IMPACT OF THE CRYOGEN FREE REVOLUTION ON NEUTRON SCATTERING LABORATORIES

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Abstract

A global shortage of helium gas can seriously jeopardise the scientific programmes of neutron scattering laboratories due to the use of cryogenic sample environment in the majority of the neutron scattering experiments. Recently developed cryogen-free technology allows a significant reduction or even a complete elimination of liquid helium consumption. Here we review the impact of the cryogen-free revolution on cryogenic equipment used at large neutron facilities, such as cryostats, dilution refrigerators, superconducting magnets and other cryogenic systems. Particular attention is given to the newly developed superconducting magnets for neutron diffraction and spectroscopy experiments. Use of the cryogen-free approach, as well as cutting-edge superconducting magnet technology and advanced neutron optics allows researcher to achieve extraordinary performance in their experiments, opening up new opportunities in neutron scattering research.

1. Introduction

In the last decade neutron scattering facilities have experienced a booming popularity. Two new major facilities, SNS in the United States and J-PARC in Japan, started operating and major new facilities such as ESS (European Spallation Source), CSNS (China Spallation Neutron Source) and PIK Russian neutron research reactor are either under construction or in an advanced design stage. Leading players in the neutron scattering field such as ILL in France and ISIS in the UK have significantly upgraded their capabilities. This rapidly growing interest in neutron scattering can be explained by astonishing new opportunities offered for users of neutron scattering facilities due to considerable progress in a number of areas such as neutron optics, neutron detection, large and complex dataset analysis, neutron scattering instrumentation and sample environment.

The majority of neutron scattering experiments are performed under cryogenic conditions [1]. There are fundamental reasons for this. Firstly, the thermal motion of atoms is reduced at low temperature, significantly improving the precision of structural measurements. Secondly, a cryogenic temperature range allows the study of low temperature phase transitions. Conventionally a cryogenic sample environment was provided by a liquid helium cryostat such as the ‘Orange cryostat’ [2]. However, extensive use of cryogenic equipment requires significant resources, creates a number of logistical issues including the considerable cost of the required cryogens and poses health and safety problems.

As we can see in Fig.1 the cost of liquid helium in the UK has doubled in the last five years and continues to grow. However, in the foreseeable future, we may experience a much more serious problem. According to the Helium Privatisation Act (1996) the US Federal Helium Reserve could be sold by 2015 creating a fear in the low temperature community that the situation with recent gigantic ^{3}He cost jumps may repeat itself with ^{4}He. There is also very strong environmental aspect of this problem. According to William Halperin: “Helium gas is a natural resource which is not replaceable and when released it will rise through, and escape from our atmosphere and be gone forever.” [3].

For neutron facilities this problem is of special importance because during the last two decades they are permanently in the top-ten list of liquid helium users in the scientific world somewhere after large accelerators, fusion reactors and MRI and NMR laboratories. Large-scale neutron facilities like ILL, SNS or ISIS used to consume a few tens of thousands litres of liquid helium annually. This situation posed one of the major risks on sustainability of a neutron facility's operation.

Luckily the revolutionary new cryogen-free approach emerged just at the beginning of the 21st century offering an effective solution. So, what is the essence of this approach?

In the case of a conventional cryostat, the cooling power required to maintain the cryogenic conditions is provided by evaporating liquid cryogens: helium and nitrogen. In the case of cryogen-free systems the cooling power is provided by a closed cycle refrigerator (CCR) also known as a cryo-cooler. There are two options available in order to reduce or, in the ultimate case, eliminate the usage of cryogens. The so-called "dry" systems do not contain liquid cryogens at all. These are built around a CCR that utilises the cooling power produced by the cold head [4-6, 34]. The other option is based on the idea of re-condensing the evaporating helium back to the cryostat by a CCR [11].

This revolutionary breakthrough became possible due to a new generation of CCRs developed during the last decade. Initially cryogen-free systems were using conventional Gifford-McMahon (GM) cryocoolers [12]. However despite obvious advantages, such as operational simplicity, the use of these systems was limited by the high level of mechanical vibrations produced by GM cryocoolers and the necessity of expensive and regular service inspections. A new generation of CCRs based on the pulse tube refrigerator (PTR) technology offers an effective solution to both problems [12 - 14]. A unique feature of the PTR is the absence of cold moving parts. This considerably reduces the noise and vibrations generated by CCR, a critical issue for a number of scientific instruments [6, 11, 12]. Furthermore the PTR offers lower maintenance costs and less disruption as expensive high-precision seals are not required and the cold head can be operated without service inspection.

Out of all the large-scale scientific facilities, the cryogen-free revolution produced most of its impact on neutron facilities because a cryogen-free approach based on CCRs is only efficient for small cryogenic devises alike cryostats, but not for bigger cryogenic systems such as large superconducting magnets and cavities of accelerators. In the last case industrial scale liquefiers are much more effective.

Here we have a brief review of impact of cryogen-free technology on the major cryogenic systems in a neutron facility experimental hall: cryostats, dilution refrigerators, superconducting magnets and neutron instrument components.

2. Cryogen-free cryostats

A cryostat is a device that allows maintenance of the cryogenic environment of a sample as long as required. In modern cryogenics there is a great variety of different types of cryostats and each type has advantages and disadvantages compared to others. Since the late nineteen seventies the Orange cryostat designed at the ILL [2] has been the workhorse of all neutron scattering facilities. This is a top-loading liquid helium bath cryostat with liquid nitrogen cooled infrared radiation shield and a base temperature of ~1.5K. However, at the beginning of 21st century, new cryostats based on cryogen-free technology have emerged. Even though all these cryostats developed at different facilities have the same basic principals of operation, the concept has generated many individual designs [4 – 10]. Photos of an example of top-loader system (based on PTR) developed by ISIS and manufactured by AS Scientific are presented in Fig. 2 [6, 10]. The system consists of the outer vacuum vessel (1); the top-loading insert with an internal diameter of 50, 75, or 100 mm (2); the CCR (3); the infrared radiation shield attached to the first stage of CCR (4); the thermal link between the second stage and the insert base flange (5). A sample is loaded into the insert on the end of a sample stick. The infrared radiation shield is made of highly conductive copper and is covered with high emissivity aluminium foil. The neutron beam access to the sample is provided through the thin aluminium or vanadium foil windows in the cryostat tails. In order to enable laser alignment of samples on the reflectometer instruments sapphire windows might be fitted.

For all designs of CCR-based top-loading cryostat, those based on PTR systematically reach lower base temperatures (2.9 - 3.5 K) than a GM based one (4.3 – 6K).

The system cooling time of under three hours for PTR based systems is also shorter than the approximately four hours for the GM based systems. The vibration level measured in the PTR cryostat is an order of magnitude less than that in a similar system based on a standard GM cryocooler [6].

For a rough estimation of the CCR-based top-loading cryostat efficiency, we can compare a cost of liquid helium required to run a standard Orange Cryostat at ~ 30 £/day (the liquid helium price as in 2011) with the cost of the electric energy consumed by a CCR ~ 8£/day [10]. This difference can only rise in the foreseeable future.

Currently ISIS has five PTR-based and eight GM-based top-loading cryostats and 21 Orange cryostats. Most neutron facilities, including ISIS, also have a number of bottom-loading CCR cryostats which can be used as a cryo-furnace with temperature range 2.8 – 700 K [5, 9]. The fleet of new cryogen-free cryostats allows the running of more than half of all the low temperature sample environment experiments at ISIS.

However, experiments which need the temperature lower than 2K still use Orange or similar cryostats. At ILL, a third stage Joule-Thomson refrigerator added to the GM 10K cryo-cooler reduced the temperature down to 1.8K [1]. Adding of the similar Joule-Thomson stage to the CCR-based top-loading cryostat for reaching 1.5K was later suggested in [5]. This idea has been realised in a prototype of the 1.5K cryogen-free top-loading cryostat based on a PTR, which has been developed and successfully tested at ISIS [7, 16]. The sample temperature range of this system is 1.45–300 K in the continuous flow regime. A sustainable cooling power of 55 mW at 1.82 K and temperature stability of ± 0.1 K across the temperature range 1.5-200 K has been achieved. From a user perspective, the system offers operating parameters very similar to those of an Orange cryostat, but without the complication of cryogens. The cryostat may also be used with ultra-low temperature inserts such as the Kelvinox VT dilution refrigerator. Preliminary tests have successfully demonstrated the operation of a Kelvinox VT insert at 75 mK.

High interest in cryogen-free cryostats in the neutron scattering community has stimulated industrial companies to develop commercial products for neutron scattering sample environment based on the technology. Currently Oxford Instruments, Ice Oxford, AS Scientific, Cryogenics, Janis and other companies have working prototypes at different stages of development.

3. Cryogen free dilution refrigerators

According to ISIS user statistics almost a quarter of all experiments require sample temperature below 1K. Such sample environment is usually provided by dilution and ^{3}He evaporation refrigerators. Many dilution refrigerators available at neutron facilities are based on the Grenoble design incorporating sintered silver heat exchangers [17, 18]. For many years this used to be the standard of conventional dilution refrigerators. Usually this kind of dilution refrigerators is built in a liquid helium bath cryostat with all the consequences typical for liquid cryogen based cryogenics.

The development of a cryogen-free dilution refrigerator started at the very beginning of the 21st century alongside development of cryogen-free technology in general [19-21]. The design of modern powerful cryogen-free dilution refrigerators in most cases is based on the prototype developed by Kurt Uhlig [22], where PTR is used to pre-cool the dilution unit. These powerful dilution refrigerators are usually used in experiments with heavy and large sample cells like those used for high pressure measurements. The photo of a similar powerful cryogen-free dilution refrigerator (E-18) developed by Oxford Instruments in collaboration with ISIS [7] is presented in Fig. 3. The fridge is capable of cooling large (diameter 200mm; height 250mm) and heavy (up to 20 kg) samples and provides access for the neutron beam through a set of thin aluminium alloy windows. The base temperature of the refrigerator is 15 mK and cooling power is approximately 370 μW at 100 mK. In experiments where E-18 has been used to cool down 110 cubic-centimetre solid and liquid ^{4}He sample cells (attached to the mixing chamber in Fig. 3) the base temperatures of 30 mK for solid sample and 50 mK for liquid sample (up to 25 bar) have been held for weeks [23-25]. The paper published in Physical Review B [23] became the first publication where a powerful cryogen-free dilution refrigerator has been used in a neutron scattering experiment. Today dilution refrigerators of this type are commercially available from a number of companies such as Leiden Cryogenics [26], Air Liquide [27], Oxford Instruments [28], LTLab [29], Cryoconcept, Janis, BlueFors and Ice Oxford.

In neutron scattering experiments, an ultra-low temperature sample environment is predominantly required for small samples. For this purpose compact dilution and ^{3}He refrigerator inserts can be used with the Variable Temperature Inserts (VTI) of a cryostat or superconducting magnet. These systems consume cooling power produced at the VTI heat exchanger. For Cryoconcept TBT®, dilution refrigerator insert developed by FRMII [8] and Oxford Instruments Kelvinox JT® (all of which incorporates Joule-Thompson stage [19]), the VTI temperature can be around 4K. For standard Oxford Instruments Kelvinox VT® and Heliox®, as well as for Leiden Cryogenics dilution insert, the temperature of the heat exchanger should be less than 2K. All of these systems can be easily made cryogen-free if used with cryogen-free cryostats. This can be achieved by inserting the refrigerator in the cryogen-free top-loading cryostat [16], or in a re-condensing cryostat with a VTI [7]. However the range of applications of these systems is limited by their relatively low cooling power (30 – 40 µW at 100 mK) and small sample space (diameter of the sample is <40 mm and height < 80mm).

Recently Oxford Instruments has developed a lower power cryogen free dilution refrigerator with cryogenic circulation [30], where the temperature of circulating ^{3}He never exceeds 50K. However, this refrigerator design does require more ^{3}He than a standard one with similar performance. The recent dramatic increase in the price of ^{3}He [31] has made the fridge commercially unviable.

4. Cryogen-free superconducting magnets.

Neutron scattering is an invaluable tool for solid state magnetism. In many experiments a sample needs to be exposed to a high magnetic field, low temperature and a neutron beam simultaneously [1]. Nowadays these conditions can be mostly satisfied in superconducting magnets. In the middle of the 1990s, the ILL ordered, for the first time, two niobium-tin high-field vertical split-pair magnets giving fields up to 8T and 12T [32]. But, they were not very satisfying as there were frequent leaks and quenches. The appearance of high stability digital power-supplies and new NMR-style stainless steel cryostats allowed these problems to be resolved and in the middle of 1990s, the Hahn-Meitner Institute (HMI) in Berlin ordered three magnets which became the standard for neutron scattering sample environment [33]. Most neutron-scattering magnets are split-pair magnets made of NbTi and Nb_3Sn filamentary superconducting wires. The magnet is usually built in a conventional liquid helium bath cryostat with a thermal shield cooled by liquid nitrogen. A set of windows made of neutron transparent materials provides a neutron beam access to the sample. A good example is the HMI magnet designed for neutron diffraction [33]. The maximum magnetic fields are 12 T at 4.2 K and 14.5 T at 2.5 K and the vertical opening angle around the beam direction is ±2°. The total thickness of Al-screens and Al-rings in the beam path is 30 mm. At the time, it was most advanced magnet of its kind.

However, in the last decade the rapid developments in superconducting magnet technology [38] and the cryogen-free revolution have made it possible to build a new generation of advanced magnets for neutron scattering [34 - 39]. The new magnets are usually designed to satisfy specific requirements of a particular advanced neutron scattering instrument such as magnet aperture and opening angle tailored to match the coverage of the instrument detectors, or compatibility with special collimating systems. Then the optimal combination of instrument and magnet allows much faster experiments with much higher resolution. All cryogen-free superconducting magnets fall into two separate groups according to their cryogenic design: dry or re-condensing systems.

In dry systems, both stages of the CCR are connected directly to the infrared radiation shield (first stage) and the magnet (second stage) [9, 34]. Therefore the magnet is suspended in vacuum and provision of the thermal equilibrium relies on effective heat transport through the magnet's body, which is not easy to achieve. Higher temperature gradients result in lower maximum magnetic fields and slower field sweeping rates. For example, the maximum magnetic field of pioneering cryogen-free neutron diffraction magnet developed by the Japan Atomic Energy Research Institute [34] is just 10 T in comparison with 14T achieved by a similar magnet submerged in liquid helium [36]. Other important parameters of the JAERI magnet are the vertical opening angle: ±3° and the total thickness of Al-screens and Al-rings in the beam path is 52 mm. These are comparable with the 12T HMI magnet [33].

Split-pair high-T_c superconducting (HTS) beam-line magnets have been recently developed by HTS-110 Ltd. for HMI and the Australian OPAL neutron facility [35]. Cryogenically, both magnets are dry cryogen-free systems. The OPAL neutron beam-line magnet was designed to provide 5T horizontal-field for a range of neutron scattering techniques including reflectometry, SANS and time-of-flight inelastic scattering. The magnet can be mounted on a goniometer so that it can be tilted up by up to 15° from vertical.

Initially re-condensing cryostats were developed for superconducting NMR magnets [11]. In this case, a superconducting magnet is immersed in the liquid helium. The radiation shield is cooled by the CCR's first stage and the second stage re-condenses helium directly in the helium vessel. Thus the re-condensing system does not consume any liquid helium in normal operation. The main advantage of this system is that all magnet operating procedures, for example cooling, running up to the field and quenching remain the same as for a standard magnet in a bath cryostat. This approach also provides a homogeneous temperature distribution, which is crucial for optimum magnet performance [38]. The maximum magnetic fields are significantly higher for re-condensing magnets in comparison with similar dry systems. All standard accessories like current leads, magnet power supply, helium transfer siphon, etc. can be used with this system. One more important advantage is the ability of the magnet to stay at field for more than forty eight hours before quenching in the case of CCR power failure. This feature also makes it possible to move the cold magnet between different neutron scattering instruments and/or the cryogenic laboratory. The cooling power required for re-condensing is provided either by PTR [36, 39] or by GM cryocooler [37]. PTR has advantages because it has no cold moving parts and, therefore, has lower level of mechanical vibrations and requires little maintenance.

In the next three subsections the design and the experience of operating the 14T magnet for neutron diffraction and the 9T wide angle chopper magnet for neutron spectroscopy developed by Oxford Instruments in collaboration with ISIS [36] will be discussed. We also review preliminary scientific data [39] which demonstrates the magnet's performance. To the best of our knowledge these two magnets can be used as examples of today's most advanced magnets for neutron scattering sample environment.

4.1 14T magnet for neutron diffraction measurements at ISIS

The 14T magnet for diffraction measurements at ISIS is a high-field split-pair magnet which consists of NbTi and Nb_3Sn superconducting coils. The 3D model of the magnet is presented in Fig. 4(a) Today, the maximum magnetic field for a state-of-the-art split-pair magnet is 15T, with an opening angle in vertical direction to ±3°, but by limiting the maximum field to 14T it is possible to increase the detector viewing angle to ±7.5° [38]. In order to optimize the magnet design for using on Wish long-wavelength diffractometer at ISIS, we have chosen an asymmetric split -5° to +10°, which for single crystal studies, allows access to at least one extra scattering plane out of the main horizontal plane. In order to minimize the risks associated with quench during magnet operation at 14T, it was decided to restrict the maximum magnetic field to 13.7T. In the future, it is planned to use dysprosium inserts [33] which should push the magnet maximum field up to ~16T.

The magnet split is supported by aluminium rings (Fig.4 (b)). The total thickness of Al alloy in the beam path is 27 mm. The surfaces of the aluminium rings which are not a part of the neutron scattering aperture are plated with cadmium in order to reduce secondary neutron scattering. In most cases the 14T magnet is used with the radial collimator (Fig.4(c)) which allows the magnet background scattering to be significantly reduced. The main 14T magnet technical specifications are given in Table 1.

Table 1 Main technical specifications of 14T magnet for diffraction measurements

Maximum field	13.7T

Field homogeneity:	0.45% (over 10 mm sample volume diameter)
Persistent mode stability	<0.01%/hr
Stray field (at max field)	≤ 0.007T at 2m distance from magnet centre
Vertical opening	-5° to +10° (to the back of the 10 mm cylindrical sample height)
Horizontal opening	340°
Magnet bath temperature	4.2K

Fig. 5 shows the diffraction pattern of the ‘spin ice’ sample $Ho_2Ti_2O_7$ (courtesy of R. Aldus, UCL) previously used in neutron scattering investigation of the material’s kagome-ice plateau [40]. The data was obtained at 150 mK and 1.6T. The diffraction data demonstrate the very good angular coverage and excellent data quality (low noise) possible when using the collimator and the cadmium shielding.

The diffraction pattern of a pyrochlore antiferromagnet sample at 70 mK (0 T and 13.5 T) (courtesy of Ross Stewart, ISIS) is presented in Fig. 6. Very good angular coverage and low noise in the data have been achieved by using the collimator and cadmium shielding.

4.2 9T wide angle chopper magnet at ISIS

The 9T wide angle chopper magnet for spectrometry at ISIS is a split pair magnet which consists of NbTi and Nb_3Sn superconducting coils, and is similar to the 14T diffraction magnet. The 3D model of the magnet is presented in Fig 7(a). The magnet split is supported by non-magnetic stainless steel wedges rather than aluminium rings (Fig. 7(b)). This design allows the amount of the material in the beam path to be minimised. The chopper instruments derive their power from being able to survey large volumes of reciprocal space in a single measurement, and so it is vital to build a magnet that can exploit this advantage. This would necessitate a trade-off between maximum field and a wide aperture [38]. Detailed modeling allowed us to design and manufacture the 9T magnet with ±15 ° opening in the vertical plane and ±45° (two openings separated by 180°) opening in horizontal plane. In addition we also have 50mm diameter hole at 90° to allow the beam in and out, which allows coverage of larger angles in the horizontal scattering plane. Such a magnet can be particularly valuable for exploring the competing phases and the microscopic interactions in the field of strongly correlated electron systems using instruments such as Merlin and Let at ISIS (with the unprecedented continuous angular coverage of their detectors). 1.5mm thick boron nitride tiles have been attached to the beam entry slots of the magnet former in order to reduce the background of the magnet (Fig.7 (c)). A 2mm thick boron nitride tube has also been installed in order to shield the bore of the magnet. The main 9T magnet technical specifications are given in Table 2.

Table 2 Main technical specifications of 9T wide angle chopper magnet

Maximum field	9T
Field homogeneity:	0.4% (over 10 mm sample volume diameter)
Persistent mode stability	<0.01%/hr
Stray field (at max field)	≤ 0.00045T at 4m from magnet centre
Vertical opening	30° (±15 °) (to the back of the 25 mm cylindrical sample

	height)
Horizontal opening	90° (±45°)
Magnet bath temperature	4.2K

Fig. 8 demonstrates the difference between neutron scattering signals from a $Ba_2PrRuIrO_6$ sample in the magnet (Fig. 8 (a)) and from the empty magnet (Fig. 8 (b)). The measurements have been performed on the Merlin instrument at ISIS (data courtesy of Devashibhai Adroja). Most of the magnet's small background signal is produced by thin aluminium windows in the outer vacuum can, infrared radiation shield, and VTI. The background signal from secondary scattered neutrons is almost negligible.

Figure 9 shows data from the first experiment on the Let instrument at ISIS (courtesy of Radu Coldea). The sample is a single crystal (~6g) of $CoNb_2O_6$, a quasi-one-dimensional Ising ferromagnet. Below a critical magnetic field the observed spin excitations are from pairs of 'kinks' in the ordered phase (see Fig. 9a). However, above a critical field the system undergoes a quantum phase transition to a quantum paramagnet and the spin excitations observed are those of spin-flips in the paramagnetic phase (Fig. 9b). For more details on this system, see reference [41]. Both sets of data were taken with an incident neutron energy, E_i= 4 meV and have a resolution at the elastic line of approximately 1% of $\Delta E/E_i$. This data is shown without subtraction of a background signal. The data is clean and demonstrates the low background and high energy resolutions achievable.

4.3 Low temperature sample environment

Both the 14T and 9T magnets at ISIS share similar top parts of the re-condensing cryostat with a VTI, but have different bottom parts accommodating the split pair magnets and sample space.

The cooling power for helium re-condensing is provided by a CryoMech PTR PT410. In a regime without a VTI, helium circulation and no ramping of field, the pulse tubes on both magnets perform so efficiently that all the helium in the cryostat is re-condensed and there is an excess of cooling power between 600 and 650 mW [36]. This extra cooling power allows re-condensing of the helium flow passing through a VTI during the operation of a dilution refrigerator insert. We have recently demonstrated zero helium boil-off operation of the re-condensing 9T superconducting magnet cryostat with a Kelvinox VT dilution refrigerator insert running at a base temperature of 63 mK during a 24 hour period [42]. This approach allows cryogen-free operation of combined high magnetic field and ultra-low temperature sample environment for neutron scattering experiments. This method offers significant reduction in liquid helium consumption as well as the advantage of operational simplicity and significant improvement in user safety. Estimations based on half a year of 14T magnet cryostat operation statistics allow us to conclude that one re-condensing magnet cryostat saves up to 7500 L/year of liquid helium.

5. Special cryogen-free sample environment systems

In the last decade cryogen-free technology has spread far beyond traditional low temperature/high magnetic field sample environment and allowed conceptually new systems to be created. In this section I present a few examples of unconventional sample environment based on the cryogen-free approach.

There has been growing demand for investigations of the mechanical behaviour of materials at cryogenic temperatures due to the recent progress in cryogenic technologies. Applications include cryogenic texture processing of alloys, strain sensitivity of superconducting magnet wires, cryogenic structural steels and low temperature shape memory alloys for space applications. ISIS has developed a cryogen-free cryogenic testing chamber for neutron scattering measurements of internal stress in engineering materials under loads up to 50kN and at temperatures from 30K to 300K [43].The schematic diagram of the stress rig is depicted in Fig.10. Two CCRs provide the cooling power for keeping the sample at cryogenic temperatures. A similar approach has been used in the design of cryogenic load frames developed at the LANSCE [44] and JAEA [45] facilities. A distinctive characteristic of the JAEA device is the lower base temperature ~4.8K, which allows the study of stress-strain effects in type two superconducting materials simultaneously with critical current measurements.

Another example of cryogenic sample environment is the cryogen-free cryostat for cooling a 10 GPa Paris Edinburgh cell down to 3K built by the ILL [5]. The 18 kg cell is attached to the second stage of the CCR and is enclosed in a tight chamber filled with exchange gas. It takes only 4 hours to reach 3K from room temperature. A cryogen-free cell based on a different design developed by FRMII allows high pressure (up to 0.2GPa) sample environment to operate in the temperature range 20K – 600K [8, 9]. The cooling down time of this cell is similar to the ILL one.

Significant reductions in the neutron beam-time required for measurements with the new generation of neutron scattering instruments creates the need for remote and fast sample handling to enable a high throughput of samples in a controlled environment. A cryogen-free automated sample changer has been developed at SNS to serve this purpose [46]. The samples are contained in hermetically sealed vanadium cans attached to the sample handler. The system uses a CCR for cooling. The time to cool the sample from ambient temperature to ~10K is approximately 7 min. A resistive heating coil can be used to extend temperature range from 10K to ~350K.

6. Cryogen Free technology in neutron scattering Instruments

Compactness and reliability of CCRs allowed them to occupy a new niche as coolers for components of neutron scattering instruments. Here I present a few examples of this approach.

The most widespread application of this kind is the CCR-based cryogenic beryllium filter. Polycrystalline beryllium Bragg cut-off occurs at an energy of 5.2 meV [47]. A block of beryllium transmits low energy neutrons up to this value, but scatters neutrons of higher energy. The beryllium filter based on this beryllium property is getting more and more popular in designs of neutron spectrometers. The transmission of the filter for neutrons of wavelengths beyond the cut-off depends mainly on phonon scattering. This can be largely removed by cooling the filter below 100 K and so removing the energy gain scattering from thermally excited phonons. Keeping beryllium filters at cryogenic temperature in the Tosca (ISIS) [48] and Vision (SNS) [49] instruments requires five CCRs, whereas in case of the Lagrange (ILL) [50] and the recently upgraded Macs (NIST) [51] spectrometers one or two CCRs provide enough cooling power.

Some instruments might require cryogenic sample environment for the majority of experiments. In this case a cryogenic sample chamber can be incorporated in the design of the instrument. For the cryogenic chamber embedded in the Tosca instrument, the necessary cooling power is provided by three CCRs, which allows measurements in the range between 10K and room temperature [48].

A cryo-pump is a vacuum pump that traps gases and vapours by condensing them on a cold surface. In neutron instruments cryo-pumps based on CCRs are used to improve the level of vacuum and accelerate pumping-out of large vacuum chambers. For example, after venting the sample chamber of SNS instrument Arcs, a combination of a large mechanical pump with Roots blower and a cryo-pump restores the vacuum to the 10^{-6} mbar range in under 15 min [52]. CCR based cryo-pumps have also been used for the ISIS instruments Maps [53] and Merlin [54].

An elegant exploitation of superconductivity has been realised in the cryogen-free compact Cryogenic Polarization Analysis Device (Cryopad) developed at the ILL [55]. This device takes advantage of Meissner shields made of niobium to properly define the magnetic field and zero-field regions crossed by the incident and scattered neutron beams. Using Cryopad, all the components of the complicated expression of the final polarization vector of a neutron beam can be measured, which provides unique information on magnetic structures and nuclear/magnetic interferences occurring in the scattering process.

7. Conclusions.

The cryogen-free revolution started in the early 2000s and is not over yet. Every year cryogen-free technology conquers new areas traditionally occupied by conventional cryogenics. However the first decade of using cryogen-free technology has allowed the accumulation of vast operational experience. Evaluation of the experience gained by ISIS and other large neutron facilities, which are arguably the most intense users of cryogen-free equipment, give us an opportunity to determine the advantages and disadvantages of these systems.

Advantages common for all cryogen-free systems:

- Operational simplicity. Most of the cryogen-free systems need only maintenance service of the CCR compressor after installation. Usually this means just changing compressor filters every two-three years;
- Reduction, or in some cases, complete elimination of liquid cryogen top-ups, which is logistically demanding and expensive. Cryogen-free technology also reduces system downtime caused by top-ups;
- Significant reduction in technical resources. No need for specially trained personnel to prepare conventional cryostats for an experiment and perform top-ups;
- Cryogen-free technology is safer. The involvement of technical personnel regularly handling cryogens raises quite serious safety issues. The highest risk is associated with asphyxiation due to the lack of oxygen replaced by rapidly evaporating nitrogen or helium. This is a potentially lethal hazard. Cryogen-free technology significantly reduces and in some cases completely get rid of all cryogenic hazards involved;
- Thermodynamic efficiency. Despite the fact that a cryocooler is less thermodynamically efficient than any industrial liquefier, the total efficiency of cryogen-free systems is comparable with conventional cryogenic systems. This can be explained by the significant losses with conventional cryogenics due to transporting and storing cryogens and transferring them to cryostats. Cryogen-free technology eliminates these losses because cooling power is directly supplied from a cryocooler to the cooling apparatus;
- Cryogen-free technology significantly reduces the system size. The systems do not have a nitrogen vessel and do not require extra volume for holding liquid helium, which is necessary for providing sufficient hold time in the case of conventional cryostats;
- Cryogen-free technology is much more environmentally friendly than conventional cryogenics. Helium losses are minimized and the strategic gas can be saved for further generations.

Evaluation of our operational experience also reveals some disadvantages of cryogen-free systems:

- The most serious disadvantage of cryogen-free systems is the limited cooling power of the CCR. The most powerful GM cryo-coolers and PTRs achieve no more than 1.5W cooling power at 4.2K. In conventional liquid helium based systems, one can easily achieve more than an order of magnitude higher cooling power just by opening a needle-valve, but increasing helium consumption as a result. In comparison with conventional ‘Orange cryostat’ this means longer sample change time. However, in the case of re-condensing systems one can accelerate sample cooling by allowing helium boil-off.
- Cryogen-free equipment significantly increases demand for electricity and cooling water supplies, which in the case of massive scale operations can be a major issue.
- The cost of the cryo-cooler (~$30000) can be a significant addition to the total cost of a cryogen-free cryostat;
- CCR operation generates significant noise, mechanical vibrations and magnetic field disturbances. This difficulty has been drastically alleviated by the absence of cold moving parts in the PTR

design. However, in cases of measurements extremely sensitive to such disturbances, special additional arrangements might be required;

- Some of the PTRs experience deterioration of cooling power (from 10% to 20 % after ~ 5 years in operation), although the fast development in CCR technology promises significant improvement of PTR reliability in the near future.

Cryogen-free systems already make up a significant part of neutron facility cryogenic equipment and the situation continues to change rapidly. In the early 2000^s, at the planning stage of the new research reactor FRM-II in Munich, a decision was made to base all FRM-II sample environment equipment on cryogen-free technology, excluding all conventional cryogenic systems from operation [8]. Similar internal development programs intended to gradually substitute all conventional cryogenic systems with cryogen-free systems are being carried out at ISIS [10] and ILL [5]. The benefits of cryogen-free technology lie not only in reducing helium consumption and eliminating the risks associated with global helium availability, but also in the astonishing operational simplicity of cryogen-free systems, reduction in required technical resources and much improved safety. All these aspects are crucial for efficient exploitation of any large scale neutron facility. A combination of cryogen free technology with advanced neutron optics, neutron scattering instrumentation and more powerful neutron sources open up extraordinary new opportunities across broad areas of science.

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Figures:

Fig. 1 Cost of liquid helium in the UK.

Fig. 2 Photos of a pulse tube refrigerator top-loading cryostat. The system consists of an outer vacuum vessel (1); top-loading insert (2); CCR (3); infrared radiation shield (4); thermal link between the second stage and insert base flange (5).

Fig. 3 Powerful cryogen-free dilution refrigerator developed by Oxford Instruments in collaboration with ISIS [7].

Fig. 4 (a) 3D model of 14T magnet for diffraction measurements at ISIS; (b) The split magnet is supported by aluminium rings; (c) The radial collimator.

Fig. 5 Single crystal diffraction from 'spin ice' sample $Ho_2Ti_2O_7$ at 150mK and 1.6T (courtesy of R. Aldus).

Fig. 6 Diffraction pattern of pyrochlore antiferromagnet sample at 70 mK (0 T and 13.5 T) (courtesy of Ross Stewart, ISIS).

Fig.7 (a) 3D model of 9T wide angle chopper magnet for spectrometry at ISIS (b) The split magnet is supported by nonmagnetic stainless still wedges (c) Boron nitride tiles attached to the beam entry slots of the magnet former.

Fig. 8 Difference between neutron scattering signals from (a) $Ba_2PrRuIrO_6$ sample in the ISIS 9T magnet, and (b) from the empty magnet. (data courtesy of Devashibhai Adroja, ISIS).

Fig. 9 Spin excitation spectrum of $CoNb_2O_6$ taken at 50 mK and with an incident neutron energy of 4 meV (courtesy of Radu Coldea) using the ISIS 9T magnet (a) 4T (b) 7T.

Fig.10 Schematic diagram of the Engin-X cryogen free stress rig at ISIS [43].