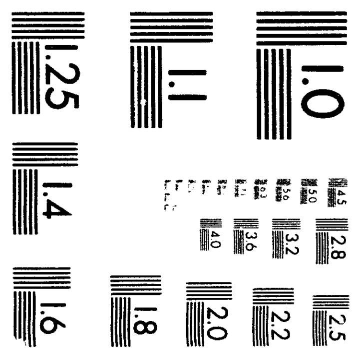

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# SURFACE PHYSICS WITH COLD AND THERMAL NEUTRON REFLECTOMETRY

Progress Report

for the Period from April 1, 1991 - September 30, 1993


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## I. ABSTRACT

Within the past two and one half years of the project "Surface Physics With Cold and Thermal Neutron Reflectometry" a new thermal neutron reflectometer was constructed at the Rhode Island Nuclear Science Center (RINSC). It was used to study various liquid and solid surfaces. Furthermore, neutron reflection experiments were begun at different laboratories in collaboration with Dr. G.P. Felcher (at Argonne National Laboratory), Dr. T. Russell (IBM Almaden) and Drs. S.K. Satija and A. Karim (at the National Institute for Standards and Technology). The available resources allowed partial construction of an imaging system for ultracold neutrons. It is expected to provide an extremely high resolution in momentum and energy transfer in surface studies using neutron reflectometry.

Much of the work reported here was motivated by the possibility of later implementation at the planned Advanced Neutron Source at Oak Ridge. In a separate project the first concrete plans for an intense source of ultracold neutrons for the Advanced Neutron Source were developed.

II. Status of the Project of Reflectometry at the University of Rhode Island

II.1 Description of the Reflectometer at the Rhode Island Nuclear Science Center

During the current project period we have set up the thermal neutron reflectometer at RINSC. The layout of this instrument is sketched in Figure 1. It is used for the study of surfaces and interfaces of nonmagnetic materials and it meets the design characteristics originally envisaged. The possibility of future upgrading to allow work with polarized neutrons on magnetic materials is discussed in the renewal application attached to this report.

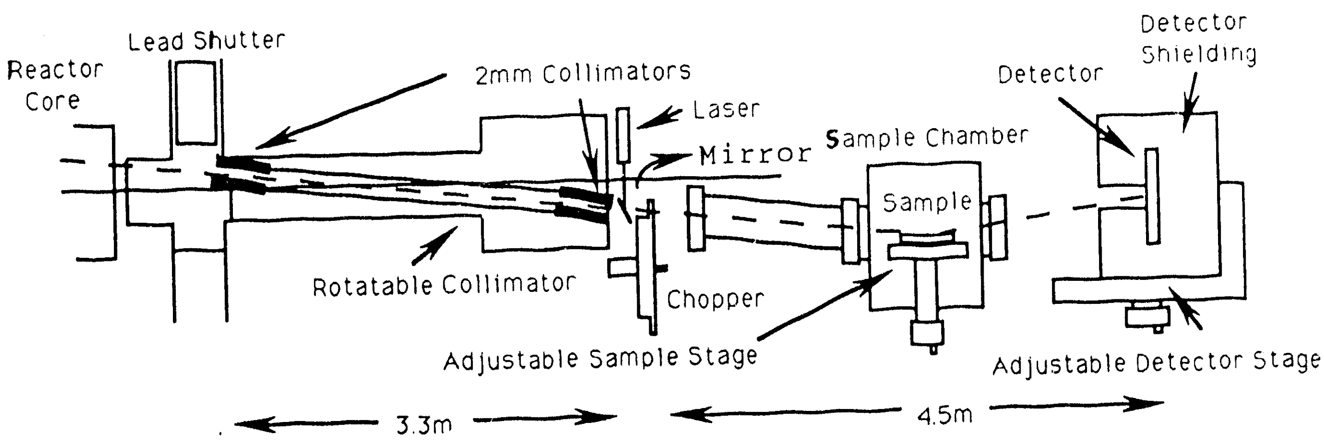


Fig. 1: Schematic view of the reflectometer at the Rhode Island Nuclear Science Center. A beam of thermal neutrons in pencil-like or slit geometry is selected by a collimator installed in a rotatable beam tube plug. A variable speed disk chopper provides a time of flight resolution of ≈ 5%

The present setup of the reflectometer at RINSC is shown in Figure 1. It has been described by Jeng et al. (1992), and in further technical detail in students' reports (Ross, 1990; Quagliato, 1992; Ehrle, 1993). A nearly horizontal beam of thermal neutrons is extracted from the $H_2O$ moderated, open-pool MTR-type research reactor operated at a power level of 2 MW. The thermal flux at the beam tube nose is about $5 \times 10^{12}$ $cm^{-2}s^{-1}$. A pencil-like beam with a divergence of $10^{-3}$ rad is defined by a collimator consisting of 30 cm long boron and lead containing polyethylene blocks, spaced 3 m from one another. Their 2 mm diam. holes were aligned with a precision of the order of $10^{-4}$ rad. Circular collimators, instead of slits, are used to facilitate measurement of diffuse scattering from surfaces and interfaces, but specular reflectivity data can also be taken with relaxed collimation. Scattering on air is reduced by evacuation of the collimator section. The beam plug containing the biological shielding and the tubular collimator can be rotated within the horizontal beam tube around a horizontal axis which is well defined by bearings made of plastic material. By rotating the plug the beam direction is changed continuously from slanting downward to upward, with a maximal angle of 0.03 rad as referred to the horizontal plane. In this way, the angle of incidence, $\alpha$, onto the horizontal sample surface can be adjusted to optimum conditions for a given experiment. The entire collimator unit is designed to be easily replaceable.

Although the facility is installed at a steady-state reactor we use the time of flight technique for wavelength variation since it allows the use of a constant angle of incidence, and hence of a constant beam geometry, for a given experiment. The chopper used consists of a 30 cm diameter disk with boron and cadmium shielding and a single slit of variable width. Alternatively, a different chopper disk with two slits can be used, providing a higher intensity for certain experimental requirements as well as a further improvement in shielding efficiency, due to the use of $^6LiF$ as the shielding material. The speed of rotation is variable from $n \approx 500$ to 2500 rpm and thus can be adjusted to optimal conditions where, in the relevant range of momentum transfer perpendicular to the sample surface, $Q$, the two contributions to resolution are matched: (1) The contribution $(\Delta Q/Q)_t = \Delta t/t$ due to the dispersion in flight time $t$ for an effective chopper opening time $\Delta t$ and a flight path of $L = 4.5$ m, and (2) the contribution $(\Delta Q/Q)_\alpha = \Delta\alpha/\alpha$ due to the angular beam divergence $\Delta\alpha \approx 7 \times 10^{-4}$ rad. The resolution is given by

$$\Delta Q/Q = \sqrt{(\Delta Q/Q)_t^2 + (\Delta Q/Q)_\alpha^2}. \qquad (1)$$

A typical value for $\Delta Q/Q$ is about 10% for $n = 1800$ rpm, $\alpha = 5 \times 10^{-3}$ rad, for a width of 3 mm of a single slit, and for a wavelength of $\lambda \approx 0.2$ nm.

The sample chamber is installed at a distance of ≈ 2.2 m from the chopper. It is equipped with an ion pump providing an oil-free sample environment when ultrahigh vacuum is required. The flight path between chopper and sample is also evacuated. Small angle scattering of the beam entering and exiting the chamber is avoided by the use of quartz glass windows. The sample can be mounted on the cold head of a closed-cycle helium cryogenic unit, allowing the sample temperature to be varied in the range from 10 K to room temperature with a fluctuation $\Delta T$ not larger than ≈ 0.5 K. A different mounting allows measurements at a temperature up to about 400°C.

For the measurements of specular reflectivity performed so far we used a single $^3$He detector mounted at a distance of 2.3 m from the sample and shielded from background by 20 cm of borated paraffin. A linear, position-sensitive $^3$He detector with a measured resolution of about 1 cm is available for diffuse scattering measurements.

Alignment of sample and detector is ensured by use of a He-Ne laser beam coinciding with the neutron beam. For this purpose a thin 45° mirror (silicon wafer) was installed in the neutron beam near the chopper position. The laser beam also allows a measurement of the angle $\alpha$ of beam incidence on the sample. Most reflectivity measurements were performed at an angle $\alpha = 0.26 \pm 0.02°$, with the beam slanting downward.

The beam intensity is monitored using a fission monitor detector in transmission, and the time of flight spectra are acquired with the help of a Nucleus personal computer and multiscaling system triggered by a photosensor signal at the time of chopper opening.

### II.2 Test Experiments and Applications

Fig. 2 shows the measured direct beam spectrum (points) as a function of wavelength, compared with a Maxwellian spectrum (dashed curve) for the same beam geometry and a thermal flux of $1.1 \times 10^{12}$ cm$^{-2}$s$^{-1}$. The effective temperature $T_{eff}$ = 60°C characterizing the spectrum is ≈20°C above the temperature of the $H_2O$ moderator of the reactor, as is common for a light-water moderated reactor source. The reduced flux and deviations from the Maxwellian distribution at long wavelengths are explained by absorption and scattering in the beam windows and along the 4 m of air traversed by the beam between the chopper and the detector. The increased intensity measured in the epithermal region is due to the radial beam-tube geometry which is usually used for this type of reactor.

At the time when this renewal application is being written the change-over of the RINSC reactor from highly enriched fuel elements to a compact core with low-enrichment fuel elements is being carried out. This upgrade is expected to provide an increased thermal neutron flux, and a further increase will result from the anticipated increase of reactor power from 2 MW to 3 MW which has been

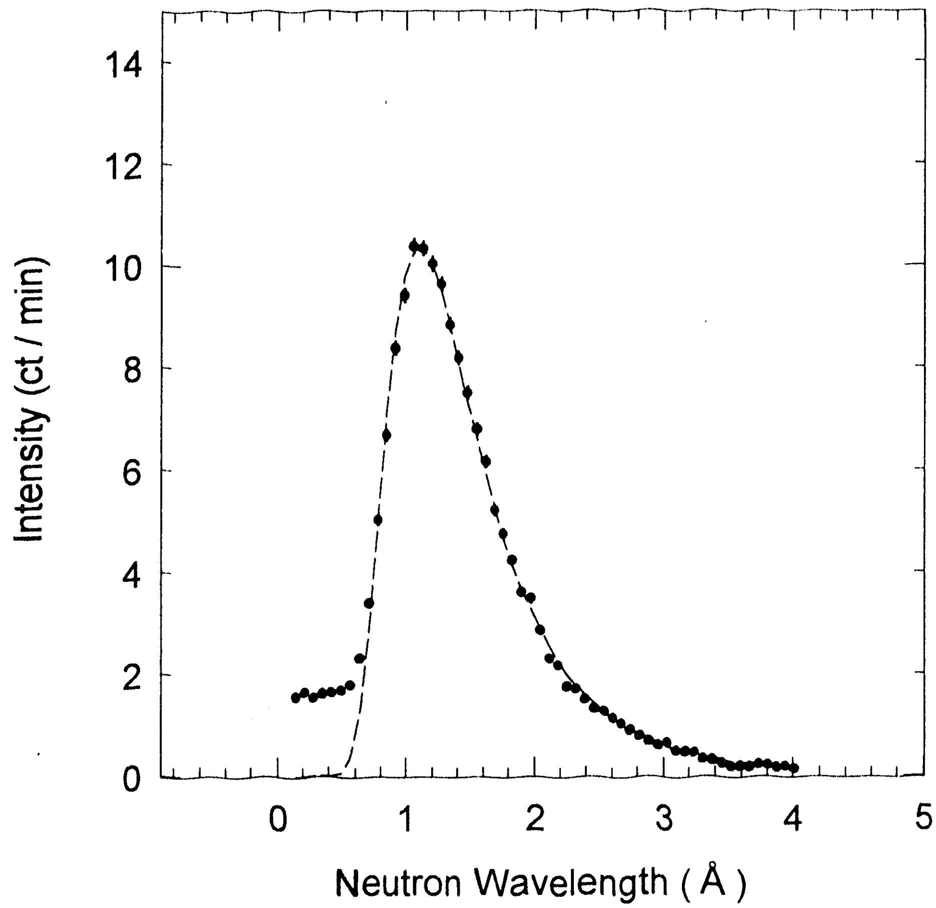


Fig. 2: The measured time of flight spectrum for the reflectometer (points) is compared to a Maxwellian spectrum corrected for beam absorption (dashed curve).

made possible by the installation of a new cooling tower for the secondary cooling water. A further intensity gain in the cold neutron wavelength region is expected if the current plans to install a Cold Neutron Source (possibly based on a solid moderator like methane) can be realized. A shift to longer neutron wavelengths would obviously be especially beneficial for neutron reflectometry.

The measurements performed so far on the thermal neutron reflectometer were mainly intended to test and characterize the new instrument and beam. Using the full neutron beam total cross section measurements as a function of wavelength and temperature were performed for single crystals of copper, lead and aluminum. The results are in agreement with previous cross-section data. Specular reflectivity data were obtained for liquid $D_2O$, for a polarizing supermirror, and for Fomblin oil (a perfluorinated polyether of the form $CF_3$-[$(O\text{-}CF\text{-}CF_2)_n$ - $(O\text{-}CF_2)_m$]-O-$CF_3$, with $n/m \approx 100$) (Jeng et al., 1992). This type of Fomblin oil had been used earlier as a thin film coating the glass walls in ultracold neutron (UCN) containment experiments determining a precise value of the neutron lifetime (Mampe et al., 1989). So far, our attempts to measure capillary waves at an oil-water interface "marked" by a thin surfactant layer have not been successful, encountering a number of technical difficulties related to sample preparation and low neutron flux. The neutron flux was too low for the measurement of diffraction (first order) from a ruled optical reflection grating, since such an experiment requires a highly monochromatic beam.

## III. Reflectometry with Ultracold Neutrons

To demonstrate the excellent resolution both in momentum and in energy transfer, attainable if extremely slow (very cold or ultracold) neutrons are used for reflectometry, a series of reflectivity measurements were performed at the Institut Laue-Langevin (ILL), using the so-called UCN Gravity Diffractometer (Scheckenhofer and Steyerl, 1977) installed there at the high-intensity Turbine Source of ultracold neutrons (Steyerl et al., 1986). The results of precise beam profile measurements for beams reflected on neutron mirrors as well as for beams diffracted from a linear, ruled diffraction grating, were summarized by Steyerl et al. (1992).

The main results are as follows: Given the intensity at the ILL source and the high resolution afforded by the use of gravity in the UCN diffractometer (where 1 cm of fall height corresponds to a change of neutron energy by 1 neV) it is straightforward to achieve a value of $\delta E = 2 \times 10^{-11}$ eV for the minimum resolvable line broadening in quasi-elastic UCN reflection. This value also corresponds to a resolution $\delta Q \approx 5 \times 10^{-6}$ nm$^{-1}$ for line broadening induced by small angle scattering at the surface. These values for $\delta E$ and $\delta Q$ are unsurpassed by any other neutron technique available at present.

## IV. Project of a Two-Mirror Imaging System for Ultracold Neutrons

### IV.1 Overview

The imaging system for ultracold neutrons sketched in Fig. 3 is presently under construction at the University of Rhode Island. Its

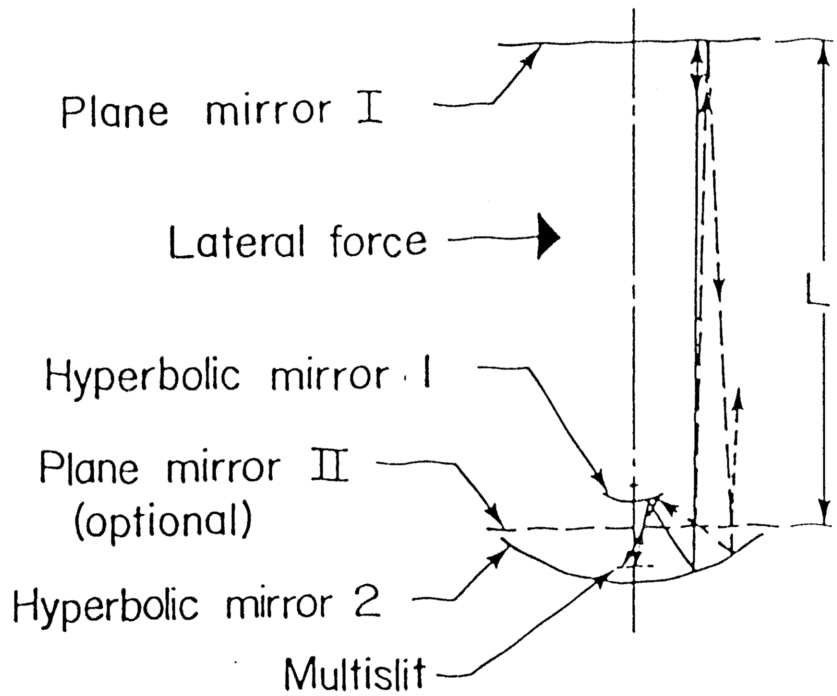


**Fig. 3**: Proposed imaging system for measuring very small deflections of an ultracold neutron beam induced by slightly non-specular surface reflections or by the action of very small forces (as indicated schematically). A multislit grating is imaged onto itself by the two hyperbolic mirrors 1 and 2. In the "storage mode" of operation a pulsed UCN beam is admitted into the space between the planar mirrors I and II where the neutrons bounce back and forth a number of times before being released back to the imaging mirrors when mirror II is moved out of the beam. For the prototype version the neutrons make only one round trip, and mirror II is not needed. The total beam deflection is registered as an image shift.

design relies on the unique property of ultracold neutrons to experience total external reflection on suitable mirror materials at any angle of incidence, even normal to the surface. In the same way as for the optics of visible light, reflection at normal incidence is a crucial requirement for the possibility to design the imaging systems with very small aberrations needed, e.g., for neutron microscopy (e.g., Frank, 1987; Steyerl et al., 1988a) or for the investigation of extremely small surface effects proposed here. The two-mirror system was discussed previously by Steyerl et al. (1988b) and by Jeng et al. (1992). It is expected to improve the resolution in momentum transfer to $\Delta Q = 10^{-8}$ nm$^{-1}$. This is about two orders of magnitude better than the present "state of the art". So far, a best resolution of $\Delta Q \approx 10^{-6}$ nm$^{-1}$ has been attained, both for existing UCN diffractometry (Steyerl et al., 1992) and for perfect crystal dynamical diffraction making use of the central peak in the Borrmann fan (Bonse et al., 1979; Rauch et al., 1983).

The level of sensitivity expected for the UCN two-mirror system would be unmatched by any known experimental technique using beams of massive particles other than the neutron. In addition to surface physics applications the proposed system could be used for the investigation of very small forces, as for instance in a search for an electric charge of the neutron. The current upper limit on a possible residual electric charge, $q_n$, of the neutron was set in an experiment of Baumann et al. (1988) who used an optical imaging system for cold neutrons and obtained the result $q_n = (-0.4 \pm 1.1) \times 10^{-21}$ proton charges.

Fig. 3 shows the basic design of the UCN imaging system. Using a horizontal beam geometry a multi-slit object is imaged onto itself, with unit magnification. Imaging with very small residual aberration is accomplished by the use of two cylindrical mirrors with optimized hyperbolic shape (labeled 1 and 2 in Fig. 3). A pulsed neutron beam, with a pulse duration of about 0.5 s and an interval of several seconds between the pulses, penetrates the object slits - a stack of transparent sections separated by opaque regions of the same width - and is admitted into the space between the hyperbolic imaging mirror 2 and the planar mirror I used in back-reflection setting. The flight path between these mirrors is about 1 m long. After one round trip the neutrons return to the object plane.

The operation of this system can be visualized by noting that the image formed by the returning neutrons can be made coincident with the multi-slit grating by a precise adjustment of the rocking angle of mirror I about a vertical axis. For this setting, the returning neutrons would find the open spaces of the grating and thus be completely transmitted through the multi-slit, provided that the optical system performs perfectly well. That is to say, that any blurring of the image due to aberrations or other imperfections are negligible. In this setting the multi-slit, now serving as the image analyzer, will be perfectly transparent, and all the returning neutrons will be counted by the detector. The detector is positioned below the multi-slit, as indicated in the three-dimensional view presented in Fig. 4, in order for the neutrons to gain enough kinetic energy by falling through a certain distance to be able to penetrate the detector window.

By a very small adjustment of rocking angle the planar mirror I can be rotated to a position where all the returning neutrons hit the opaque (rather than the transparent) sections of the multi-slit and thus will all be blocked. In this other extreme case, the transmitted beam intensity will be a minimum. The rocking angle necessary to change from maximum transparency to an adjacent position of minimum transparency (i.e., by half an interference fringe) is estimated to be of order 10 $\mu$m/1 m $\approx 10^{-5}$ rad for a multi-slit with widths of 20 $\mu$m for the open and the opaque sections, and for a distance of 1 m between the mirrors.

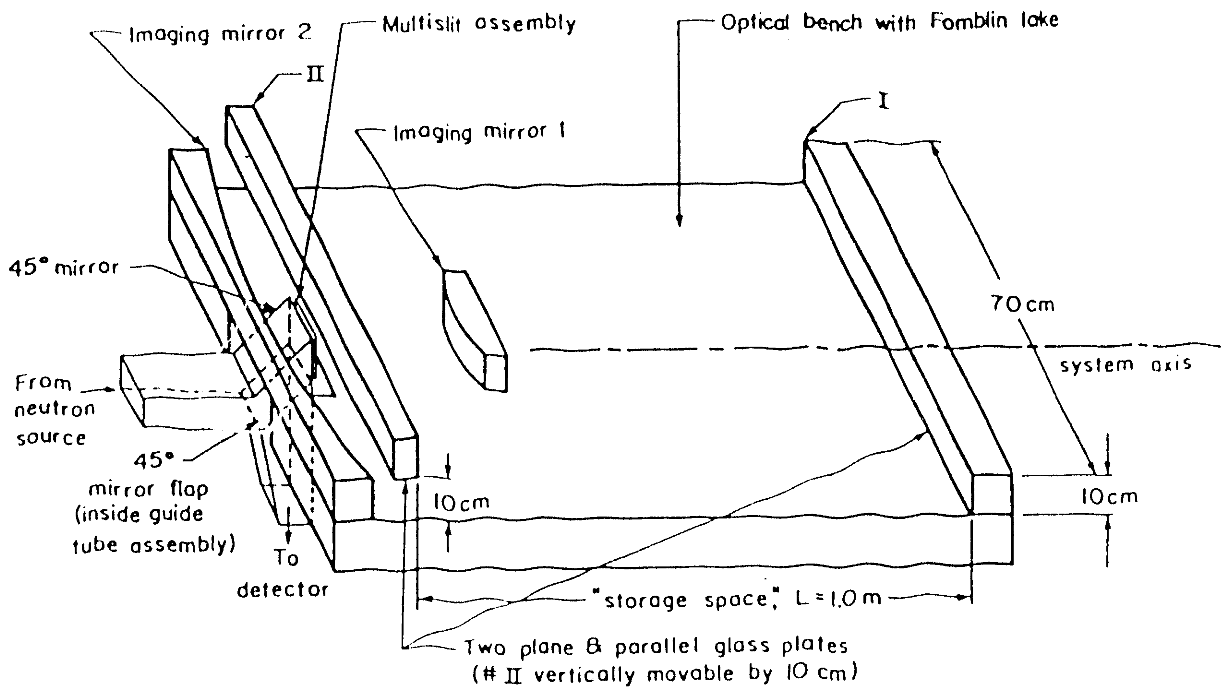


**Fig. 4**: Three-dimensional view of the UCN imaging system. A lake of Fomblin oil on a vibration-isolated table serves as the horizontal mirror on which the UCN hop along their way. The optional flat II is needed only for the "storage mode" of operation. It is moveable in the vertical plane to allow, sequentially, the storage space to be filled, closed, and emptied.

The main purpose of this system is to measure very small deflections of the neutron beam induced, not by a rotation of a mirror but, e.g., by small lateral forces acting on the neutron beam. The possibility of mirror rotation is necessary for adjustment and calibration. The forces to be studied could be due to the Coulomb interaction of a small, residual electric charge of the neutron with a strong static electric field set up in one arm of the imaging system, or due to the gravitational interaction with a laboratory mass. The beam curvature due to such forces is shown schematically in Fig. 3. Alternatively, a slightly non-specular reflection on a sample mirror inserted into the neutron path in a way indicated in Fig. 5, will give rise to a deflection of the beam by a small angle, as compared to the specular beam.

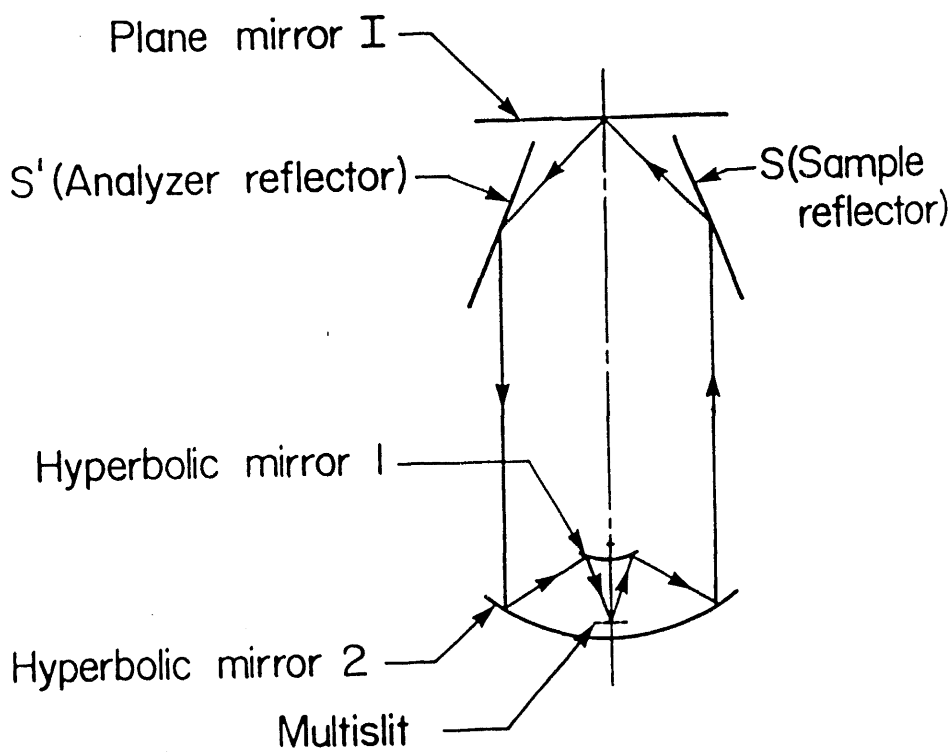


**Fig. 5**: Experimental scheme allowing the beam profile for reflection at a sample reflector (S) to be scanned with extremely high angular resolution, corresponding to a Q-resolution of $\Delta Q \approx 10^{-9}$ Å$^{-1}$. Scanning is performed by the precise rotation of sample S and the analyzer mirror S'.

## IV.2 Present Status of Construction of the Ultracold Neutron Imaging System

Using the funds available for the current budget period (grant Nr. DE-FG02-91ER45445) it was possible to procure the following basic components of the UCN imaging system:

(a) The vacuum chamber including a three-layer mu-metal magnetic shield. A detailed drawing of this system is shown in Figs. 6a (side view perpendicular to the axis) and 6b (side view along the axis). The chamber is designed and has been tested for a vacuum of about $10^{-6}$ mbar. It is equipped with a cryogenic and a mechanical primary pump and all necessary vacuum gauges. The magnetic shield consists of three concentric circular cylinders of mu-metal of 2 mm thickness. The spacing between the cylinders is optimized for best shielding. The shielding system provides sufficient shielding from, both the static magnetic field of the earth and neighboring equipment, and the dynamic magnetic fields induced mainly by the operation of the overhead crane. The calculated shielding factors for static fields are in excess of 1,000 for a magnetic field longitudinal to the system axis, and of 5,000 for a field perpendicular to the axis.

(b) The plane mirror labeled I in Fig. 4. This mirror is used in back-reflection, and it serves to reverse the flight direction of the parallel beam of neutrons formed by the imaging mirrors 1 and 2. This mirror is made of fused silica without surface coating, and it meets the required specifications: (i) Overall flatness with a maximum shape deviation of 0.25 optical wavelengths per inch; (ii) Mean square micro-roughness less than 1 nm. This important component of the imaging system will be delivered and tested in early 1994.

(c) The vibration isolation system. As shown in Figs. 6a and 6b, the optical mirror system is set up on a bench which is mechanically separated from the surrounding vacuum vessel by soft bellows, and which is supported by the vibration control system. The vibration isolation system is designed to suppress the maximum vibration velocities for all imaging and back-reflecting mirrors to the acceptable level of about 0.5 $\mu$m/s at the low frequencies of about 2 Hz which are most critical for the Imaging System. This frequency corresponds to the time needed by a neutron to complete a full round trip through the system.

Since soft, welded bellows separate the optical table from the vacuum vessel and all other components like the pump system and guide tube connection to the Source, it is not necessary to provide vibration isolation for the instrument as a whole but the selective isolation provided for the optical table only is sufficient.

As a basis for the lay-out of the vibration-isolation system, vibration spectra were measured very close to the anticipated site

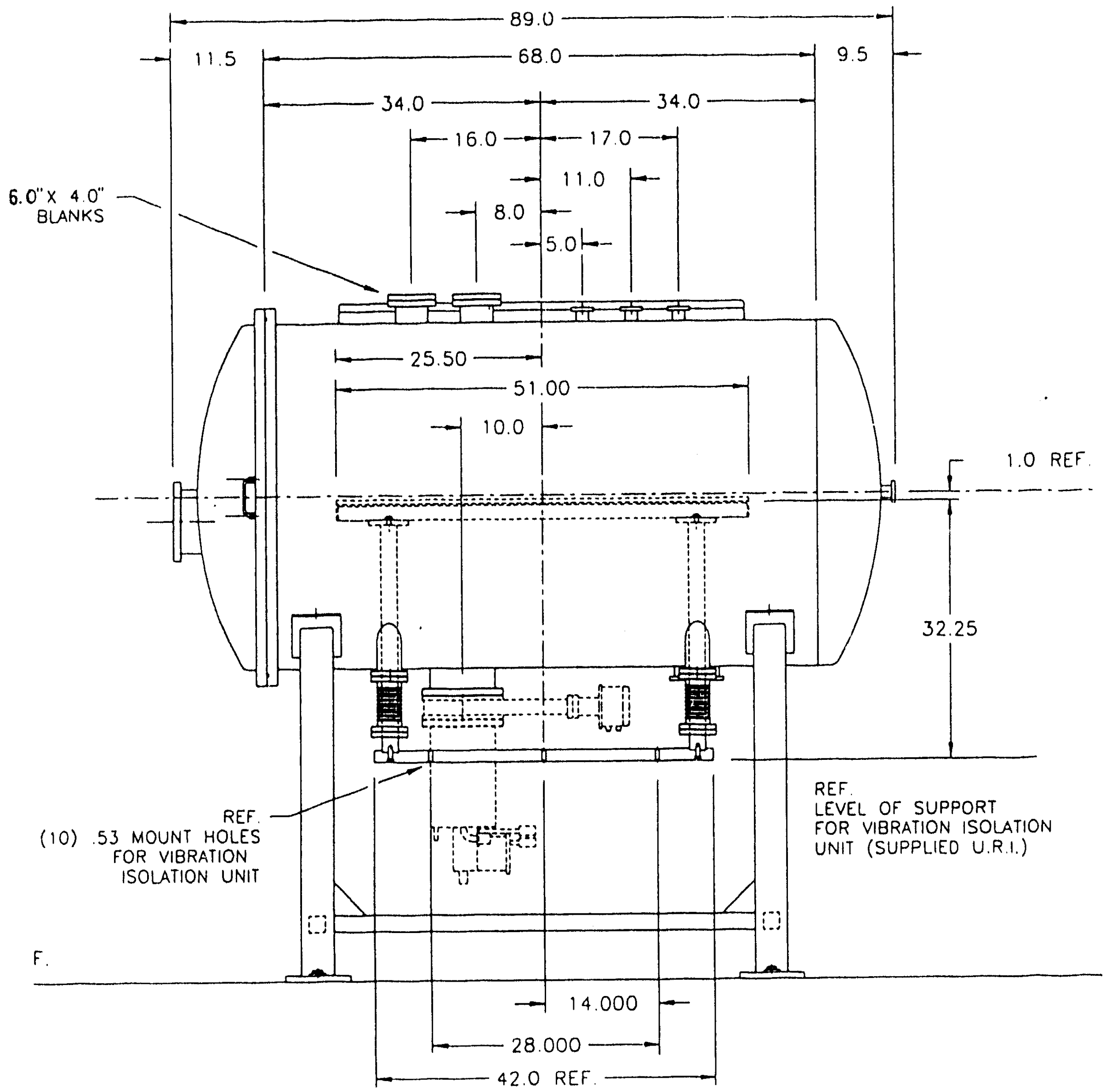


**Fig. 6a**: Side view of the UCN Imaging System (viewed perpendicular to the system axis).

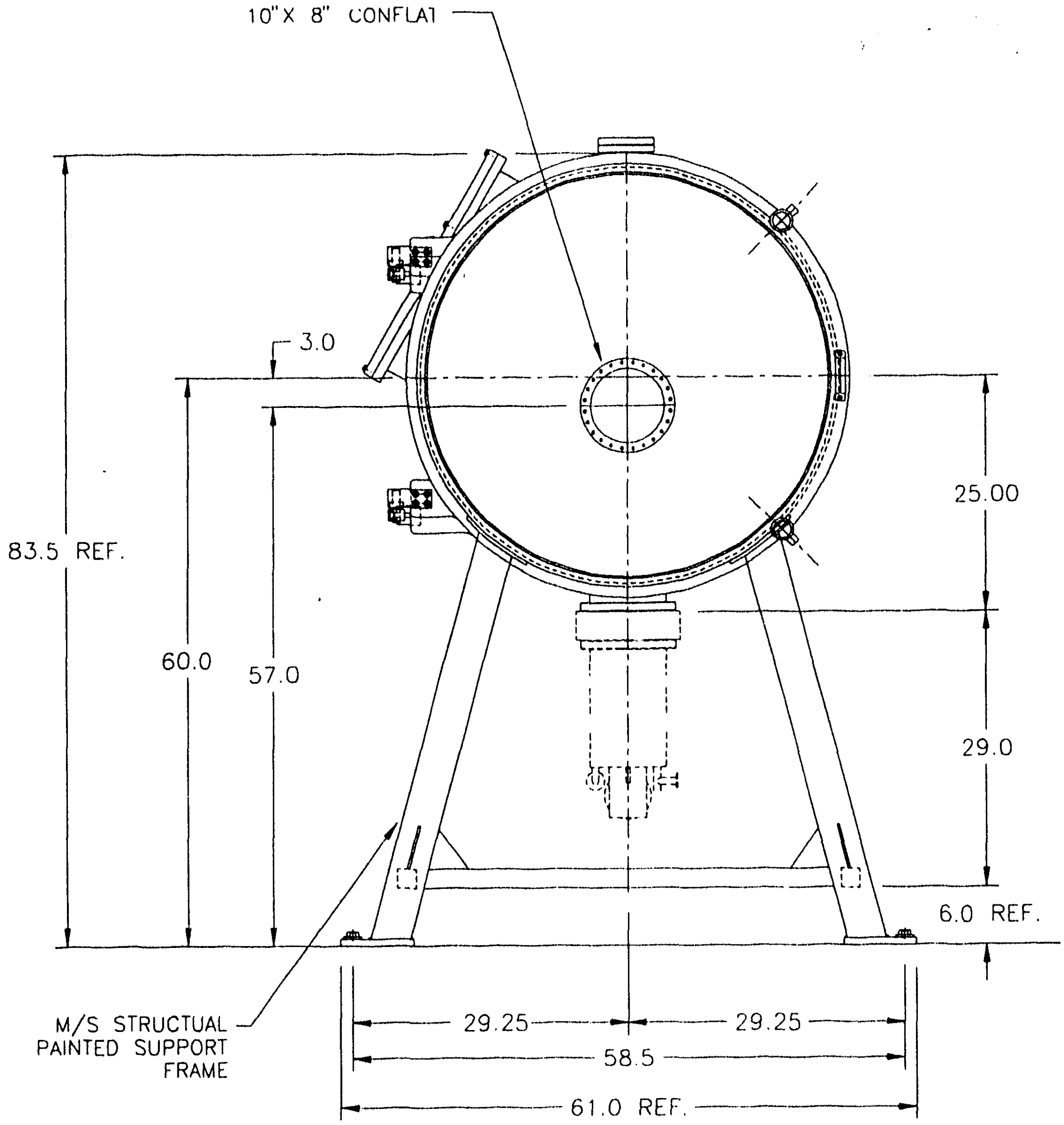


**Fig. 6b**: Side view of the UCN Imaging System (viewed along the system axis).

of the UCN Imaging System on "Level D" of the ILL reactor, at full reactor operation (i.e., with the primary water pumps and all other sources of vibration in operation). The spectra were taken using a commercial Bruel & Kjaer vibration analyzer system, and they included both vertical and horizontal motions. On the basis of this data we chose an "active" control system with a feed-forward and feedback servo-loop control system acting on six degrees of freedom (for position and rotation). The term "feed-forward control" is used to indicate that the motion of the floor is used to counteract the normal reaction to the system to it before the system has the time to respond to this motion. The vibration control system has been ordered and it will be delivered in early 1994.

Among the items which exceeded the present budget limits and could therefore not yet be procured, are the two curved imaging mirrors. They constitute the main components of the imaging system. A very extensive search for potential bidders with the necessary experience in high-quality mirror production was conducted in the U.S. The search was complicated by the requirement of cylindrical mirrors for our neutron-optical system. Most potential suppliers are only equipped for the mirror systems with an axis of revolution that are commonly used in imaging systems for visible light. The difficulties encountered so far led us to consider a relaxation of our original specifications on optical quality.

The relaxation concerns the maximum tolerable total aberration, i.e., the degree of image blurring. An increase of total aberration from the original value of 20 $\mu$m to about 100 $\mu$m would affect the sensitivity of resolving a small lateral beam deflection, deteriorating it by a factor of about three. The deterioration is smaller than the factor of five by which image blurring increases, since the loss in image quality is partly compensated by an increased neutron count-rate. Unfortunately, even for these relaxed specifications it was not possible so far to procure imaging mirrors at a cost commensurate with the present budget limits. The latest estimates provided by the optical industry indicate that the cost for the two imaging mirrors will be between \$ 150,000 and \$ 175,000.

Completion of the UCN imaging system will require also the purchase of a piezo-electric system capable of controlling the rocking angle of the plane mirror with the required precision of about $10^{-7}$ radians. The remaining components of the UCN Imaging System can be fabricated at low cost in the URI workshops: The bench for the optical mirror system, the neutron guides connecting the experiment to the UCN "Turbine Source", the multi-slit and the neutron detector. All these items are described in further detail in the attached renewal application.

## V. Current Reflectometry Experiments Conducted at Argonne and at NIST on a Collaborative Basis

Two sets of neutron reflectometry investigations are currently being carried out by a postdoctoral fellow, Dr. H. Lin, in collaboration with scientists at Argonne National Laboratory (Dr. G.P. Felcher), at the IBM Almaden laboratories (Dr. T. Russell) and at the National Institute for Standards and Technology (Drs. S.K. Satija and A. Karim):

### V.1 Study of an Iron-Chromium Multilayer

Layered structures of Fe and Cr have been the object of intensive study (Barthélémy et al., 1990; Parkin et al., 1991; Endoh, 1992) ever since giant magnetoresistance has first been observed in this system (Baibich et al., 1988; Barthélémy et al., 1989). More recently, granular mixtures of various pairs of materials have also been shown to exhibit a significant magnetoresistivity (Berkowitz et al., 1992; Xiao et al., 1992). In the layered Fe-Cr system the magnetoresistance was shown to be related to the tendency of neighboring iron layers to align antiferromagnetically for certain intervals of chromium layer thickness. Several possible mechanism have been proposed which could explain the persistence of antiferromagnetic ordering through the intervening chromium layer.

While the ordering phenomenon is very interesting in itself and the various theories trying to understand it are under discussion, its relation to the appearance of magnetoresistance is established by the notion of strong spin dependence of conduction electron scattering at the chromium-iron interfaces, an effect related to the spin-dependent impurity scattering observed in bulk ferromagnetic transition metals (Fert and Campbell, 1976; Campbell and Fert, 1982). In this view the observed decrease of electrical resistivity at higher applied magnetic field strength is ascribed to the transition from antiferromagnetic to ferromagnetic interlayer ordering. For pronouncedly spin-dependent electron scattering at the chromium-iron interfaces the change in magnetic ordering will obviously be observed as a significant change in electrical resistivity.

The Argonne experiments in which Dr. Lin is participating use a novel Fe-Cr multilayer system prepared at Argonne National Laboratory. The superstructure is sputter-deposited on a substrate of monocrystalline magnesium oxide. The measurements are performed at the polarized neutron reflectometer POSY I at the Intense Pulsed Neutron Source (IPNS). At the time the present renewal application was written the data evaluation was in progress. The analysis uses a fitting routine suited for polarized neutron reflection from magnetic multilayer systems with parallel or antiparallel interlayer ordering of magnetization in the ferromagnetic layers.

In a phenomenological approach similar to an analysis by Krebs et al. (1990), the experimental data can be interpreted with respect to the parameters in a model where the magnetic superstructure is considered to be the result of competition between three contributions to the total energy: (1) the exchange energy between the ferromagnetic layers; (2) the anisotropy energy; and (3) the Zeeman energy in an applied magnetic field. In general, the competition between these terms may give rise to deviations from a strict alignment of layer magnetization in a simple ferromagnetic (...+++...) or antiferromagnetic (...+-+-...) superstructure.

We have also developed a new algorithm for calculating reflectivities for a more general magnetic structure where the magnetization in a layer can point in any arbitrary in-plane direction. It is not based on the well-known matrix method (Felcher et al., 1987) but consists in a generalization of Parratt's recursion method for the reflectivity in a nonmagnetic superstructure (Parratt, 1954) which allows to take into account surface and interlayer profile blurring in a simple, approximate way, using the well-know Debye Waller type factor. Our adaptation of this method to magnetic superstructures allows us to interpret polarized neutron reflection data also with respect to possible deviations from strict ferromagnetism or antiferromagnetism.

V.2 Investigation of a Diblock Copolymer System

In a way similar to magnetic multilayers, the strong interest in the properties of block copolymers derives, both from their extreme economic value and from the fact that they serve as unique model systems for a number of theoretical problems related to critical phenomena and scaling laws (De'Bell and Lookman, 1993). Due to the enormous versatility in morphology and chemical properties characteristic of polymers in their different states (in solution or in melted or solid form) they provide unique laboratory systems for the study of various aspects of phase transitions. As an example of recent applications of block copolymers we emphasize their use in micro-electronics and biomedicine as surfactants, compatibilizing agents, and adhesives (e.g., Bucknall et al., 1992).

For all these applications it is essential to understand the behavior of block copolymers at surfaces and interfaces. For the group of diblock copolymers (consisting of two kinds of polymers chemically linked together in a long chain), surface induced lamellar ordering has been well studied in the system PS-PMMA (polystyrene-polymethylmethacrylate) deposited in thin films on silicon substrate (Anastasiadis et al., 1989; 1990; Menelle et al., 1992). Previous neutron reflectivity measurements have shown that PS is located preferentially at the air-copolymer interface, and PMMA at the silicon substrate. The concentration profile is well described as a multilayer comprised of PS and PMMA microdomains oriented parallel to the surface of the substrate. The interface between the microdomains was found to be about 5 nm thick, independent of the molecular weights of the two block copolymers.

Our current study focuses on the swelling of this system in environments of, either a preferential solvent for PS or for PMMA, or of a good solvent for both blocks. Ordinary (protonated) cyclohexane and deuterated cyclohexane are chosen to investigate the asymmetric case. They are a preferential solvent for polystyrene. So far we have performed neutron reflectivity experiments on ordered dPS-hPMMA and dPS-dPMMA films (where the letters h and d indicate, respectively, "protonated" and "deuterated"). The samples are in equilibrium with h-cyclohexane and d-cyclohexane vapor. These experiments are performed at the BT-7 reflectometer (using a vertical sample geometry) and at the new NIST/IBM/U. Minn. reflectometer (using a horizontal sample geometry), both installed at NIST. The data is presently being analyzed.

## VI. Theoretical Work Related to Neutron Reflectivity

Our work on the theoretical analysis of reflection and refraction at rough surfaces (Steyerl et al., 1991) was continued by a graduate student (L.R. Iyengar). She obtained numerical data on diffuse scattering distributions for a variety of parameters characterizing the surface roughness. Furthermore, an analysis of reflectivities of magnetic multilayers consisting of a sequence of two different ferromagnetic materials was carried out for temperatures near the critical point. Such systems had been first analyzed by Fishman et al. (1987) for the example of a multilayer consisting of EuS and EuO. Aspects of dynamical behavior were theoretically investigated by Schwenk et al. (1988).

In a separate study we considered a possible application of very slow neutron reflectometry to investigate the liquid-vapor phase transition very near the critical temperature. Experimental work on this subject is usually complicated by the role of gravity in laboratories on Earth. (This is the reason why several experiments have been performed under conditions of micro-gravity in a Space Shuttle.) According to our analysis (Ludecke et al., 1991) it appears feasible to obtain the exponent $\beta$ describing the critical behavior with minimal gravity correction in comparatively simple experiments determining the reflectivity of very cold neutrons from liquid-vapor interfaces. The specific example used in this analysis was helium-4.

## VII. Personnel, Students and Guest Scientists

So far, four graduate students have been engaged in the development of the thermal neutron reflectometer at RINSC and with experimental reflectometry work. Steven J. Quagliato obtained his degree of Master of Science in December, 1992. U-Ser Jeng is expected to receive his Ph.D. degree in 1994. A further graduate student (Lakshmi R. Iyengar) is involved with analytical work in connection with surface roughness and magnetic multilayer properties. Recently, Levon Esibov has joined our group. His graduate research will be focused on the development of a three-dimensional spin analysis option for the thermal neutron reflectometer at RINSC.

The position of post-doctoral fellow has been filled in June, 1992, by Dr. Hong Lin. She has carried out reflectometry studies at

Argonne National Laboratory (on a magnetic multilayer, in collaboration with Dr. G.P. Felcher) and at the National Institute of Standards and Technology (on polymers, in collaboration with Dr. T. Russell, IBM Almaden, and Drs. S.K. Satija and A. Karim at NIST). These studies are continuing.

Two students of a German Technical College (Fachhochschule Heilbronn) were able to obtain scholarships from German sources allowing them to join our group for six months each. Gerhard Ross performed much of the early overall design work for the reflectometer (Ross, 1990) and Thorralf Ehrle laid out and installed the cryogenic system for the reflectometer (Ehrle, 1993). Recently, Dr. Winfried Drexel of the Institut Laue-Langevin, Grenoble, joined our group for four months (from April 1 until July 31, 1993) to help us in the coordination of the collaboration with the ILL.

Joseph Micciche (technician, level III) has provided technical help from the beginning of this project. Further technical support is available from the Machine and Electronic shops of the Physics Department.

The principal investigator (A.S.) has devoted approximately 50% of time during the academic year and 70% of time during the summer months to the various aspects of this project. The second project in which the P.I. is presently involved (together with post-doctoral fellow Dr. M.L. Crow) is concerned with complementary research: The development of a Source of Ultracold Neutrons for the Advanced Neutron Source presently planned at Oak Ridge.

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## X. Theses and Major Reports by Students

Th. Ehrle (1993), "Cryogenic Cooling System for Material Samples Used in Neutron Scattering", Techn. Report, Univ. Rhode Island.

S.J. Quagliato (1992), M.Sc. Thesis "A Neutron Reflectometry Study of Liquid Deuterium Oxide, Fomblin Oil and a Supermirror", Graduation Dec. 1992 from the Univ. Rhode Island.

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