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The cold neutron source for the FRG-1 research reactor at the GKSS research centre

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Abstract

The changing requirements and the demands for higher neutron flux, especially for high wavelength or cold neutrons ($\lambda > 5$ Å) in the beam tubes of the research reactor leads to the installation of the cold neutron source and other upgrading measures (reduction of core size by a factor 2, installation of a beryllium block reflector around the beam tubes, new neutron guide tubes etc.) at the research reactor.

The increasing amount of cold neutrons will be used mainly in small angle neutron scattering (SANS) experiments. Such experiments using larger wavelengths are necessary for the investigation of larger defects in materials. This technique (SANS) is used in fundamental physics, material sciences, chemistry and biology.

The paper gives a short overview on existing Cold Neutron Sources (CNS), describes the design - design principles, design considerations and especially safety requirements - of the CNS now under construction for the FRG-1 at GKSS, Geesthacht, some aspects of the upgrading of the research reactor. Important design features to be mentioned are the use of supercritical hydrogen, cooling by forced convection and the triple containment for safety reasons. The safety aspects will be discussed in detail.

The paper also discusses the good operational experience gained with the CNS in operation since more than 10 years at DR-3 of the Risø National Laboratory. The RISØ-CNS is the design basis for the new CNS at GKSS.

The cold neutron source for the FRG-1 research reactor at the GKSS research centre

1. Introduction

So called cold neutron sources find today an increasing interest. The combination of the installation of a cold neutron source, installation of neutron guides and improvements of the research reactor design will increase the low energy (high wavelength) neutron flux by some decades. The portion of the CNS to this improvement is more than one decade. To realize this decade or an additional decade it is absolutely necessary to analyse first the present situation at the research reactor facility. An optimization should include the reactor core, the reflector, the utilization, the beam tube arrangement and the neutron spectra. At the GKSS research centre such an analysis was done showing interesting possibilities for increasing the low energy neutron flux e.g. by a factor of 130 for 10 Å neutrons. Therefore it was decided and contracts signed to realize this latest in 1987.

2. What is a cold neutron source (CNS) /7/

A cold neutron source (CNS) is no neutron source like a reactor (fission source) or a (γ, n)-neutron source or spontaneous fission source (Cf^{252}). In a CNS no neutrons are generated. Only neutrons having thermal energies are slowed down to subthermal (cold) energies using a simple, complex, complicated and expensive facility. In general a CNS is a small volume filled with a low temperature moderator in the reflector of a research reactor (Fig. 1, 2).

2.1 Existing CNS

The CNS-situation (Table 1)

- CNS sources in reactors shutdown like FR-2 and EL 3 are out of operation,

- More than ten (10) CNS are in operation mainly in heavy water reflected, high power beam tube research reactors.
- The success of the operation and utilization enhanced the decision in many research centres to install such powerful instruments, too. For this reason six CNS are under construction and some projects with great chance of realization are existing.

It is evident that there is an enormous increasing need for such research tools.

2.2 Cold neutrons

Neutrons are grouped into energy ranges. Reactor physicists are more familiar with corresponding values in (eV) but solid state physicists prefer to speak in wavelength λ (Å). The relation between energy and wavelength for neutrons is

$$\lambda = \frac{0{,}286\ (\text{Å})}{\sqrt{E\ (\text{eV})}}$$

Table 2 gives some figures for the energy range 10 eV ... 10^{-6} eV (epithermal, thermal, cold, ultra cold neutrons) and the corresponding wavelength from 0,09 Å ... 286 Å. Especially the energy region from $5 \cdot 10^{-3}$ eV ... 10^{-5} eV (λ = 4 Å ... 90 Å) is the range of the so called cold neutrons.

2.3 Cold moderator

2.3.1 General demands

If an effective moderator is wanted some general demands have to be met:

- avoid upscattering of cold neutrons e.g. no water layers between CNS and experiment
- low atomic number Z of the moderator material for effective slowing down
- extremely low temperature for this moderator material for getting as much as possible cold neutrons
- low absorption cross section
- efficient scattering cross section
- sufficient high density of the reflector to enable effective slowing down
- no radiation damage by the γ-radiation
- heat transfer conditions must be evaluated satisfactory

All these conditions leads directly to hydrogen and deuterium as cold moderator. A high atomic density can be achieved in using organic H, D compounds or ice. In research reactors with more than negligible power both possibilities cannot be used as

- organic compounds will be cracked in high γ-radiation fields
- due to the high γ- and neutron fluxes a considerable amount of energy is absorbed in the moderator and its cladding. This energy (heat) cannot be removed if ice is used.

Therefore the only realized possibilities for effective cold neutron source moderators are hydrogen or deuterium as gas or liquid at temperatures between 20 K and 35 K.

2.3.2 Properties of hydrogen

Of fundamental interest for the design of a cold neutron source are the cryogenic, nuclear, heat transfer, explosive and corrosive properties of hydrogen. In hydrogen molecules the spin of the two protons can be aligned (orthohydrogen ↑↑) or opposite (parahydrogen ↓↓). From Fermistatistics for $I = \frac{1}{2}$ the expectation value for the distribution of ortho- and parahydrogen in an assembly is 3 : 1 at high temperature (e.g. 300 K). This equilibrium distribution is not fixed. As parahydrogen ($I = 0$) is the ground state a separation of hydrogen into ortho- and parahydrogen at low temperatures is possible. But this separation is not stable. In practice the amount of parahydrogen will be greater than 25 % at low temperature. As nuclear properties are different for ortho- and parahydrogen it is of main interest to know in what phase or in what mixture the hydrogen exist. The total cross sections are of the order of 20 different in the low energy region and even different by a factor of around 5 for 25 meV thermal neutrons. In calculations a 50 % mixture between ortho- and parahydrogen is assumed.

An additional, important effect is the molecular binding of the H-, D-molecules and their collective motion. It's absolutely necessary to take this into account using the model of Koppel, Young /1/ or/and Swaminathan, Tewari /2/. Otherwise errors in calculating the net slowing down effect of more than 20 % will occur /3/.

Hydrogen is inflammable between 4 % and 70 % and explosive between 18 % and 59 %. The critical points are $T_{krit} = 33{,}3$ K, $p_{krit} = 13{,}0$ bar. The density of liquid hydrogen (1 atm) is $\rho = 0{,}0708$ g/cm^3 and of supercritical (gas) hydrogen ρ (25 K) = 0,068 g/cm^3, ρ (30 K) = 0,059 g/cm^3 at 16 bar.

2.4 Principle design aspects

Real cold neutron sources have to take into account

- the size of the reactor core is limited. Therefore the CNS can be positioned only in a flux gradient and not always in a max. flux position

- the optimum flux position is strongly dependent from the choosen reflector material (H_2O, Be, D_2O). The max. of the thermal flux peaks in the reflector are of the order of ca. 2 (H_2O), 8 (Be), 10 (D_2O) cm distance from the reactor core. The distribution pattern is sharp (H_2O) and flat (D_2O, Be).
- the low temperature CNS must be surrounded by canning material for H_2, vacuum for cryogenic isolation and the beam tube canning material
- the optimum size of the CNS has to be determined for max. cold neutron flux and minimum cryogenic power. The CNS (H_2 and canning material) are heated by n- and γ-radiation. This heat has to be removed to secure low temperature H_2 at the source position.

In the following the above mentioned conditions are discussed in detail.

2.4.1 Diameter D

If neutron guides are used between the CNS and the experiment to avoid r^2 reduction of the neutron flux the diameter of the CNS should be choosen in that way: the CNS has to be transfered to the entrance cross section d of the neutron guide under the conditions of total reflection

$$L \cdot 2 \operatorname{tg} \gamma_c (\lambda) = D - d$$

L = distance between CNS and neutron guide = 130 cm
γ_c = critical angle of reflection = $0{,}099^\circ \cdot \lambda$ (Å) for Ni

		D = 13 cm	D = 15 cm
e.g.	λ = 5 Å	94 %	100 %
if d = 6 · 10,8 cm^2	λ = 8 Å	80 %	98 %
	λ = 10 Å	71 %	91 %

In addition the emissivity and radial dependence of emissivity of the CNS increases with increasing diameter. These three reasons force the tendency to a large source diameter. But the cryogenic cooling power P is ca. proportional to the volume of CNS ($\sim r^2$) and the price for a cryogenic plant is roughly $(P/Po)^{0,7}$. Therefore doubling the diameter will increase the cost by 160 %.

2.4.2 Thickness

Effectiveness of scattering is in a first order proportional to $N \cdot \sigma$ and $N = L/A \cdot F \cdot \rho \cdot d$.

As ρ is more or less identical for gas H_2 (T = 25 K) and liquid H_2 (T = 20 K), the optimal thickness must be the same for both types of CNS. The optimal size depends slightly from the reflector. In our case we calculated d = 4,2 cm.

2.4.3 Source materials

Common as source materials are pure Al or AlMg alloys with wall thickness of 1 mm to 6 mm Al depending on the demands on mechanical stability. The mechanical stability has to be discussed taking into account: overpressure accidents, low temperature behaviour, fast neutron embrittlement and production of Si through thermal neutrons: Al^{27} (n, γ) Si^{28}. Most of the CNS build worldwide using Al as canning material as Al has only a low absorption cross section. For the two CNS at Orphee /4/ thin stainless steel (25 % Ni) is used as canning material.

The selection of the source material and the thickness of this material has a great influence on safety and on the needed cryogenic power and therefore on investment and operation cost. In addition: the depression of neutron flux through the canning material should be considered, too. e.g. for 5,5 Å neutrons the flux depression is calculated for

12 mm Al to ca. 11 %
1,6 mm steel to ca. 20 %.

Therefore the decision for selecting the source material and the wall thickness of this material should be made very carefully.

2.4.4 Nuclear heating

Components of nuclear heating are: γ-heating in the canning material, γ-heating in H_2, fast neutron heating in H_2, capture γ's and β's in the canning material. These components have to be calculated and/or experimentally determined very carefully as they are strongly dependent from the special situation: distance of CNS from the core, moderator between CNS and core, reactor spectrum, power density e.g. An example for different distances and different reflectors between reactor core and CNS.

	4 cm Be (W)	2,5 cm H_2O (W)
γ-heating Al CNS	445	615
fast n-heating H_2	255	325
γ-heating H_2	60	80
γ-heating Al piping	65	90
capture γ + β Al	20	20
H_2-blower	80	80
heat losses in piping (2x20m)	200	200
	1.125 W	1.410 W

This example demonstrates that small design changes have great influence on the total amount of the needed cryogenic power. A surplus in cryogenic power will be better than a lack in cryogenic power.

2.5 Gain of cold neutrons

In a weakly absorbing media the energy distribution of the neutrons obey the Maxwellian distribution

$$\Phi\ (E,\ T) \sim \frac{E}{(KT)^2} \cdot \exp\ (-\ \frac{E}{KT})$$

Therefore in zero order the gain G (E)

$$G\ (E) = \frac{\Phi\ (E,\ 25\ K)}{\Phi\ (E,\ 300\ K)}$$

Under these conditions the max. theoretical gain is

$$E = 5\ \text{meV} \mathrel{\hat{=}} \lambda = 4\ \text{Å} \quad G = 16$$
$$E = 1\ \text{meV} \mathrel{\hat{=}} \lambda = 9\ \text{Å} \quad G = 93.$$

In fig. 3 this theoretical G (E) is compared with measured gains at five reactors. These results must be interpretated:

- DR-3 As the CNS (gas H_2) is installed in a tangential beam tube the measured gain will be infinite if the CNS is not filled with water in the case "without cold moderator". Otherwise nearly no neutrons will be measured.
- HFBR This source has a thickness of 7 cm which is by far too much. Therefore the design seems to be not optimal.
- FRJ-2 The reduced gain for higher wavelengths maybe a question of statistics or of optimization (source diameter to neutron guide cross section) for these wavelengths.
- Orphee Measurements were done at full power.
- Dido No optimal design.

The GKSS CNS will have at $\lambda = 5$ Å a gain $G \geq 11$.

2.6 Safety related aspects

The design basis accidents for a gaseous cold neutron source are

- instantaneous rupture of H_2- or He-buffer. Protective action: Selection of steel (1.4541) and increased wall thickness (1 mm per 1 bar which corresponds to power reactor rules). H_2-corrosion has to be considered.
- hydrogen/oxygen explosions. Protective action: triple containment philosophy in building (see fig. 4). Triple containment means hydrogen, Al (Nr. 1), vacuum, Al (Nr. 2), He, Al (Nr. 3). Pressure in hydrogen, vacuum and He are surveilled and combined with alarms, shutdown of cooling plant and reactor and etc. Therefore inside buildings no H_2/O_2-mixture can occur.
- loss of cryogenic power (no standby cooling, no shutdown of the reactor).

  The CNS will melt between 10' and 20' as the heat produced in the Al-canning cannot be removed. It can easily be shown that the melting will stop after having a contact between the molten Al and the beam tube.

- rupture of CNS
  Pressure increase up to 12 bars. Protective action: vacuum chamber withstands this pressure. He chamber is designed for 30 bar
- freezening of H_2 in heat exchanger and piping (no control or stop of cryogenic plant). Protective action: unisolated piping for heat removal through radiation to avoid blocking of standby cooling loop and thereby pressure increase in moderator chamber.

The design of a CNS can be made that way: all instrumentation may fail but nothing happens to the reactor or third persons. Therefore a CNS has to be treated only as a complex and expensive experiment.

3. Main design features of the FRG-1 cold neutron source

At the GKSS research centre Geesthacht in June 1984 the decision was made to sign a contract for the installation of a CNS with gaseous (supercritical) hydrogen as the cold moderator. The vendor is a consortium of the company Interatom and the Danish nuclear research centre at Risø. In the following the principles of the design of this CNS will be described (fig. 5, 6).

The CNS has an inner diameter of 155 mm and an effective inner thickness of 42 mm. These dimensions were choosen to have total reflection conditions even for 10 Å-neutrons for more than 93 % of the neutron guide cross section.

The operation conditions of the supercritical H_2 in the CNS are: 13,5 bar to 18 bars, 25 K to 35 K. The H_2 low temperature can be choosen by the operator due to experimental needs. To circulate the cold gaseous H_2 two (1 of 2) blowers are in operation. The heat input is removed in a H_2/He heat exchanger with He at 19 K/25 K temperature. The supply system for the cold He (compressor, cold box, heat exchanger, water coolers) is commercial available and needs no special development.

The H_2 system consists of

- 2 blowers for cold hydrogen (1 of 2)
- H_2/He heat exchanger
- 3 blowers for warm helium (2 of 3) for standby cooling
- relief box with relief valves
- H_2 buffer (6 m^3, 18 bar)
- H_2 house with pneumatic controlled valves for H_2 refilling procedure (1 of 2)
- chimney for H_2 release

- additional H_2 cooler (H_2O or Freon) for allowing standby operation without exceeding max. temperature at the CNS canning.

Standby operation: The cryogenic system is out of operation, the research reactor is on full power.

A vacuum at 10^{-6} mbars during operation of the CNS is necessary in many subsystems

- for temperature insulation and safety reasons in the inpile part, piping, joint box and relief box
- for temperature insulation in the cold box.

During normal operation only 1 blower in the joint box is in operation and the H_2 circulates from the inpile part to the joint box and back. The H_2 buffer volume of 6 m^3 works as a pressure reservoir.

If the cold He-circuit (cryogenerator) is out of operation the H_2 circuit consists of the inpile part (where the nuclear heat must be removed), piping, joint box (1 blower is in operation), piping, relief box, piping, H_2 house (2 blowers are in operation), Freon-cooler (1 of 2) and back.

In general it is necessary to demonstrate that for all operational modes of the CNS and of the reactor a safe operation of the CNS is possible. For having a high availability it should be impossible to have operational conditions which may damage the CNS e.g. operation of the reactor and no cooling of the CNS. Therefore we are looking at all combinations of operational conditions of the CNS and the reactor as there are:

CNS operation:

- no H_2 filling, no cryopower
- H_2 filling, standby cooling
- startup
- operation
- operation, H_2 circulation stopped

Reactor operation - shutdown
- startup
- full power operation.

An investigation shows that startup or operation of the reactor should not be possible in the cases no H_2 filling in the CNS or H_2 circulation stopped. These combinations may or will cause severe damage of the CNS if no protective actions take place.

4. Experience with the Risø CNS /5, 6/

The design of the GKSS cold neutron source is based on experience with the supercritical CNS installed in the Danish DR-3 reactor at Risø National Laboratory. The Risø CNS has been in operation since 1975. The Danish DR-3 reactor at Risø is a 10 MW heavy water cooled and moderated research reactor (Pluto type). The reactor is operated 23 days followed by a 5 days shutdown period.

The CNS plug with the moderator chamber is placed in a horizontal beam hole in the reactor. The thermal neutron flux at the moderator chamber is $5 \cdot 10^{13}$ n/cm^{2}s. The Risø CNS is cooled by two Philips-Stirling four cylinder two stage cryogenerators. The cryogenerators and the joint box are placed inside the reactor hall.

The gain factor at different hydrogen temperatures for the Risø CNS has been measured (fig. 3). It is seen that the gain factor is greater than 10 for energies below 5 meV.

It has been possible to operate the reactor independent of the CNS during the 10 years of operation. Occuring leaks in the containment or failures of the cryogenerators have only resulted in switch over to standby cooling. The overall availability over 10 years is greater than 95 %.

It is not possible to make maintenance on the CNS during reactor operation due to the necessity to cool the moderator chamber when the reactor is on power. All major repair and maintenance must then take place in the shutdown periods. Most of the manpower for maintenance has been used on the cryogenerators. Especially cleaning of the heat regenerators have been very time consuming, other maintenance jobs can be mentioned:

- replacement of ball bearings in the fast running hydrogen fans
- location and repair of leaks of H_2 and He into the vacuum system.

In order to cut down manpower and cost of spare parts in connection with maintenance, replacement of the cryogenerators with a new turbo cooling plant is considered.

5. Experiments with cold neutrons

Defects, voids and disorders in solids are in the range of a few Å to some hundred Å. Nuclear and magnetic (if ferromagnetic material is used) elastic small angle neutron scattering (SANS) is a powerful technique to investigate them using cold neutrons. GKSS intends to increase their SANS activities for investigations e.g. in the following fields

- precipitation structure in iron-copper alloys and pressure vessel steels as a function of irradiation (fluence)
- determination of hydrogen distribution in submarine welded alloys
- structural defects in amorphous alloys
- structure of polymers
- development of so called supermirrors.

Other examples can be found in /8/.

6. GKSS efforts in upgrading /7/

The operation of the cold neutron source will increase the cold neutron flux for

5 Å neutrons by a factor of 11
8 Å neutrons by a factor of 20
10 Å neutrons by a factor of 30.

To amplify these factors GKSS will undertake additional measures as follows

	factor
a) reduction in core size by a factor of ca. 2, reduction of the uranium enrichment from 93 % to 20 %, enlarge the U-235 content per fuel element from 180 g U-235 to more than 270 g U-235	> 1,5
b) Be metal reflector 32 cm thick around the beam tubes	1,45
c) 0,5 cm water gap around the beam tubes	1,15
d) renewal of beam shutter	1,20
e) renewal of neutron guides (better reflectivity)	?
f) increasing operation period (from 800 MWd/a to > 1.200 MWd/a)	1,5

All these measures will improve the conditions for the experiments with cold neutrons for

5 Å neutrons by a factor of 50
8 Å neutrons by a factor of 90
10 Å neutrons by a factor of 130.

This remarkable upgrading enables not only to shorten experimental time but it opens the possibilities for new experiments.

Table 1: cold neutron sources

Reactor	Location	Power (MW)	Reflector	Source	Cooling Power (W)	Positioning
			I. out of operation			
FR-2	Karlsruhe	44	D_2O	LH_2	50	horiz.
EL-3	Saclay	17	D_2O	LH_2	80	horiz.
			II. in operation			
DIDO	Harwell	27	D_2O	LH_2	40	horiz.
FRJ-2	Jülich	23	D_2O	LH_2	1000	horiz.
HFR	Grenoble	57	D_2O	LD_2	5000	vert.
Orphee	Saclay	14	D_2O	2 x LH_2	2 c 700	vert.
HFBR	Brookhaven	60	D_2O	LH_2	600	horiz.
DR-3	Risø	10	D_2O	GH_2	620	horiz.
FiR1	Helsinki	0,1	H_2O	LH_2	low	horiz.
KUR	Kyoto	low	C	LH_2	20	horiz.
HERALD	Aldermaston	5	H_2O	LH_2 + LD_2	200	horiz.
			III. under construction			
FRM	Munich	4	H_2O (Be)	LH_2	700	vert.
BER-2	Berlin	5 (10)	Be	GH_2	1900	horiz.
FRG-1	Geesthacht	5	Be	GH_2	1500	vert.
HFR	Grenoble	57	D_2O	LD_2		horiz.
Rutherford Appleton Lab.+		-		GH_2	580	horiz.
HWRR	Beijing	10	D_2O	LH_2		

+) Spallation source

Table 1: cold neutron sources

IV. projects

1. HFR-reactor	Petten, Netherlands
2. JRR-reactors	Tokai Mura, Japan
3. Pluto	Harwell, Great Britain (cancelled GH_2-project)
4. MPR-30-reactor	Djakarta, Indonesien

Table 2: neutron energies

Energy E (eV)	wavelength λ (Å)	neutrons
10	0,09	
1	0,29	epithermal
0,5	0,40	
10^{-1}	0,90	
$2{,}5 \cdot 10^{-2}$	1,81	thermal
10^{-2}	2,86	
$5 \cdot 10^{-3}$	4,04	
$2{,}5 \cdot 10^{-3}$	5,72	cold
10^{-3}	9,04	
10^{-5}	90,4	
10^{-6}	286	ultra cold

$$\lambda = \frac{0{,}286 \text{ Å}}{\sqrt{E\ (\text{eV})}} \qquad \text{Å} = 10^{-8} \text{ cm} = 0{,}1 \text{ nm}$$

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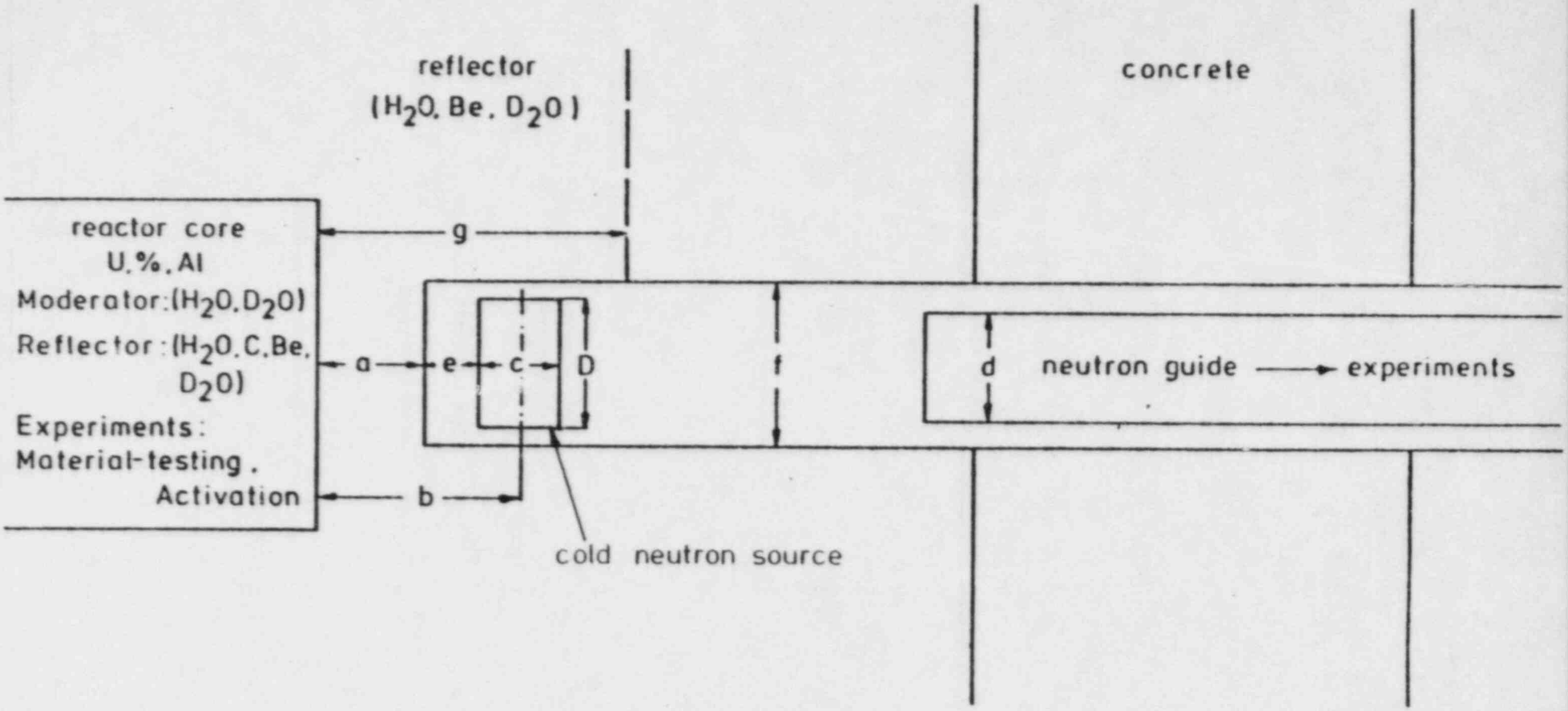
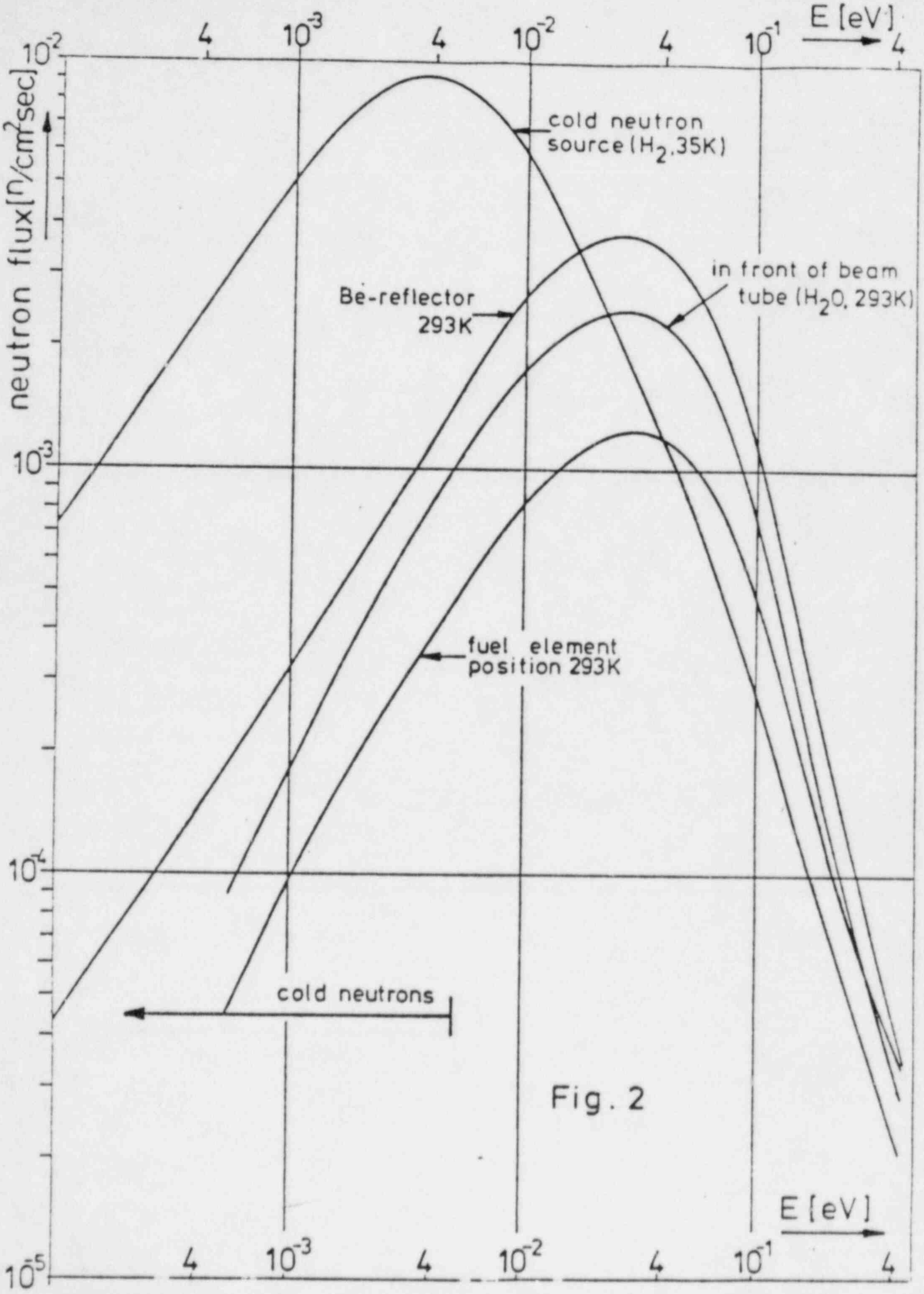
/8/ J. Stein: Experiments with cold neutrons, IAEA-Seminar, Tripolis, Sept. 1984



a = distance core - beam tube
b = distance core - CNS
c = thickness CNS
d = inner diameter neutron guide
D = inner diameter CNS
e = distance beam tube - CNS
f = inner diameter beam tube
g = thickness reflector

Fig. 1



E [eV]
neutron flux [n/cm²sec]
cold neutron source (H_2, 35K)
in front of beam tube (H_2O, 293K)
Be-reflector 293K
fuel element position 293K
cold neutrons
Fig. 2

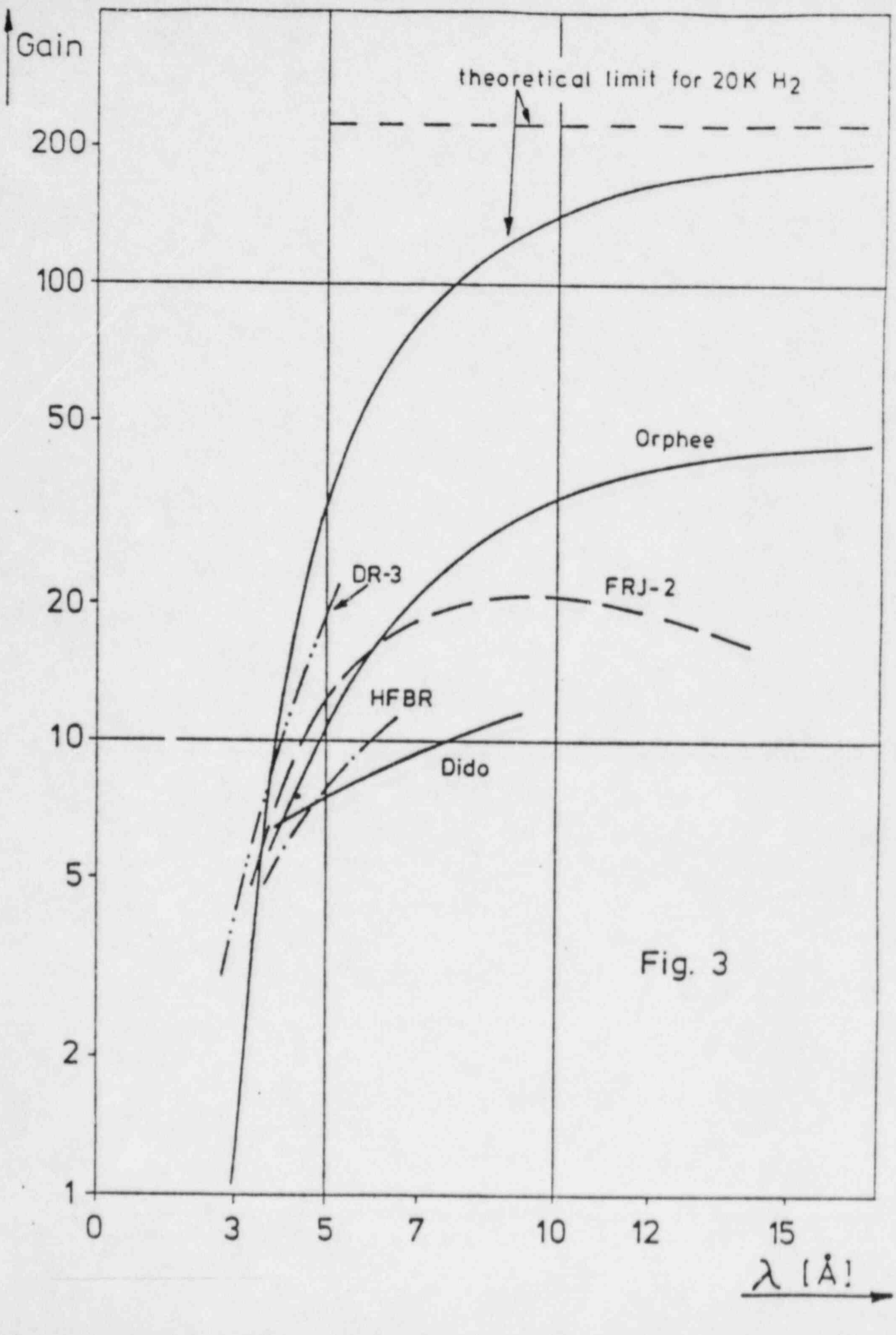
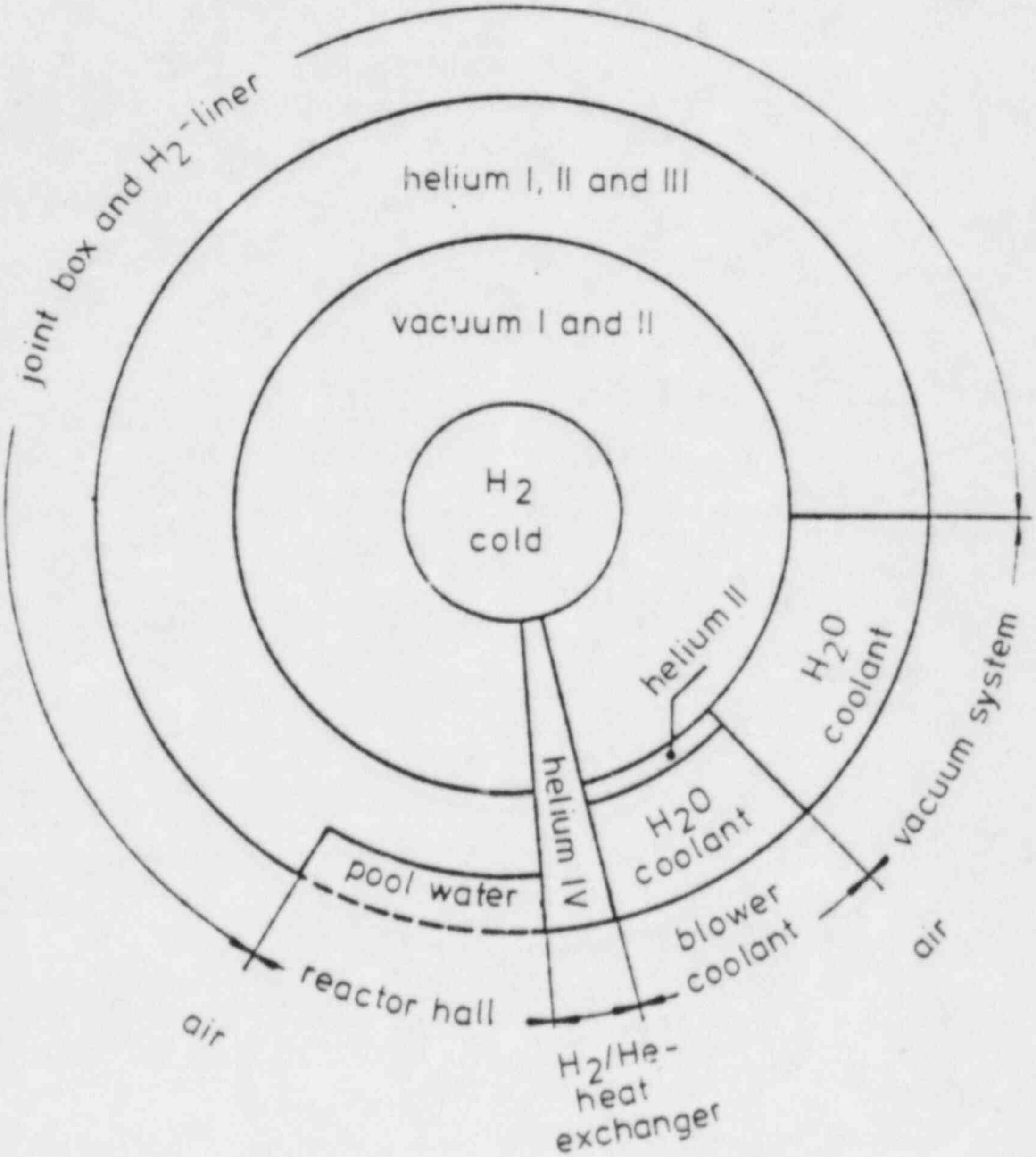

Gain
theoretical limit for 20K H_2
200
100
50
20
10
5
2
1
Orphee
DR-3
FRJ-2
HFBR
Dido
0
3
5
7
10
12
15
λ [Å]

Fig. 3



Triple containment CNS

Fig. 4

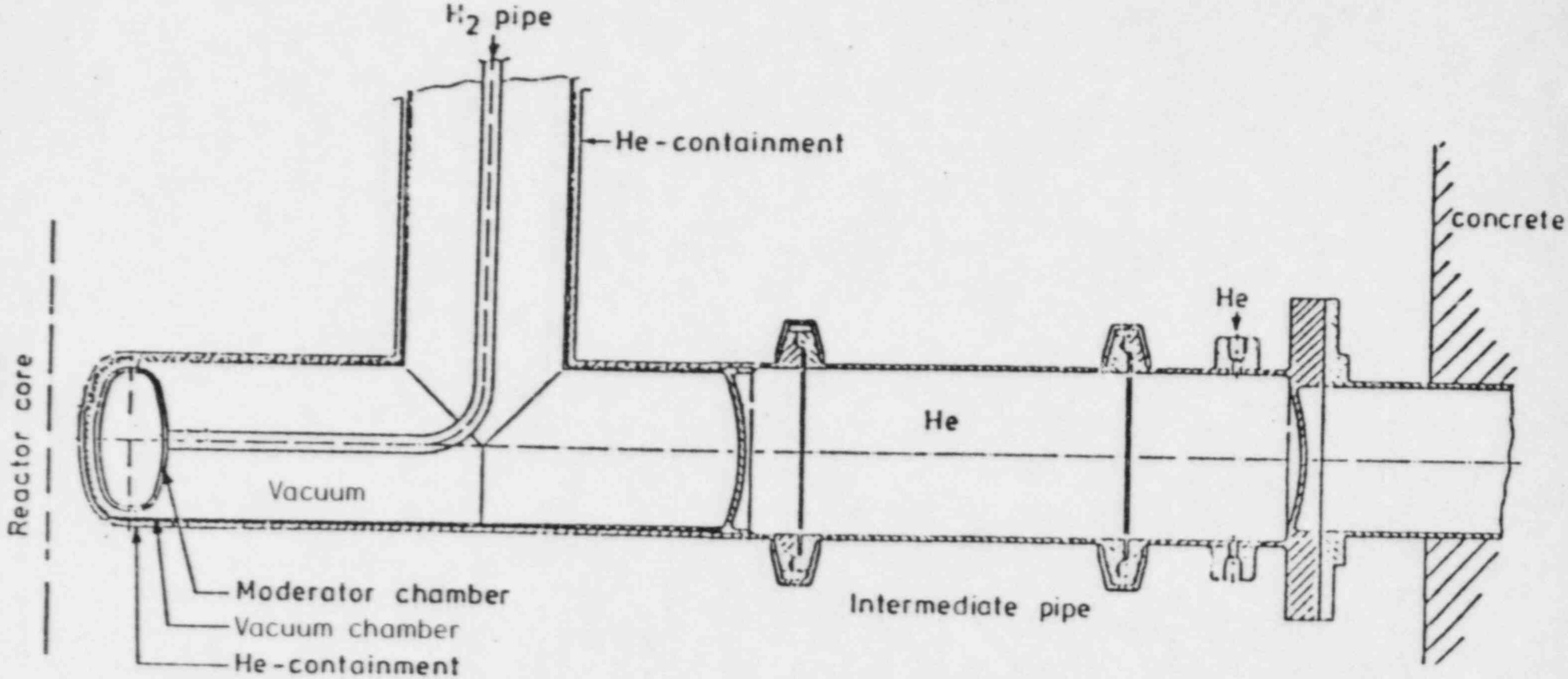

H_2 pipe
He - containment
concrete
He
He
Reactor core
Vacuum
Moderator chamber
Vacuum chamber
He - containment
Intermediate pipe

Fig. 5

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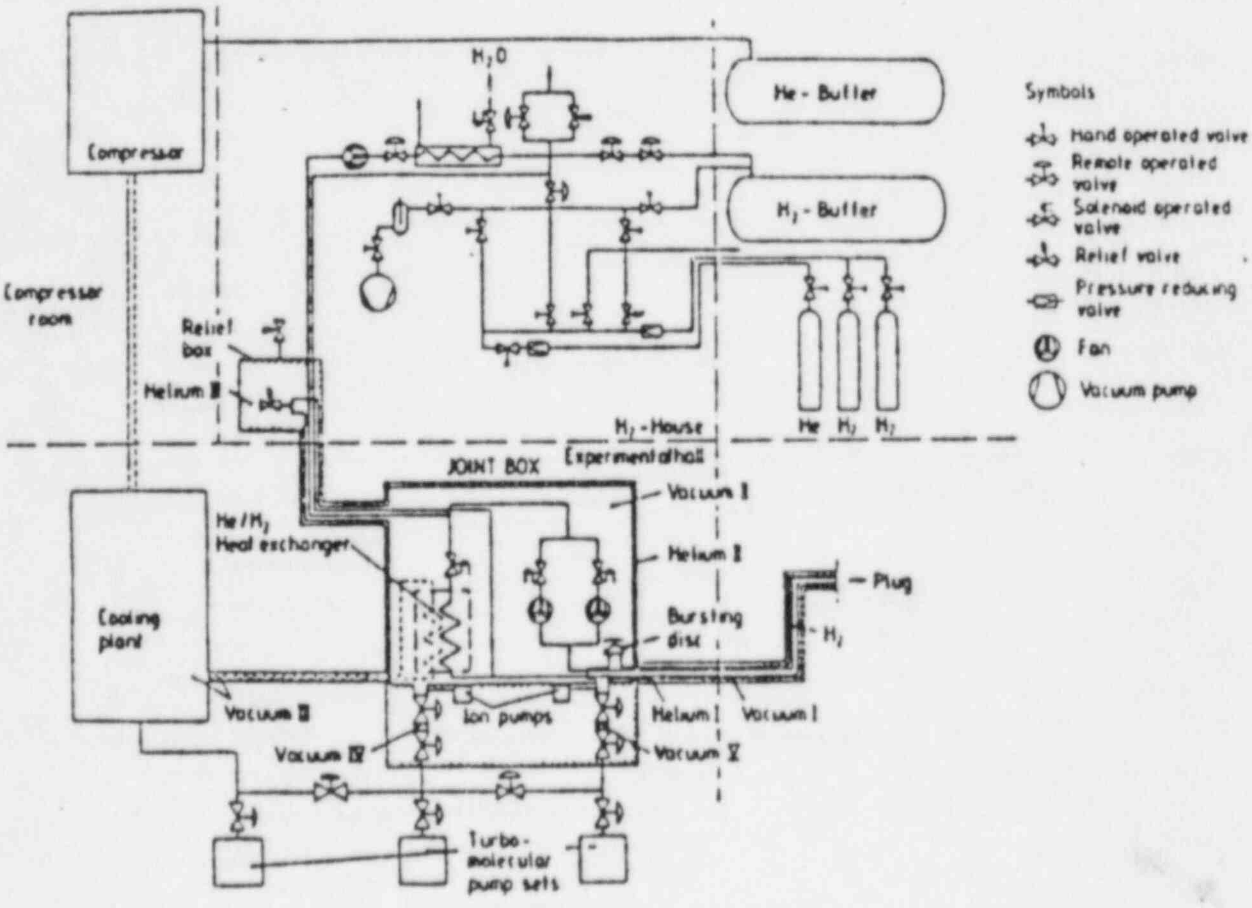

Fig. 6 principle diagram of FRG 1 cold neutron source