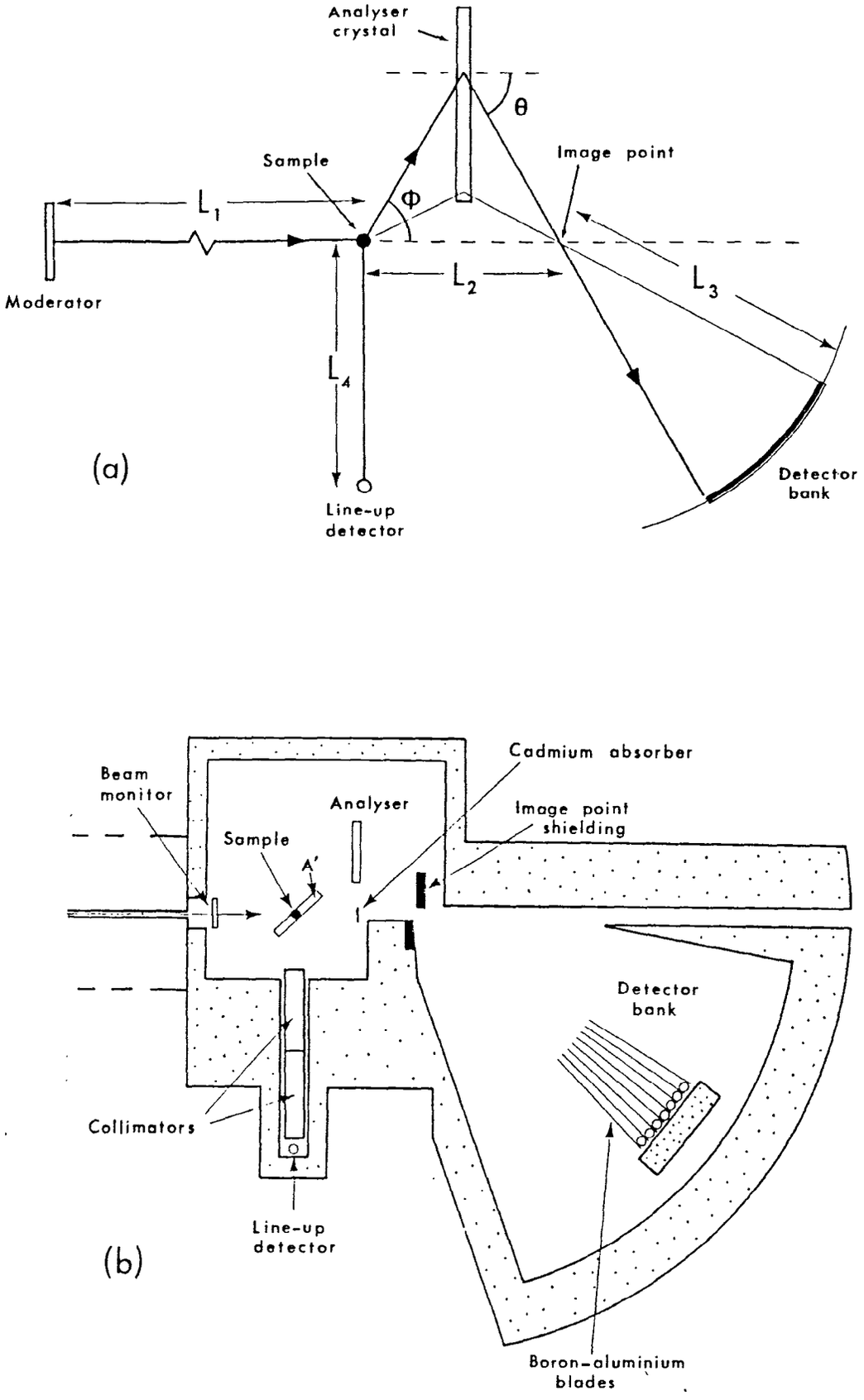

Fig. 9 Schematic diagrams of the Constant-Q Spectrometer at LANSCE, an instrument for the measurement of elementary excitations in single crystal samples.