

# Neutrons for Research, Engineering and Medicine in Germany

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**Why neutrons?** Neutrons are essential, precious, and powerful. Their unique properties showing the structure and dynamics of materials have led to numerous advances and discoveries in basic materials science and made them an invaluable tool in industrial product development and manufacturing. They are vital to a number of scientific disciplines, including condensed matter, materials research and nuclear physics. In addition, they are essential for materials irradiation testing and the production of materials, especially radioisotopes for industry and medicine. Thus neutrons not only enable scientific advances, but also are crucial to the development of applied technologies, production of materials and nuclear medicine. [1]

Neutrons have a wavelike character, with wavelengths typically ranging from 0.01 to 100 nm. The way in which such particles are scattered elastically from materials reveals the spacing of the constituent atoms or the size of the molecules of which they are composed, ranging from crystalline materials through polymers and biological macromolecules, all the way up to strain-scanning of metals and alloys in engineering components. Neutrons can easily discriminate between isotopes of an element, are particularly effective at revealing the position of hydrogen-containing molecules such as water, or hydrogen atoms as part of the full structure of the biological molecules significant for pharmaceuticals or genomic research, as well as protons in hydrogen storage materials and fuel cells. A full understanding of the factors' controlling function in all of these systems frequently requires neutrons combined with other methods, and increasingly complemented by computer simulation. The latter also means that neutron scattering measurements provide an essential tool to benchmark computer simulations, with far-reaching consequences for many other fields of science.

The energy of such neutrons is typically of the order of meV, comparable to that associated with the motion of atoms and molecules in solids and liquids. Further, the fact that the neutron also possesses a small magnetic moment means that it can also probe the structure and excitations of electronic spins in magnetic materials – indeed, it is the most incisive tool of magnetism at an atomic scale. This is not only important in illuminating the search for new recording media, including the single-molecule magnets that may provide the qubits for future quantum computers, but also provides unique insights into the mechanism of high-temperature superconductivity.

Neutrons are electrically neutral, enabling them to penetrate deeply inside materials without disturbing them significantly. This enables the study of structure within dense materials, or held in complex apparatus for sample environment, facilitating studies under extreme or reactive conditions. This property also renders them non-destructive. Finally, it should be noted that the neutron itself provides an important subject for research, particularly when cooled down to very low temperatures or energies. The determination of some of its fundamental properties – for example its lifetime and the presence or lack of a very weak electric dipole moment – enables us to explore some of the most fundamental principles of physics, often in ways that complement the work performed at facilities for high-energy physics such as CERN. [2]

Last but not least, neutrons induce nuclear reactions. These enable to transform elements as it has been

performed in the doping of Si by transmutation of Si to P, producing the by far most homogeneously n-doped Si for industry. Nuclear medicine urgently needs radioisotopes for molecular imaging and therapy. For instance, the worldwide need for Technetium-99m of about 30 Million annual applications is almost entirely produced by 6 research reactors [3].

## How it began

After World War II nuclear technology was out of reach for the young Federal Republic. Things changed abruptly with the speech “**Atoms for Peace**” given by U.S. President Dwight D. Eisenhower to the UN General Assembly in New York City on December 8, 1953. In sight of the rise of the nuclear age and the development of the hydrogen bomb, the US were put under considerable pressure by the worldwide public to create an “Atoms for Peace” program that encouraged the peaceful use of nuclear energy and would help to avoid the destruction of humanity using this new technology. In this speech Eisenhower presented his ideas for peaceful uses: nuclear energy should be used for the production of energy e.g. electricity or heat; for applications in medicine and controlling epidemics and for helping to feed a growing population. All this should happen under the umbrella of an international atomic organization to ensure the safe and friendly usage of radioactive materials and technology.

As a follow-up the *International Conference on the Peaceful Uses of Atomic Energy* in Geneva, Switzerland took place in 1955. It brought together world leaders to discuss peace and aimed to reduce international tensions. It led to the foundation of the International Atomic Energy Association (IAEA) as a sub-organization of the UN to implement the “Atoms for Peace” program, opening up nuclear research to civilians and countries that had not previously possessed nuclear technology on July 29, 1957 in Vienna. This cleared the way for peaceful use of nuclear technology in both parts of Germany.

## Building the Atom-Egg

The Federal Republic of Germany (BRD) now had access to nuclear technology and nuclear materials for peaceful use. The so-called Atom-Ministry was founded, headed by Franz Josef Strauss. In 1953 Strauss became Federal Minister for Special Affairs in the second cabinet of *Chancellor Konrad Adenauer*.

The Federal Government and the states founded several nuclear labs like Gesellschaft für Kernforschung mbH and later on Kernforschungszentrum Karlsruhe GmbH (KfK), Kernforschungsanlage Jülich (KFA Jülich),



News from the German Agriculture Society (cover June 13, 1957) with the Atom-Egg under construction and a horse plough in the front symbolizing the raise of the new era.

Hahn-Meitner-Institut für Kernforschung (HMI), Gesellschaft für Kernenergieverwertung in Schiffbau und Schifffahrt at Geesthacht (GKSS), Physikalisch Technische Bundesanstalt at Braunschweig (PTB) and others, all intended to build research reactors for learning how to handle nuclear technology. Franz Josef Strauss did not forget his Bavarian homeland and agreed with the Prime Minister of Bavaria, Wilhelm Högnner (Social Democrats Party, SPD), to have the first nuclear reactor in Bavaria! Werner Heisenberg was also interested in these plans as he wanted to relocate the famous Max Planck Institute for Physics (originating from the Kaiser Wilhelm Institute) to Munich.

The first ideas were to locate this new Bavarian research reactor in Munich at Technische Hochschule München (TH-München) in the center of the city on Gabelsbergerstrasse. However, this idea was soon discarded and arable land in the north of Garching were considered. On June 6<sup>th</sup>, 1956 a part of the Bavarian Cabinet visited the selected site and had dinner at the local Gasthaus in Garching centre: Garching Neuwirt. Only 5 days later, on June 11<sup>th</sup>, Prime Minister Högnner gave the permission for the young Prof. Heinz Maier-Leibnitz to go to the US to buy a swimming pool reactor. He went immediately for a two-week trip and bought it while he was there. It took 2 months to obtain the detailed engineering design of the reactor. So building it started in November 1956. A year later, first criticality of the Forschungsreaktor München FRM was achieved on October 31<sup>st</sup>, 1957.

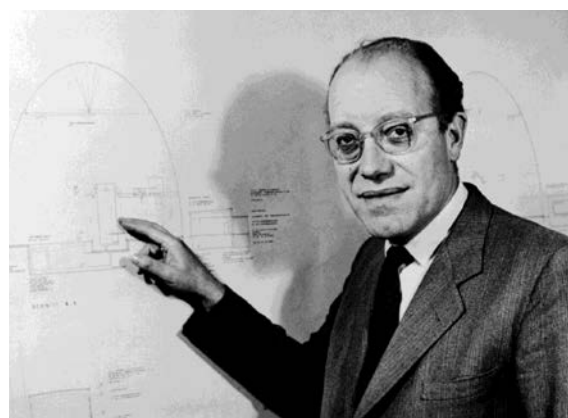
Garching is proud to be the home of the first nuclear reactor in Germany. Therefore, when the FRM was built, the local government proudly incorporated the Atom-Egg into their coat of arms. Thanks to this decision, Garching today is world-renowned for the Research Campus of the Technische Universität München, of which the Atom-Egg was like a seed. [4]

What happened in West Germany was mirrored in the East. The Soviet Union, within the Atoms for Peace program, delivered research reactors to almost all of the republics/states of the Union and countries of the Soviet Economy Zone. Only 6 weeks after the Atom-Egg reached criticality the Zentralinstitut für Kernforschung of the GDR at Rossendorf near Dresden started the operation of the Rossendorf research reactor on December 16, 1957. This neutron source already had a heavy water moderator and originally produced 2 MW<sub>th</sub>. It was later upgraded to 5 MW<sub>th</sub> and then to 10 MW<sub>th</sub>. Just before the fall of the Iron Curtain a refitting programm of the reactor was started, but the final decision to decommission it was taken after reunification. On June 27, 1991 the nuclear operation came to an end.

### Heinz Maier-Leibnitz

Heinz Maier-Leibnitz was a Professor at THM (renamed 1970 in Technische Universität München, TUM) from 1952 to 1979. From the beginning, he saw this research reactor as an instrument to do science rather than contributing to the development of nuclear technologies. Heinz's motto

was "Do something new" or more precisely "Do it differently to the Americans" who at that time were much further ahead in terms of using neutrons for science, which was later on honored by attributing the physics Nobel prize to Bertram Brockhouse and Clifford Shull in 1994. Maier-Leibnitz had also an impressive number of PhD students. Many of them became engaged at the research reactors of the upcoming National Labs like KFA Jülich. The two research reactors MERLIN and DIDO went into operation with about an order of magnitude higher neutron flux compared to the Atom-Egg. Following the motto of Maier-Leibnitz, the Atom-Egg but also the Jülich reactors became literally the nuclei of innovative instrumentation and science with neutrons that spread all over Europe and the world in the coming decades.



Prof. Heinz Maier-Leibnitz explains the construction of the Atom-Egg (Copyright TUM).

Certainly, the discovery with the greatest impact on science with neutrons was the invention of neutron guides, almost happening accidentally during the trial of Tasso Springer, at that time PhD student of Maier-Leibnitz, while protecting an emergent neutron beam by a metal tube in order to prevent people to run through the beam. Tasso Springer realized that with the tube protecting the beam he counted more neutrons at his detector. Other important milestones for science with neutrons of Maier-Leibnitz and his students where precise measurements of cross sections of neutrons with matter, high precision nuclear spectroscopy, the first operating time-of-flight diffractometer with a 150 m long neutron guide, high-resolution spectroscopy by neutron back scattering, the Steyerl turbine for shifting thermal neutrons in the wavelength range of very cold neutrons with neutron wavelength similar to visible light, the first small-angle scattering camera for detecting objects on the scale of nanometer to micrometer, irradiation of matter at very low temperature (4.6 K) to study the influence of radiation on matter and last but not least hadron cancer therapy by fast neutrons. [4]

### From Garching to Europe

There was a second political impact which fostered science with neutrons in Europe. On January 22, 1963 Charles de Gaulle and Konrad Adenauer signed the Élysée Treaty. This ambitious treaty intended to create a Franco-German cultural identity. As a kind of show case a large Franco-German research institution should be founded. After an initially very small progress, Heinz Maier-Leibnitz, Louis Néel, Jules Horowitz, Robert Dautray, Félix Bertaut, and others took the initiative for a high-flux neutron source, intended to outstand all other neutron sources. Furthermore, they wanted to compete for dominance with the

USA in the field of neutron research. Again, science was serving diplomacy. [5, 6]

On January 19, 1967 the Science Ministers Gerhard Stoltenberg and Alain Peyrefitte signed the contract for the Institute Laue-Langevin (ILL) situated at Grenoble in the French Rhone-Alpes region given the task to build and operate a high flux neutron source (HFR). Heinz Maier-Leibnitz became the founding director. On November 6, 1970 the UK joined the ILL contract thanks to its science secretary Margaret Thatcher. First criticality was achieved in 1972, the ILL became a tri-national neutron source. The single element compact core with about 8 kg 93% enriched Uranium is heavy-water cooled and moderated at a nominal thermal power of 58 MW, yielding an unperturbed thermal neutron flux in the moderator of  $\phi_{th} = 1.3 \times 10^{15} \text{ ncm}^{-2}\text{s}^{-1}$ . Some 10 years before the High Flux Irradiation Reactor (HFIR) at Brookhaven National Lab was the first neutron source with such a compact core and an even slightly higher thermal flux. However, light water cooling and moderation mainly trap this high flux in the core, so being optimized for the production of isotopes by neutron-capture, but less suited for extracting thermal neutron beams. The high thermal flux of the ILL reactor some 12 cm outside the core in the D<sub>2</sub>O moderator was the technical precondition for further pioneering techniques of this source. Auxiliary moderators like the D<sub>2</sub> cold source operated at liquid hydrogen temperature (25 K) (today two cold sources) and the hot source consisting of a block of graphite heated to about 2000 °C, were placed in the heavy water reflector, near to the position of maximum thermal flux, thereby shifting the thermal wavelength of  $\sim 0.12 \text{ nm}$  to longer wavelength  $\sim 6 \text{ nm}$  and shorter wavelength  $\sim 0.05 \text{ nm}$ . A circular primary shielding allows the extraction of the neutrons by in total 16 beam ports facing the different moderators or wavelength shifter. These neutrons are almost loss-free transported by cold and thermal neutron guides to today more than 45 experimental stations, mostly situated in the meanwhile erected two neutron guide halls. [5, 7]

Many of the first-generation instruments followed the handwriting of Maier-Leibnitz and his students, like neutron guides, small angle scattering to characterize objects on the meso-scale, high-resolution spectroscopy by backscattering, high-precision nuclear spectroscopy, intense ultracold neutron source, etc. Heinz-Maier-Leibnitz's follower as director of the ILL was his former student Rudolf Mößbauer (1972 to 1977), at that time already Nobel prize winner for the experimental realization of nuclear resonances, the so-called Mößbauer effect. He was the director who made the decisive step from building a high brilliance neutron source to create the world-leading place in science with neutrons. Again different to what happened beyond the Atlantic, Mößbauer

insisted on the service character of the ILL, the brilliant neutrons source should serve the best scientists from the three partner countries, coming from national labs and the universities – and as another step further beam time should be allocated solely based on the scientific merit of the measuring proposal [6]. Competition is the key to scientific excellence and the ILL counts today apart from the three founding nations additional 10 Scientific Member countries from Europe. With its today more than 45 instruments it has become the world leading center in science with neutrons.

With the success of ILL, many of the German neutron scientists shifted their main activities to Grenoble – on the detriment of the neutron sources at home, which had a one or two orders of magnitude smaller flux. Research reactors with beam ports like those at Karlsruhe, Braunschweig, MERLIN at Jülich and others were shut down. However, the remaining ones – the TRIGA reactor at University Mainz, the FRM, DIDO at Jülich, BER-II at HMI, FRG-1 at Geesthacht – were more and more needed to prepare the scientific case for proposals at ILL, in order to be competitive in the hunt for beam time at ILL.

### Up-grade of the BER-II reactor at the Hahn Meitner Institute

Prof. Hans Dachs from the Hahn-Meitner-Institute (HMI) was one of the first to realize that Germany would need a national source with neutron fluxes at least approaching those of the ILL in order to strengthen the German neutron science community for the competition for beam time at ILL. Already in 1982 the HMI launched the plan to upgrade its 2<sup>nd</sup> research reactor BER II (first criticality in 1970) from 5 to 10 MW<sub>th</sub>. The upgrade measures comprised the installation of a Beryllium reflector surrounding the core and most importantly the installation of a liquid H<sub>2</sub> cold neutron source which were to feed a suite of new scattering instruments in a new experimental hall. However, the Berlin colleagues were also the first to experience the changing perception of nuclear technology by society. In early 1989, just before the final operation license for the rebuilt neutron source was expected, the Berlin Senate changed because of elections to Red-Green (SPD + AL<sup>1</sup>). The new AL Senator for Environment Michaela Schreyer refused the licensing, very much in dissent with the coalition partner SPD. This and further open conflicts led to the end of the Red-Green coalition in autumn 1990. After the next election the Berlin Senate changed to Black-Red (CDU + SPD) and on March 26, 1991 the operation license of the upgraded BER II was finally granted. [8]

Prof. Ferenc Mezei, who joined the HMI in 1984 and is a great expert in neutron optics and instrumentation was responsible to add a suite of cold-neutron instruments all of which had been designed to allow for polarized-neutron capabilities. He came up with the idea of so-called multi-spectral beam extraction which was later realized at BER II to deliver thermal and cold neutrons in one and the same guide. So, Germany had in the beginning of the 90<sup>th</sup> a brand-new neutron source with an unperturbed flux in the Be-reflector of  $1.5 \times 10^{14} \text{ n/cm}^2$  with a clear emphasis on cold neutrons and a unique broad spectrum from thermal to cold neutrons at one of its altogether 24 instruments [9]. The conversion of BER II to operate with low enriched uranium was completed in the years 1998 to 2000 without reduction in neutron flux.

The instrument FLEXX aiming on resolving magnetic and structural excitations with up to now unprecedented



View on reactor, administrative building and neutron guide hall of the ILL at Grenoble (Copyright ILL).

<sup>1</sup> "Alternative Liste", which later merged into the party "Die Grünen"



resolution is one example of the innovative and highly competitive instrumentation at BER II [9a]. The success of the instrument also builds on the renowned sample environment capabilities developed for the neutron instrumentation at the BER II reactor, a hallmark of research with neutrons in Berlin. Exemplarily, fingerprints of a novel phase, a quantum spin liquid, have been detected in the compound  $\text{YbMgGaO}_4$  by experiments on FLEXX [10]. The underlying triangular lattice structure in this compound favors the emergence of liquid-like magnetic excitations named spinons. Knowledge about these excitations might help to better understand high-temperature super conductivity and is highly relevant for quantum information technology. Interestingly, a part of the experiments have been performed at the ILL, showing the competitiveness of the new instrumentation at BER II.



| The spectrometer FLEXX at BER II of HZB for measurements of structural and magnetic excitations in matter. This instrument combines a classical triple-axis spectrometer with neutron spin-echo technique, which boosts its resolution by more than one order of magnitude [10a]. Spin-echo techniques are also extensively used in nuclear magnetic resonance (Copyright Klaus Habicht, HZB).

Certainly, the most outstanding facility at BER II is its High Field Magnet (HFM) which delivers static magnetic fields up to 26 Tesla, the highest static magnetic fields available for neutron scattering. This special sample environment together with a versatile instrument which unites several important neutron scattering methods, namely diffraction, small-angle scattering and time-of-flight spectroscopy, provides capabilities to measure structure and dynamics on atomic scale and temperatures as low as 0.65 K. Among the first experiments at HFM scientists at HZB have studied the magnetic structure in a Rh-doped variant of a magnetic material,  $\text{URu}_2\text{Si}_2$  with hidden order of unknown origin, further contributing to solve a long-standing puzzle. The high magnetic field changes the energy of electrons in the material and leads to long-range magnetic order [11].

In July 2013 the Supervisory Board of HZB (formerly HMI) decided to concentrate on research with synchrotron radiation – BESSY II and new synchrotron projects – and to stop operation of BER II. So, in the end of this year BER II will cease its service, despite its highly competitive instrumentation.

### ELLA, a new guide hall for cold neutrons at DIDO

When Prof. Tasso Springer returned to Forschungszentrum Jülich from his ILL directorate beginning 1983, he faced a similar situation for DIDO like the Berlin colleagues. DIDO, with a first criticality in 1962, already built with a  $\text{D}_2\text{O}$  moderator and meanwhile upgraded to 23  $\text{MW}_{\text{th}}$ , in 1986 was upgraded by a new cold source and a completely



| View into the ELLA hall at DIDO with its suite of small-angle cameras (Copyright JCNS, FZ Jülich).

newly built neutron guide hall called ELLA<sup>2</sup>, equipped with  $^{58}\text{Ni}$  neutron guides. DIDO delivered an unperturbed thermal neutron flux of  $3 \times 10^{14} \text{ ncm}^{-2}\text{s}^{-1}$ , i.e. at that time the most intense neutron beams in Germany.

A perfect example of the newly achieved competitiveness became the suite of small angle cameras in the ELLA guide hall, covering structures from 1 nm to beyond 1  $\mu\text{m}$ . A well-known problem at winter times is the filter blockage of diesel fuel. Fuel oils contain alkanes that precipitate at low temperature as large crystals of wax are plugging filters. By means of the small angle cameras at ELLA self-assembling additives of crystalline-amorphous diblock copolymers have been characterized which combat this behavior by decreasing the size and altering the shape of the wax crystals; e.g., smaller sized crystals are less likely to clog the filters. [12]

DIDO with its relatively high thermal flux was equipped with several irradiation thimbles. Since 2004 one of those was used to produce the fission isotope  $^{99}\text{Mo}$  by irradiating  $^{235}\text{U}$  targets.  $^{99}\text{Mo}$  decays within 60 h to  $^{99\text{m}}\text{Tc}$  which is the most used radioisotope for molecular imaging in nuclear medicine – only in Germany 3 million applications are needed per year. So, for the first time Germany could contribute to the world-wide supply chain of this important radioisotope.

DIDO stopped operation in 2006, but we will come back to its revival later on.

### TRIGA Mainz

As a follow-up of the Atoms for Peace program a group around E. Teller in the US developed in the late 1950ies a research reactor type of low thermal power that is inherently safe, the so-called TRIGA reactor fueled with  $\text{UZrH}$  fuel. Here the passive inherent safety derives from the large negative prompt temperature coefficient of this particular fuel. In 1967 such a TRIGA reactor started operation at the University of Mainz. Typically, this kind of reactor can be used in a pulsed or continuous mode. TRIGA Mainz delivers in its continuous mode at a power of 100  $\text{kW}_{\text{th}}$  a thermal flux of some  $10^{11} \text{ ncm}^{-2}\text{s}^{-1}$  at its beam ports and at its irradiation thimbles a maximum of  $2 \times 10^{12} \text{ ncm}^{-2}\text{s}^{-1}$ . However, in its pulsed mode the peak power reaches up to 250  $\text{MW}_{\text{th}}$ , resulting in a neutron fluence in the order of  $10^{15} \text{ cm}^{-2}$  in one pulse (fwhm = 30 ms). Pulses might be repeated every 12 to 15 minutes. TRIGA Mainz stands out by the combination of low averaged thermal power, its compact design, accessibility of the beam ports and its high intensity in a pulse. This

<sup>2)</sup>externes  
Neutronenleiter  
Labor

makes it ideally suited for basic research in nuclear physics and radiochemistry and less suited for neutron scattering. So radiochemistry of short-living fission products with half-lives down to 1 s are a specialty of the research at the TRIGA reactor, also in collaboration with the GSI at Darmstadt. [13]

The most visible flagships of research at the TRIGA Mainz are its two UCN sources. A thermal beam gets pre-moderated by a solid H<sub>2</sub> moderator. These cold neutrons then hit a solid D<sub>2</sub> crystal at liquid He temperature which acts as a so-called super-thermal column, where the cold neutrons further lose energy through inelastic scattering processes and are accumulated with kinetic energies corresponding to mK temperatures, i.e. they are ultra-cold. Here the high intensity in the pulse and the slow repetition rate of 12 to 15 minute i.e. the small averaged heat deposition at the solid D<sub>2</sub> (~5 K) crystal perfectly complement each other. Not a high UCN flux is the ultimate goal, but a high UCN density stored in a given volume is wanted. Today, the two UCN sources at TRIGA Mainz deliver a UCN density of ~2 UCN/cm<sup>3</sup> which compares very favorably with the UCN densities achieved at ILL or PSI (Switzerland) in the range of 20 UCN/cm<sup>3</sup> [14].

Due to their low kinetic energy UCNs can be stored either in magnetic gradients or bottles with almost perfect surfaces. This allows to measure precisely the neutron lifetime ~880 s and correlations between the products of its  $\beta$ -decay. These parameters are compared to the prediction of standard theory for the unification of three fundamental forces and eventually may be indications for a theory beyond the standard model.

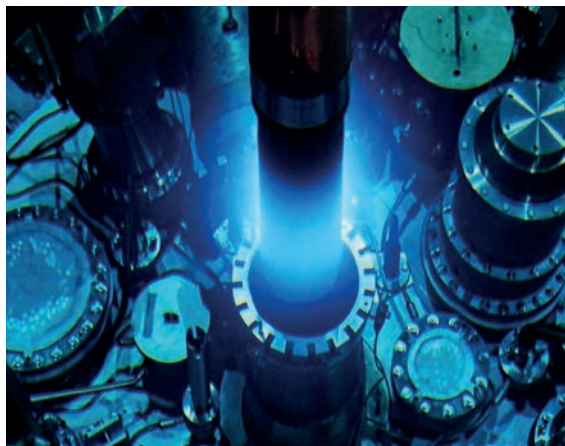
#### A dream – a spallation source for Germany

With the high intensity research reactors like HFIR, HFR or FRM II (see below) with continuous neutron fluxes in the range of  $10^{15}$  ncm<sup>-2</sup>s<sup>-1</sup>, the technical limit of safety and feasibility has been reached, simply because the heat load at the fuel plate surfaces reaches values > 400 W/cm<sup>2</sup> or the volumetric heat load exceeds locally 2 MW/ltr. On average 2.3 neutrons are freed per fission event. Hitting a heavy target like W or Ta by a proton beam with energies exceeding 1 GeV frees 10 times more neutrons per event. So, why not build a spallation source which has the potential to deliver 10 to 100 times more neutron flux in the pulse than the best continuous neutron sources, but of course in a fraction of time. In Germany this idea was picked up by the colleagues at FZ Jülich and the project of the German Spallationsneutronenquelle SNQ to be built at Jülich was launched. After detailed project engineering it turned out (1995) that this ambitious project was too expensive for Germany in those times. Instead, the idea of a new German medium-flux reactor, cheaper and hopefully faster to realize was launched.

The work put in the project SNQ was not wasted. The idea of a powerful spallation source, now as an European flagship facility persisted and FZ Jülich engaged again in the topic, but now as an European Spallation Source with Germany as host country. But in competition with two other large-scale projects, namely X-FEL (free electron laser for x-rays) and FAIR (heavy ion accelerator), both to be hosted at Germany, too, this 2<sup>nd</sup> attempt was doomed to failure in 2002. However, the idea of an ESS survived, Sweden in partnership with Denmark has been selected as host country, construction has started in 2014, and user operation is expected to begin after 2024. And Germany (FZ Jülich, HZG, TUM) is heavily engaged in building innovative instrumentation for the ESS.

#### The Heinz Maier-Leibnitz Neutron Source (FRM II)

In the mid-1980s the idea was mooted by Prof. Wolfgang Gläser at TUM to significantly increase the FRM's capacity. Finally, the concept of a new building emerged as the most secure technical solution. The whole German neutron community supported this endeavor, arguing after the stop of the SNQ project that Germany needs a national neutron source with a flux approaching that of ILL, to be realized in realistic time scales and last but not least economic. It was one of the first tasks of the newly founded (1987) national Komitee Forschung mit Neutronen (KFN) to push forward these arguments at the political level [15]. The groundbreaking ceremony for the new building took place on August 1, 1996. The research neutron source Heinz Maier-Leibnitz (FRM II) reached its first criticality on March 2, 2004 and user operation began only one year later on April 29, 2005 with an initial suite of 15 instruments. The Atom-Egg, however, ceased operating on July 28, 2000, as the staff there was needed to start operation at FRM II.



After 60 days of operation the compact fuel element of FRM II is exhausted. The fuel element is taken out of the central cooling channel (Copyright Bernhard Ludewig 2012).

The heart of FRM II is its compact core consisting of an assembly of involute shaped fuel plates with a diameter of 24 cm in the active zone. Its concept largely profited from the experience gained with the two preceding high flux research reactors with a compact single fuel element – HFIR at Oak Ridge (1965) and HFR at Grenoble (1972). At FRM II cooling is achieved by light water, whereas moderation and neutron reflection happen in an outer D<sub>2</sub>O moderator. Thus the Tritium contaminated moderator can be operated in a closed cycle and the risk of emission of Tritium is drastically reduced. For the first time metallic Uranium densities of 3 gU/cm<sup>3</sup> in the meat of the fuel plates were used allowing a further decrease of the core diameter. As a result, FRM II with a moderate power of 20 MW<sub>th</sub> has a maximal unperturbed thermal neutron flux of  $8 \times 10^{14}$  ncm<sup>-2</sup>s<sup>-1</sup> some 12 cm away from the outer radius of the core in the heavy water moderator. The moderate power of 20 MW<sub>th</sub> allows to place a D<sub>2</sub> cold source (25 K) and a hot source (graphite 2000 °C) at the maximum of thermal flux. A through-going beam tube will house a solid D<sub>2</sub> UCN source (currently under construction) similar to those installed at TRIGA Mainz, but now with a continuous UCN flux. Further a so-called converter facility at the outer edge of the moderator converts thermal neutrons to an intense beam of fission neutrons ( $2.3 \times 10^8$  n<sub>f</sub>cm<sup>-2</sup>s<sup>-1</sup> in a narrow energy range around 2 MeV. Last but not least,



Reactor	Location	Operating since	Nominal therm. Power [MW]	Cycle length [days]	HEU load [kg]	Uranium density [g U/cm <sup>3</sup> ]	Unperturbed therm. flux [ncm <sup>-2</sup> s <sup>-1</sup> ]	Moderator	Cooling	Wavelength shifter
HFIR	Oakridge, USA	1965	80	23		1.1	$1.3 \times 10^{15}$	H <sub>2</sub> O	H <sub>2</sub> O	1 cold source
HFR ILL	Grenoble, France	1972	58	44	8 - 10	1.1	$1.3 \times 10^{15}$	D <sub>2</sub> O	D <sub>2</sub> O	2 cold sources 1 hot source
FRM II	Munich, Germany	2004	20	60		3	$0.8 \times 10^{15}$	D <sub>2</sub> O	H <sub>2</sub> O	1 cold source 1 hot source 1 UCN source <sup>3</sup> 1 fission source 1 positron source

Tab. 1.

Comparison of the three compact-core high intensity neutron sources.

the neutrons in the moderator play Einstein ( $E = m \times c^2$ ), i.e. the neutron induced high  $\gamma$ -intensity in the moderator spontaneously transforms to matter and anti-matter in form of electrons and positrons. The positrons are extracted electromagnetically through a beam tube. With a thermal positron flux of  $1.2 \times 10^9$  p<sub>th</sub>/s this is the most intense source of mono-energetic antimatter in the form of positrons. In total the reactor core is surrounded by 11 beam ports, 3 of them facing the cold source. Furthermore, several thimbles introduced vertically from the top into the moderator serve for irradiation purposes like isotope production, silicon doping or neutron activation analysis. FRM II stands out by the highest flux to thermal power ratio, the broadest spectrum of wavelength shifter as there are UCN (under construction), cold, thermal, hot, fission neutrons and positrons – see Table 1 and [16].

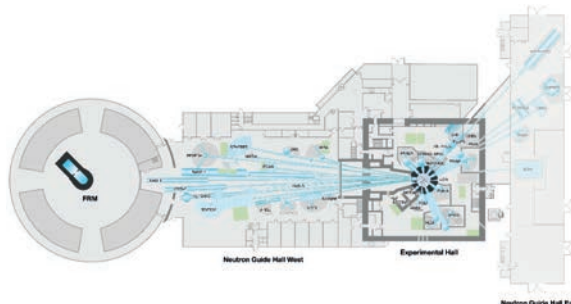
### MLZ – A national center for research with neutrons

With the first criticality of FRM II in 2004 the Council of FZ Jülich decided to close DIDO in 2006 and to transfer 6 of its best instruments to FRM II. In parallel with this decision, the neutron scattering activities within FZ Jülich were concentrated in a new institute, the Jülich Centre for Neutron Scattering JCNS. This was more than changing a label because FZ Jülich changed from an operator of nuclear reactors to an operator of neutron instruments at the world's most intense neutron sources, the ILL, FRM II and also one instrument abroad at the American Spallation Neutron Source at Oak Ridge. JCNS operates all these instruments for the German and international user



Heinz Maier-Leibnitz Zentrum (MLZ), a cooperation between FZ Jülich, HZG, TUM, MPI and further 10 university groups for the scientific use of the FRM II (Copyright: Schürmann/ TUM).

<sup>3</sup>) Under construction



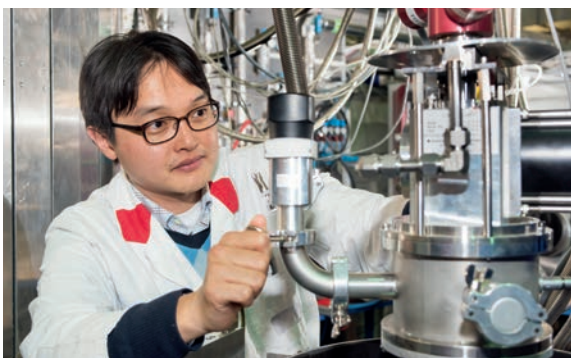
Layout of the beam transport and instrumentation at FRM II. From left to right: the Atom-Egg, which might serve as extension of the neutron guide hall west after disposal of the FRM reactor block, the neutron guide hall west fed with cold neutrons, the experimental hall around the reactor block and guide hall east (instruments under construction) (Copyright Ramona Buchner JCNS@MLZ).

community. With the transfer of the first instruments to Garching the construction of a second guide hall including offices and laboratories for the extension was realized.

The very successful merger of the neutron activities of TUM and FZ Jülich encouraged to go a step further. The research reactor FRG- 1 at Helmholtz Zentrum Geesthacht HZG (formerly GKSS) was switched off in 2010. HZG followed the Jülich model with the newly established “German Engineering Materials Science Center” (GEMS) and transferred their neutron scattering activities to FRM II. The close collaboration between the TUM, FZ Jülich and HZG at FRM II in Garching led to the establishment of a cooperation between the three institutions under the name Heinz Maier-Leibnitz Zentrum (MLZ), with the aim of getting scientific use of the FRM II on January 1, 2011. With the creation of the MLZ the Federal Republic and Bavaria also agreed on sharing the costs for providing the neutrons. Germany has now concentrated its knowledge in neutron applications around the MLZ. Today MLZ operates 28 instruments and further 6 instruments are under construction to serve about 1000 German and international researchers per year with brilliant neutron and positron beams [16]. MLZ has become one of the world leading centers in science with neutrons, with additional services of FRM II to industry and medicine.

### We have a dream

“We have a dream!” is the common motivation of physicists all over the world searching for superconductivity at room temperature. In 1986 this dream got an enormous up draught when Georg Bednorz and Alexander Müller (Physics Nobel prize 1987) discovered oxides which led to materials which are superconducting at high temperatures like 140 K. Euphemistically these ceramics are called “high”-temperature superconductors (high-T<sub>c</sub>). But, 30

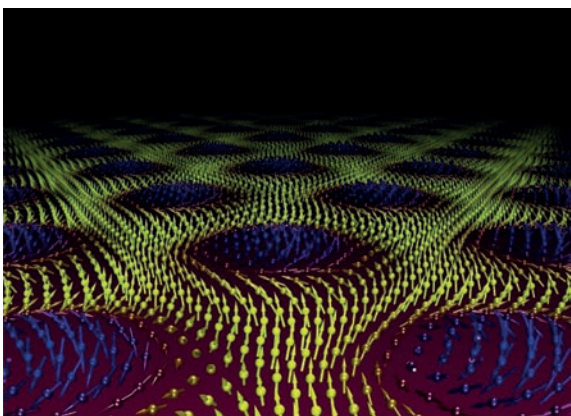


Jitae Park and Astrid Schneidewind work at MLZ on explaining the mechanism of high-T<sub>c</sub> (Copyright: Wolfgang Filser/ TUM).



years later the dream of superconductivity at room temperature is not yet reality – because its mechanism has not yet been fully understood. For conventional superconductivity at liquid He temperatures the Bardeen-Cooper-Schrieffer BCS theory explains very well the formation of electrons to Cooper-pairs mediated by lattice vibrations (phonons), which are at the origin of non-dissipative electric currents. That Cooper pairs are also important in high-T<sub>c</sub> is meanwhile common sense. To the surprise of many scientists there is strong experimental evidence that collective excitations of the electronic magnetic moments are responsible for this coupling mechanism. And that is where neutron scattering takes part. The neutron spin (magnetic moment) is ideally suited to detect these excitations and much of the evidence that magnetism might be the key to understand high-T<sub>c</sub> comes from experiments done at ILL and MLZ [17-21].

“Vortices are rather stable objects easy to move through space!” Despite the strangeness of this statement it is part of our common experiences. Hurricanes move along the landscape and remain stable for a while. As children we were amazed how water vortices in the bathtub withstood our trials to destroy them. 2009 a group of researchers around Christian Pfleiderer from TUM reported about an up-to then unknown order of magnetic moments in solid state, – so-called Skyrmions or vortices in MnSi at low temperatures. Neutron scattering with the suite of small angle cameras at MLZ was at the origin of this discovery [22]. Meanwhile, materials have been prepared which show this kind of order also at room temperature [23]. Their stability and the 100,000 times lower power needed to move them through the lattice, compared to shift a memory bit in conventional magnetic materials, make them to potential candidates for data storage or new sensors.



A lattice of magnetic vortices – so-called skyrmions – here in MnSi at 25 K as revealed by neutron scattering at MLZ [22] (Copyright S. Mühlbauer TUM).

The radioisotopes Lutetium-177, Holmium-166 and Terbium-161, produced at the FRM II by means of n-capture reactions, serve principally in tumor therapy, but occasionally in medical imaging, too. Technetium-99m is the most important and most commonly used isotope in nuclear medicine – some 30 million applications worldwide per year. There is a very wide range of applications in the field of diagnostic medicine. Technetium-99m arises as a fission bi-product of the irradiation of uranium. From 2020 on, the mother isotope Molybdenum-99 is to be produced in large quantities at the FRM II [24].

Bacteria become increasingly resistant against antibiotic. For instance, bacteria split the  $\beta$ -lactam ring of Penicillin and make it ineffective. Not long ago Leighton Coates and colleagues from the Oak Ridge National Lab detected by means of neutron protein crystallography at the MLZ, how this enzymatic reaction happens [25]. During the reaction a proton acceptor has to absorb a proton temporarily. There have been two contradicting hypotheses which molecule group will take over this task. The diffraction experiments on the protein diffractometer of MLZ revealed the amid group Glu-166 as the proton acceptor. Yet, the resistance of bacteria against antibiotic is not overcome, but to know the mechanism how the antibiotic is destroyed is certainly an important step forward to that goal.

Researchers are looking for new materials for future gas turbines, as the current “work-horse” Ni-Superalloys are reaching their service temperature limit because of the melting point of the material. One promising candidate is the cobalt-rhenium-chromium (Co-Re-Cr) system that exhibits a higher melting point in the order of 100 to 200 °C (depending on composition) than the Ni-based superalloys. Co-Re-Cr alloys are strengthened with nanoscaled tantalum carbides (TaC). Complementary neutron diffraction and small-angle neutron scattering measurements, especially in-situ at high temperatures, were performed to study the stability of these TaC precipitates. It turns out that TaC precipitates are stable at least up to 1200 °C, making the important precipitates very interesting for alloy strengthening. [26]

Nanotechnology aims to create new properties by modifying materials at the nanoscale. Polymers confined in nano pores are of particular interest since they offer a large range of applications such as coatings, lubrication, nanocomposites, biosensors or drug delivery. A recent investigation on the dynamics of polydimethylsiloxane (PDMS) chains confined in anodic aluminum oxide (AAO) revealed that the mobility of the PDMS is strongly affected by the confinement and follows a two-phase model: one free bulk-like fraction of chains and one phase of confined polymers. Access to these molecular motions on a time scale of 10 ns could only be achieved by so-called Neutron Spin Echo technology as it is established at MLZ [27].





In neutron spin echo technique the velocity of the neutrons before and after scattering at the sample is compared with highest accuracy. For this very high and geometrically precise magnetic fields are needed. At the corresponding instrument at MLZ this is achieved by superconducting magnets (the two big vessels before and after the sample). (Copyright O. Holderer, FZ Jülich)

### The future is bright

The prospects for the future of the three German neutron sources, ILL (with 1/3 German shareholding), MLZ/FRM II and TRIGA Mainz are clearly outlined. Currently, the Associates of ILL are preparing the 6<sup>th</sup> ten-years contract to last until 2033; MLZ/FRM II is expecting an operation time similar to that of ILL, i.e. beyond the year 2050; the university of Mainz intends to operate TRIGA beyond 2030.

And the new European flagship, ESS is under construction at Lund with the aim to deliver a decent service to the community with a first set of 15 instruments in the mid-twenties. ESS will have a time averaged flux as high as ILL or FRM II, but also the potential to deliver up to factor of 100 more in the pulse (pulse frequency 14 Hz, pulses length ~280 ms). With its first 15 instruments it will provide new capabilities, but less capacity. In the coming decade and beyond European neutron capacity will be provided by ILL, MLZ, the Swiss SINQ at PSI and the British ISIS near Oxford [28].

On December 12, 2018 the German and Russian Science ministers Anja Karliczek and Mikhail Kotjukow signed a German Russian Road map for an intense collaboration in the exploitation of large-scale research facilities [29]. This road map foresees that the MLZ partners, FZ Jülich, HZG and TUM build new and upgrade existing neutron instruments for the neutron source PIK at Gatchina near St. Petersburg. PIK is currently in a ramp up phase and will provide towards 2025 a neutron flux comparable to that of ILL and MLZ. Up to five "German" instruments will supply further capacities for the German and European neutron community.

The latest developments in the area of targets, moderators and neutron optics make the realization of an extremely compact accelerator driven neutron source possible. Different to spallation sources, these accelerators are of relatively low final particle energy (50 to 100 MeV), but with high currents. Such a beam would be directed to different target stations dedicated to specific applications and optimized for investigating small samples. Such high brilliance accelerator-based neutron sources HBS represents a unique infrastructure for neutron analysis (imaging methods and scattering), but also for industrial applications or clinical isotope production. They will be used in a multitude of scientific disciplines such as physics, chemistry, biology, medicine, geology, materials and engineering sciences. These HBSs ideally complement the larger international facilities such as the ILL or future ESS.

HBSs have the potential to create a network of different located sources for training, method development and specialization. FZ Jülich [30, 31] and the Laboratoire Léon Brillouin at Saclay [32] are heavily engaged in R & D for this new idea. First realizations are expected in the next 10 years.

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