

Changes in Cooling Strategies from 2005 to 2025 at Temperature from 3 K to 25 K using He or H₂ as a Coolants

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Abstract. The strategy for cooling in the temperature range from 3 to 25 K is likely to change because of the working fluid cost (in the case of helium) and safety issues (in the case of hydrogen). In both cases this requires minimizing the inventory of the working fluid for the reasons stated above. These limitations may apply for systems using coolers and definitely apply for systems that involve central refrigeration. This paper covers a range of temperature of interest to superconducting devices that use LTS and HTS conductors. The methods for minimizing the gas inventory are similar for the two gasses. Hydrogen has many advantages for cooling above 15 K. Liquid hydrogen has a heat of vaporization that is over twenty times that of liquid helium. This paper will show how one can reduce the amount of these liquids in cryostats. Hydrogen appears to be a better working fluid than helium for cooling down a device using natural convection. Hydrogen safety issues and other issues with hydrogen will be discussed.

1. Introduction

Before explaining why there is a change in cooling system design from 2005 to 2025, it is useful to look at the history of the refrigeration systems of the production and circulation of helium [1]. They are basically the same for both gasses except for the hydrogen safety issues. The cryostats for cryogenic helium and hydrogen are also basically the same. The important difference is the safety measures need for cryostats for liquid hydrogen and methane [2]. The basic design for any low temperature gas cryostat is based on the vacuum insulated vessel was invented by James Dewar in 1895 [3]. The vessel greatly reduced thermal conduction and convection losses between room temperature and the liquid cryogen vessel. With reflective coatings on the walls of the vacuum chamber, it reduces radiation heat transfer as well [4]. Hydrogen was liquefied by James Dewar in 1898 [5]. Helium was much harder to liquefy and contain. H. Kammerlingh Onnes at Leiden University liquefied helium using a double cascade cooling with LN₂ and LH₂ pre-cooling in 1908 [6]. The ability to liquefy helium led to the discovery of superconductivity in 1911 by the H. Kammerlingh Onnes' group in Leiden [7].

Superconductivity research required liquid helium and cryostats to hold the helium for the duration of the experiments. The basic helium cryostats didn't change much between 1911 and the late 1940s. The experiments and the sub-components within the basic cryostats did change. W. F. Giauque at UC Berkeley was convinced that adiabatic demagnetization refrigeration could lead to temperatures below 1 K. Giauque built a liquid helium facility based on a better double cascade refrigerator at Berkeley and a magnetic refrigerator to create a temperature in Berkeley of 0.25 K [8].



Any system of superconducting magnets that uses cryogenic hydrogen or helium, one must have reliable refrigeration. In 1933, Sam Collins of MIT began working on a small liquefier for hydrogen and helium based on the Claude cycle with two piston expanders, regenerative heat exchangers, LN_2 pre-cooling and a J-T valve to produce liquid helium or hydrogen. The project was delayed because of a lack of funds during the depression and his other work during the second world War. His first helium refrigerator was tested at MIT in 1948 [8] [9]. A version of his liquefier was used to liquefy deuterium and tritium for the first hydrogen bomb test in November 1952 [10]. The Collins liquefier became a commercial product in 1956 [11]. By the late 1970s or early 1980s large refrigeration systems became sufficiently reliable to be used on accelerator rings and separate magnets for experiments [1]. At the same time helium, cryostats became reliable enough for large accelerators and their experiments.

Cooler technology for helium or hydrogen took longer to develop. The Gifford McMahan (GM) cooler in 1959 [12] and the Pulse Tube cooler in 1963 [13] were both developed at Syracuse University. The first two stage coolers with second stage temperature as low as 20 K were also developed at Syracuse University, in 1966 [14]. These coolers were used to cool Maser amplifiers and cryogenic vacuum pumps. A two-stage cooler with a separate coupled circuit cooled by the two cold heads and a final J-T valve expansion to 4.5 K was also developed at Syracuse University in 1969 [15]. There must be separate compressor for the J-T circuit. There were heat exchangers between the top plate, both stages and the J-T valve or a more efficient wet expander. The breakthrough in cooler technology is the development of high heat capacity regenerator material at low temperatures in 1990 [16]. With the high heat capacity regenerator, the second stage of a two-stage cooler can go down to ~ 2.7 K with helium 4 as a working fluid [17]. When the high heat capacity regenerator is used in a single stage cooler, the cold head can go down to ~ 15 K [17]. With helium 3 as a working fluid a two-stage cooler can go below ~ 2 K to produce superfluid helium 4. [18]. All of this assumes that the insulation and shields are properly designed, and the cryostat vacuum is low ($< 10^{-7}$ Pa) [19]. By the late 1990s, both Sumitomo and Blufors had made real strides in developing reliable 4 K to 80 K coolers of both types.

2. The Properties of Hydrogen and Helium as a Gas and a Liquid

Hydrogen and Helium are the only elements on top row of the periodic table. Hydrogen is on the far left side of the periodic table, which makes it a highly reactive gas. Helium is on the far right side of the periodic table is a noble gas. Hydrogen is a two-atom molecule and helium is a single atom.

2.1 Properties of Hydrogen

Hydrogen has three isotopes; hydrogen without a neutron H, heavy hydrogen with one neutron (deuterium) D and super heavy hydrogen with two neutrons (tritium) T. All three isotopes of hydrogen take the same molecular form as ordinary hydrogen H_2 . Hydrogen in nature is a mixture hydrogen and deuterium. Tritium is radioactive with a half-life of ~ 12.3 years. As a result, there is virtually no tritium in a bottle of hydrogen. Deuterium is not radioactive, so it is present in hydrogen gas at ~ 155 parts per million. In molecular form deuterium can be present as HD or D_2 , which represents a slight variation in properties such as boiling temperature as a function of pressure, triple point temperature, and critical temperature and pressure. Every one of these temperatures are higher for deuterium. Ordinary hydrogen is a better working fluid than either of its isotopes.

In 1912, Arnold Eucken discovered an unusual heat capacity in liquid hydrogen [20], but it was not resolved until 1927 by W. Heisenberg and F. Hund [21]. The issue with molecular hydrogen is the spin states within the molecule. The hydrogen molecule has two protons, which are fermions with a spin of $\frac{1}{2}$. Thus, molecular hydrogen has two extremes in its spin energy state, 0 and 1. The highest energy state is 1, the ortho state when the spin states are aligned, and the lowest energy state is 0, the para state when the spin state oppose each other. It is only 0 at very low temperatures. Deuterium is more complicated because it has one or two neutrons that are bosons that have a spin of 1. A deuterium can have up to six spin states depending on the atoms in the molecule and the spin orientation. Fortunately, this can be ignored in ordinary hydrogen because can only have at most 0.015 percent deuterium in the gas. Hydrogen at 293 K is about 75 percent ortho hydrogen, and at 20.3 K the minimum amount of ortho hydrogen ~ 2 percent. The maximum heat of transition from ortho to para is 721.8 kJ kg^{-1} . Thus,

the total transition energy $\sim 527 \text{ kJ kg}^{-1}$. Without a catalyst this energy release is delayed, which can result in the sudden boil-off of up to 50 percent of the liquid hydrogen. There are a number of catalysts that can be used from ferric oxide to rare earth metals that can speed up the transition process. These catalysts must be at 80 K when H_2 is about $\sim 50\%$ para and at a lower temperature where H_2 is $\sim 20\%$ para. This suggests that liquid hydrogen already in the para state can be pumped without reverting to the ortho state, as long as the pump work is taken out at a liquid temperature 20.3 K or below.

Hydrogen embrittlement occurs in metals when atomic hydrogen is present. Embrittlement reduces the metal's ductility and with time it will induce cracking that can lead to metal failure while under stress. Things like high strength bolts can fail in the presence of water as they did in 2012 on the San Francisco Oakland Bay Bridge. Hydrogen embrittlement is a potential problem in vessels and pipes that contain hydrogen or fluids that contain hydrogen. This effect was first studied in the second half of the 19th century [22], when steam engines were powering everything. Hydrogen embrittlement affects high strength steels as well as ordinary steels even at low temperatures [23], [24]. Unfortunately, titanium is on the list of materials that are subject H_2 embrittlement. There are metals that are prone to embrittlement while they are warm, but not when they are cold. These metals include austenitic stainless steels, aluminum and its alloys, and copper and its alloys (for example beryllium copper). Hydrogen embrittlement is on the list of factors that has to be considered for hydrogen safety. For more information on H_2 embrittlement in material contact NASA or the Sandia National Laboratory in the United States. Cambridge University in the United Kingdom may be another good source of information.

2.2 Properties of Helium

Helium has two isotopes; helium four with two protons and two neutrons and helium three with two protons and one neutron. Neither isotope is radioactive. Helium three is by-product of tritium decay. In nature helium three is very rare. The primary source for helium three is from the decay of tritium that is produced in a nuclear reactor. Helium three is very expensive. It is a gas that can be a refrigerant in small coolers or dilution refrigerators. Unlike hydrogen, helium 4 zero net spin of zero. Thus, it doesn't have any spin energy gain or loss between room temperature and absolute zero. In the excited state there may be some spin related issues.

Helium 4 (the usual isotope) does not have a triple point like other substances. The triple point temperature for most things is when solid, liquid, and gas can co-exist. The defined triple point for helium 4 is the temperature where gas, normal helium and superfluid helium [25], [26] can coexist. The temperature of this point is 2.172 K and the pressure is 5.04 kPa. One must go to a pressure of 2.533 MPa to get solid helium 4. One might argue that there is another non-traditional triple point between two liquid phases and the solid phase in helium 4, but this is unlikely to be encountered.

Helium 3 is a rather boring fluid compared to helium 4. Helium 3 has normal triple point a 2.6 mK and 0.0704 kPa. One must go below the triple point temperature to have any superfluid phases [27]. There are two superfluid phases A and B coexist with the solid phase. The highest temperature superfluid exists at about 2.8 mK at the solid phase line. The lowest temperature triple point involves the gas, superfluid B and the non-superfluid liquid at $\sim 0.9 \text{ mK}$ and $\sim 0.024 \text{ kPa}$. Helium 3 combined with helium 4 do work together to get very low temperatures in a dilution refrigerator [28]

Liquid He 4 has a very high expansion coefficient (~ 25 percent per degree K) at 4.5 K. There have been instances when helium expansion was thought to be liquefaction when the heat transfer was poor between a cooler cold head and the helium in the cryostat [29]. Hydrogen at 20.3 K has an expansion coefficient an order of magnitude lower than for liquid helium at 4.2 K. Liquid nitrogen at 77 K has an expansion coefficient that is two orders of magnitude smaller than helium at 4.2 K.

2.3 A Table of Hydrogen and Helium properties that is useful for Calculation

The table that follows contains the data of hydrogen and helium [30]. The data include the boiling temperatures at 101.33 kPa, the triple point and critical temperature and pressure.

The table also includes data on density for saturated liquid and saturated gas for both fluids. The spin enthalpy and total enthalpy for both gases are also included. The final four rows in the table are the thermal conductivity and viscosity for both the liquid and the gas for both fluids at the 101.33 kPa boiling temperature.

TABLE 1. Parameters of Cryogenic Fluids [30]

| Fluid Property | He [^] | H ₂ [^] |
|---|-----------------------|-----------------------------|
| Triple Point Pressure P _{TP} (kPa) | 5.17* | 7.09 |
| Triple Point Temperature T _{TP} (K) | 2.17* | 13.8 |
| Boiling Temperature T _b at 101.33 kPa | 4.22 | 20.3 |
| Liquid Density at T _b (kg m ⁻³) | 125 | 70.8 |
| Critical Temperature T _c (K) | 5.19 | 32.3 |
| Critical Pressure (kPa) | 223.9 | 1308.2 |
| Fluid Density at T _c and P _c (kg m ⁻³) | ~69.7 | ~31.0 |
| Heat of Vaporization at T _b (J kg ⁻¹) | 20900 | 442000 |
| Spin Enthalpy from T _b to 273 K (J kg ⁻¹) | 0 | ~530000 |
| Enthalpy w/o Spin T _{TP} to 293 K (J kg ⁻¹) | ~1578000 | ~4400000 |
| Enthalpy with Spin T _{TP} to 293 K (J kg ⁻¹) | ~1578000 | ~4930000 |
| C _p of Gas at 101.33 kPa T _b to 273 K (J kg ⁻¹ K ⁻¹) | 5200 | 14200 |
| C _p of Saturated Liquid at T _b (J kg ⁻¹ K ⁻¹) | ~2500 | ~9800 |
| Thermal Cond. Gas at T _b (W m ⁻¹ K ⁻¹) | 0.011 | 0.021 |
| Thermal Cond. Liquid at T _b (W m ⁻¹ K ⁻¹) | 0.027 | 0.119 |
| Viscosity Gas at T _b (kg m ⁻¹ s ⁻¹) | 0.91x10 ⁻⁶ | 1.05x10 ⁻⁶ |
| Viscosity Liquid at T _b (kg m ⁻¹ s ⁻¹) | 3.53x10 ⁻⁶ | 1.34x10 ⁻⁵ |

*Helium 4 triple point temperature is based on two liquid phases and the gas phase.

[^]The properties above assume ⁴He and H₂ are 99.999% pure.

3. Cooling Methods

3.1 Cooling Devices with flow from a large Helium or Hydrogen Refrigerators

Three large bubble chamber magnets were built between 1970 and 1980 [31]. These magnets had a pair of cryogenically stable superconducting coils inside a large cryostat filled with liquid helium. The space between the coils allowed the beam to enter the hydrogen chamber and pictures to be taken of the hydrogen bubble tracks through windows in the hydrogen cryostat. These magnet coils were in a helium bath and the bubble chamber had up to 50 m³ of hydrogen and even more helium [32]. The refrigerators that kept the magnet coils and bubble chambers cold were large liquefiers of helium and hydrogen. After the 1968 Brookhaven Summer Study on superconductivity [30], it was realized that if intrinsic stability [33] and dynamic stability [34], one didn't need large quantities of helium to ensure cryogenic stability [35]. M. Morpurgo at CERN proposed using supercritical helium to cool magnet coils. He built a magnet that used supercritical helium as a coolant [36]. This author tested a magnet that could be cooled using supercritical helium or two-phase helium [37]. The result of the test was that two-phase cooling led to lower cooling temperatures than supercritical cooling. The problem is how does one prevent erratic behavior in a flow circuit with both liquid and gas in the pipe [38]. This problem was solved by having a heat exchanger in a helium tank that is connected to the helium refrigerator and the helium line coming back from the magnet being cooled [39]. The two-phase helium leaving the J-T valve (or wet expander) is 40 to 60 percent gas. The gas is condensed in the heat exchanger so that it enters the cooling loop as a sub-cooled liquid. When helium comes back the helium tank it is about 40 to 60 percent gas, which is re-liquefied by the refrigerator cold box. The helium system should be built to the non-flammable gas pressure vessel code [40].

Liquid hydrogen is no longer used within particle detectors, but it can be used as a cooling fluid in the temperature range from ~15 K to ~30 K for HTS magnets. If one is worried about magnet quench protection in magnets with high stored energy, the temperature should be on the low part of the temperature range. For safety reasons one should limit the amount of liquid hydrogen in any given cooling circuit. This suggests that a superconducting magnet should be cooled by circulating hydrogen through tubes.

Hydrogen tanks and tubes must be built to the flammable gas pressure vessel code [41] and the cryostat vacuum vessel must vent to hydrogen disposal system in the event of a hydrogen tube leak or a rupture. This author thinks that the use of liquid hydrogen should not be permitted in long tunnels.

Between 1977 and 1990, six thin detector magnets were built so that high energy particles could be detected outside the magnet cryostat with the magnetic flux being returned by iron outside of the outer detectors. Two magnets were built with high current density coils wound on an 1100 aluminum mandrel that was used for quench protection [42]. Four of these magnets were wound with a conductor with a Nb-Ti cable co-extruded in ultrapure aluminum [43]. One magnet coil was wound on the outside of an aluminum mandrel and three coils were wound on the inside of the aluminum mandrel.

All six of these magnets were cooled using two phase helium (about 50 percent gas) from a J-T valve and then entering a heat exchanger within a tank of liquid helium at 4.3 to 4.4 K. The heat exchanger changed the two-phase helium to sub-cooled liquid helium. The helium entered tubes around the magnet as subcooled liquid and returning to the helium tank as ~50 percent liquid mixed with helium gas. As a result, pressure drops are much lower in the cooling circuit. A liquid helium pump could also be used to circulate two-phase helium, but the pump work heat must be removed from the liquid before it goes to the device being cooled [44]. See Figure 1 below, shows both methods of cooling a magnet. The amount of liquid helium in the cooling tubes is much less than in the cryostat for bath cooled magnets.

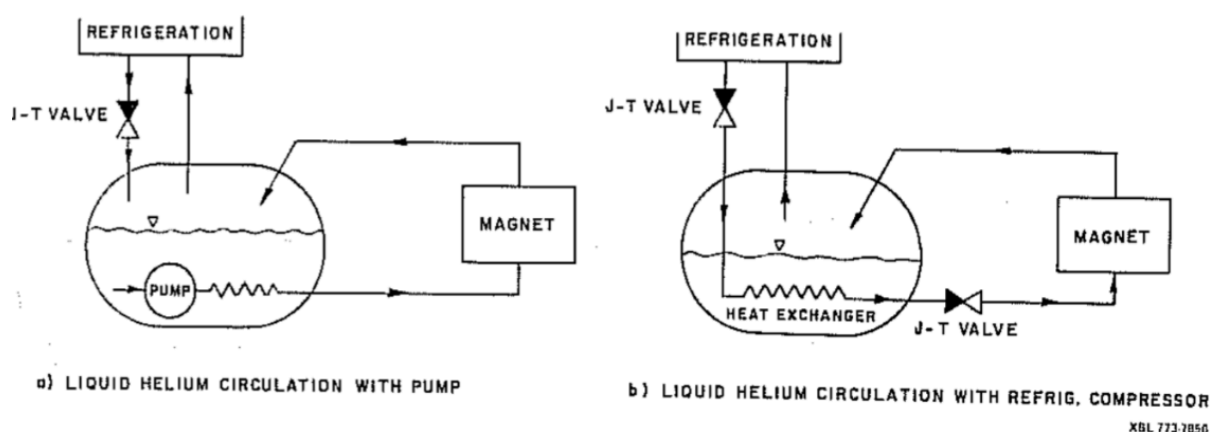


Fig. 1 Two-phase Helium cooling with a helium pump and by using the pressure from the 4 K helium refrigerator to provide the pressure to force the two-phase Helium through cooling tubes. Note, the J-T valve after the heat exchanger was not needed even for a circuit over 330 m long with 45 loops that were 2.2 m in diameter [39].

3.2 Cooling Magnets by using Coolers and Cooling Loops

The price of helium in Berkeley 1993 was ~\$1.50 per liquid liter. The price of helium in 2025 is a factor of forty higher. One should use as little liquid helium as possible. Many companies market cryogen-free magnets, which is fine if the magnet is small and if the magnetic field at the cold head is low. It is better to have the cooler cold heads some distance above the magnet being cooled. For a 4.5 K magnet with a copper strap (RRR = 130) with a current density of 2600 A m^{-2} , the ΔT in a 1 m long Cu strap is ~3.2 K. If one replaces the Cu strap with two helium filled tubes, (one with boiloff-gas going up to a cooler cold head and the other with condensed helium going down into the bath), the ΔT from the magnet to the cooler is ~0.1 K over a 1-m distance. The heat transfer rate for helium in tubes $\sim 17000 \text{ W m}^{-2}$. Figure 2 illustrates how a cooling loop where heat from a magnet goes to a cooler hold head using free convection of liquid helium and gaseous helium. From [46] it is clear that a cooling loop has better for heat transfer than a Cu strap at 4.2 K or 20.3 K. Figure 2 shows a basic two-phase cooling loop [45]. The gas rises to the cooler cold head and liquefies. The liquid drops back into cryostat. This process is use cooled down a large magnet at MSU and keep it a 4.6 K [46].

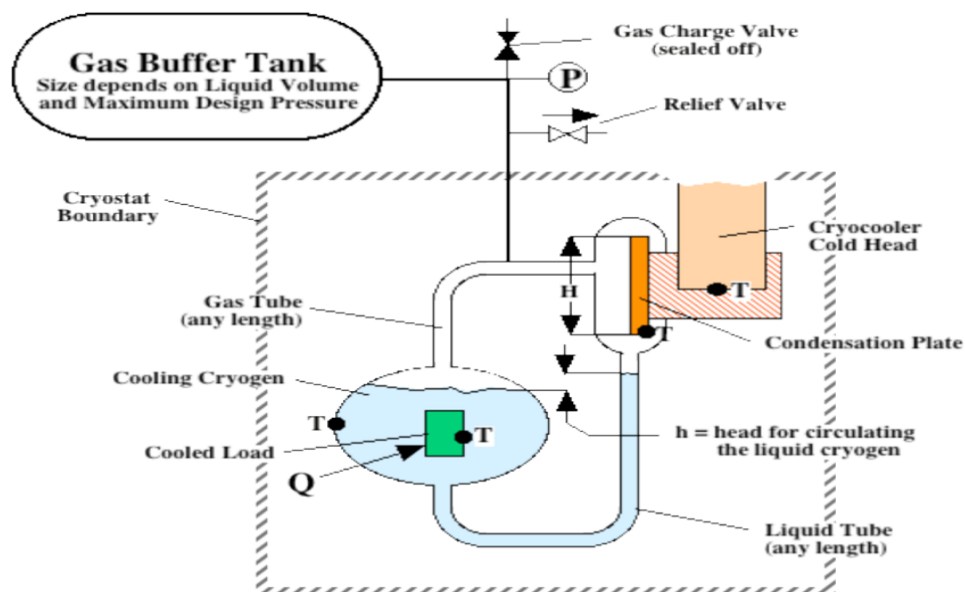


Fig. 2 A simple two-phase cooling loop for transporting heat from a cooled load by evaporation to a gas condensation plate attached a cooler cold head [45].

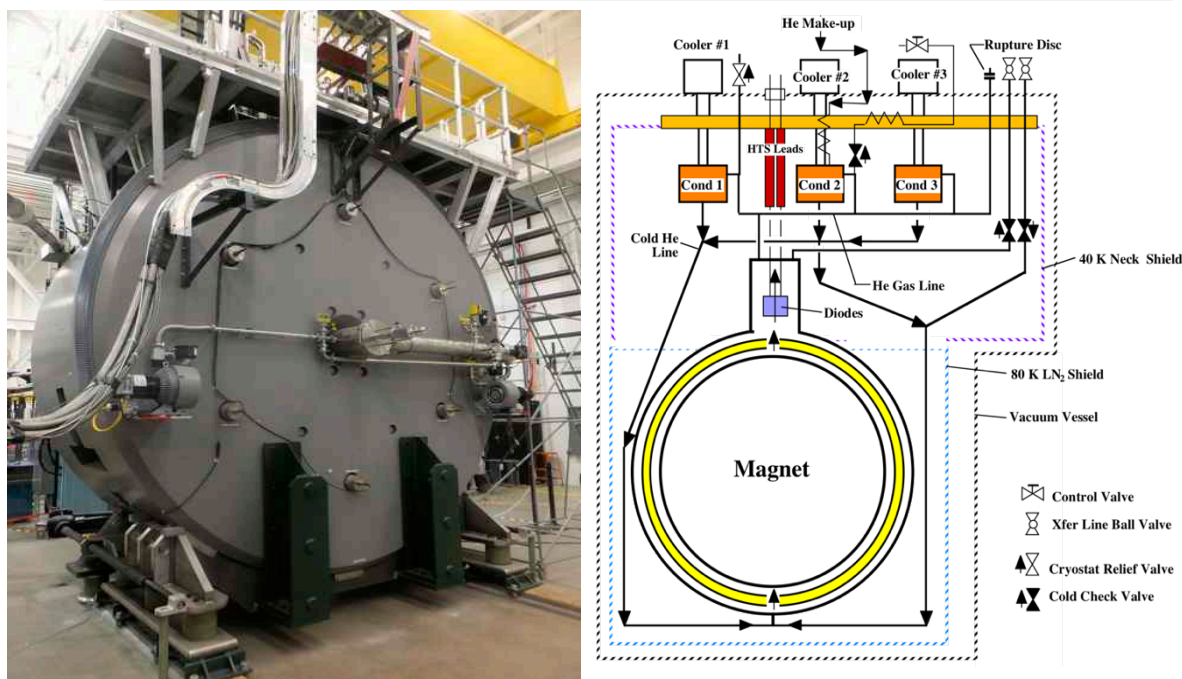


Fig. 3. Left: The MSU Cyclotron Gas Stopper Magnet [46]. Right the MSU Cyclotron Gas Stopper Magnet two-phase cooling loop the shows how three coolers cool-down a magnet coil (one of two) and liquefy helium into the cryostat with helium at ~ 4.5 K. Two coolers can keep the coil cold at ~ 4.6 K.

Other reasons for having the coolers separated from the magnets are the effects of magnetic field from the magnet; 1) on the cooler [47], 2) on the HTS leads [48], 3) the desire to keep coolers out of a high radiation zone, 4) the necessity for low frequency pulse tube coolers to have a favorable orientation, or 5) for other reasons connected with magnet use. As to coolers in magnetic fields, the cold heads must be kept at inductions < 0.5 T. Inductions > 0.04 to 0.06 T can reduce the useful life of a GM cooler. The

rotary valve on a pulsed tube cooler must be $<0.04T$ without an iron shield around the valve motor. With an iron shield around the valve motor, the induction can go up a factor of ~ 100 [46]. Pulse tube cooler's rotary valve and its motor in a remote location will have a 10 percent reduction in cooling. Compressors should be in $B < 0.02T$. HTS leads are attached between the first and second stages of a cooler. The HTS leads must be designed to carry the current where the cooler is located at the first-stage temperature [49]. Most HTS leads are affected by the field orientation. The field from the magnet must also be considered with HTS leads. High current HTS leads must be designed with a favorable self-field orientation.

When a large magnet is cooled using coolers, the shield must be made of a material with high thermal conductivity. Copper is not the best choice. This author recommends using 1100-O aluminum [49]. If the 1100-O Al shield has the same mass as the Cu shield it replaces, the shield will have a more uniform temperature distribution and the shield will be much stiffer. If the shield is coupled to the magnet magnetic field, it must have slits to reduce the circulating currents in the event of a quench. This is standard practice with any shield, but the spaces between the shield plates must be covered with a reflective tape that can't carry current.

4. Concluding Comments

Given his experience with the Michigan State Gas stopper magnet [46], this author would recommend building all LTS magnets so that the helium inventory is minimized. A large helium refrigerator can have its compressors are long distance from the cold box. The cold box can be a long way from the magnet or device being cooled. When coolers are used in place of a large refrigerator, it is often difficult to site the cooler compressors.

Some examples of where coolers should be used and where a central refrigerator is used are separated insertion devices as compared to a free-electron laser. In both cases the devices being cooled are nearly the same type of device. For a single insertion device (wiggler or undulator), it makes sense to use up to three coolers to cool it, whether it be superconducting or cold neodymium-iron magnets. When one has a free-electron laser one has many insertion devices in a row that go long distances, large refrigerators must be used, but there shouldn't be large volumes of liquid helium.

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