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A Compact Frozen Spin Refrigerator Design

D. Keller · J. Higgins

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Abstract A dilution refrigerator design is proposed that could be used as an insert to a pre-existing Oxford Instruments 1 K ^{4}He evaporation refrigerator with a 5 T magnet. This insert is designed such that all dilution components fit within the geometric constraints presented by the Oxford cryostat. This system will be used in frozen spin studies where the sample material is polarized using dynamic nuclear polarization and then cooled to well below 100 mK. The polarization can be monitored using a continuous wave nuclear magnetic resonance coil coupled to the material during the polarization process and under frozen spin mode. The dynamic polarization and measurement process sets the scale for the expected heat load of this cryostat so design characteristics are specific to the system's thermal requirements.

Keywords Frozen Spin · Dilution Refrigerator · Dynamic Nuclear Polarization · Nuclear Magnetic Resonance

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1 Introduction

The polarized solid state target [1] has been an integral part of spin dependent nuclear and particle physics experiments over the last 50 years. A polarized target system cools ($T \leq 1$K) a polarizable material while in a homogeneous high magnetic field (2.5-5 T). Photoproduction scattering experiments typically use a ^{3}He/^{4}He dilution refrigerator so that the desired target ensemble helicity can be held in a frozen-spin state ($<$ 100 mK) after the polarization enhancement from dynamic nuclear polarization (DNP). For high intensity

Contact Author: D. Keller
University of Virginia, Charlottesville, Virginia 22901
Tel.: 434-243-9955
E-mail: dustin@jlab.org

(∼100 nA) electron or proton beam scattering experiments a ^{4}He evaporation refrigerator is required in combination with a powerful pumping system. For frozen spin targets designed for beam interaction [2] a material such as irradiated NH_3 or TEMPO-doped butanol can be dynamically polarized to over 90% at 5 T and 200-300 mK. A beam of photons or low intensity (10^7 particles/sec) charge particles can be put on the target at a frozen-spin temperature of approximately 30 mK with the polarization maintained by a thin, superconducting holding coil (0.5 T) inside the target cryostat[2,3]. Such a system allows for a large kinematic coverage by relying on this small holding coil, allowing the polarizing magnet to be removed. In the frozen-spin mode, spin-lattice relaxation times as high as 4000 hours have been observed implying very slow degradation of polarization over time.

We propose a design that is optimized only for frozen spin target material characterization and will not be used in scattering experiments. This system should be capable of polarization of a sample using dynamic nuclear polarization where the nuclear polarization is monitored using continuous wave nuclear magnetic resonance (CW-NMR) measurement [4]. The system must be thermally equipped to handle solid-state RF experiments and should be loadable with frozen solid target materials in the sample holder (mixing chamber). The dilution unit will fit into a pre-existing Oxford Instruments evaporation refrigerator-magnet combination such that the system will allow quick interchange between a high power evaporation refrigerator system and a low power low temperature system. For this system there are two competing requirements in the design which have to be carefully consider, namely the maximum cooling power for the DNP process at 1 K (10-20 mW/g of material) as well as minimizing the heat load to mixing chamber during the frozen spin mode at below 100 mK.

The present work is an investigation into the geometric constraints (the Oxford Cryostat), the systems thermal requirements, the actual design of the dilution insert, and some simulations and discussion of expected performance.

2 The Oxford Cryostat

The geometric constraints of the new dilution insert are defined by the limited space in the pre-existing cryostat and magnet. The magnet is an Oxford Instruments 8 T NbTi superconducting solenoid with a bucking cancellation coil. The magnet though capable of 8 T is operated at 5 T during DNP. The magnet consists of a coil which runs at 83.80 A at 4.2 K with a field homogeneity of ±0.001%. The clear bore diameter is 77.5 mm with a current decay in persistent mode of less than 1×10^{-5} per hour. The magnet dewar holds approximately 40 L of liquid helium which is insulated with an inner-vacuum layer, a liquid nitrogen layer, and an external vacuum layer. The inner and outer vacuum layers are connected and continuously pumped during operation down to $\sim 10^{-6}$ Torr using a turbo pump system. There is also a jumper-transfer line that connects the 40 L magnet dewar to the high cooling power

evaporation refrigerator. This jumper is a vacuum insulated line that is used to continuously transfer liquid helium into the evaporation refrigerator phase separator can. The functionality of the phase separator is a consequence of continuous transfer of liquid He at low rate.

The high cooling power evaporation refrigerator consists of a set of upper heat exchangers that are cooled when vapor from the separator is pumped through, using a helium compressor pump. Liquid comes into the separator via the jumper-transfer line from the liquid helium in the magnet. The incoming helium liquid from the jumper transfer line is a mixture of helium gas and liquid depending on how much enthalpy leaks to liquid during the transfer. The phase separator contains a sintered disk that divides the separator to upper and lower halves. In the upper half the gas is pumped out at a rate that lowers the temperature of remaining liquid and separator metal to 2.5 K. The pumped cold gas is then used to cool down the upper heat exchangers. The cooled liquid in the lower part of the separator flows down to 1 K evaporator path through a series of gas-liquid heat exchangers and through the run valve that controls the flow, to the 1 K path. Below the lower heat exchangers is the cold helium reservoir which is simply a helium space made from the refrigerator shell. This whole shell is connected to a high power pump stack to pump on the liquid helium volume creating a high degree of evaporation while passing the cold helium vapor over the lower and then upper heat exchangers to optimize cooling and functionality of the refrigerator. An example of a pump stack would be a mechanical backing pump, a 350 m^3/hour and then a 1000 m^3/hour ($\sim$1 W of cooling power with this system), but the pumping system can vary depending on the goals of the project.

This Oxford system has a KF-50 mouth opening on top where the material insert goes. The insert should be of the appropriate length and design to hold the sample material below the lower heat exchangers and in the center of the homogeneous field region. There is a central opening in the heat exchangers and the separator can of 50 mm in diameter, so that the insert can sit in the center of the cryostat. This system is fitted with a manometer connected to the evaporator pump-out that reads the helium vapor pressure. This is the most accurate temperature measurement of the cold helium reservoir in the evaporation region. Other sensors are fitted to various parts of the refrigerator to monitor operation of the cryostat. A schematic of the Oxford Instruments system is shown in Fig. 1.

3 Goals and Thermal Requirments

The requirements of the design for the dilution insert are such that the mixing chamber and material sample can be held around 50 mK. The diameter of the insert should be approximately 4 cm, and the top of the dilution unit has to reside underneath the liquid level within the cryostat, allowing a total length of 150 cm. Because this is a DNP system a microwave guide to deliver the 140 GHz polarizing microwaves must pass through the central line of the insert

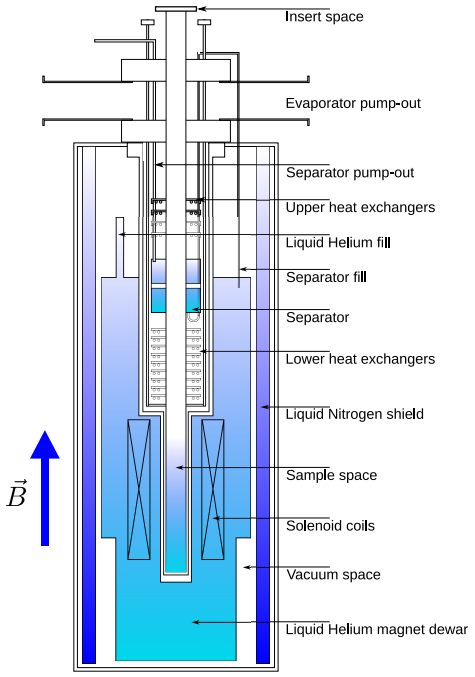


Fig. 1 The Oxford Instruments original cryostat. This diagram indicates the starting evaporation refrigerator and superconducting solenoid that the dilution unit will fit into.

to irradiate the target material in the mixing chamber area. The microwave generator is an extended interaction oscillator (EIO) tube which connects to a D-band rectangular waveguide which bends at 90 degrees and then passes into a circular waveguide delivering the microwaves through the length of the refrigerator to the sample. Though the microwave system is capable of delivering nearly 1 W of microwave power on the sample this can be attenuated to a more manageable 50 mW. The polarization of the material takes place at a much higher temperature ($\sim$1 K) so this is not a heat load that we concern ourselves with in regards to the operation of the dilution unit. However, it is necessary to make sure the waveguide does not create a heat leak down to the mixing chamber area.

At 50 mK the main heat source to mixing chamber is from the CW-NMR system. For this design the sample material does not need to be in contact with the NMR coil, allowing for the NMR measurement to probe the full volume of the target without being in thermal contact of the mixing chamber. The applied continuous RF power does produce heat through radiofrequency (RF) irradiation of the sample in the tens of μW range if configured to run with low RF power. The RF induces nuclear spin flips during polarization measurements, however the CW-NMR system used is designed to be a non-destructive measurement by running as a low constant current Q-meter which works as part of a phase sensitive circuit which responds to the changes of

the impedance in the NMR coil [4]. The RF susceptibility of the material is inductively coupled to the NMR coil as part of a series LCR circuit, tuned to the Larmor frequency of the nuclei being probed. This measurement system is expected to get better than 3% relative uncertainty in polarization during DNP [5]. The best way to deal with the heat load produced by the CW-NMR is to take infrequent polarization measurement, giving the cryogenic flows time to recover.

The advantage of mating the dilution insert with the Oxford cryostat is that the 1 K evaporation refrigerator of the Oxford system will be used as the condenser to the dilution insert. This is done by having the incoming ^{3}He-^{4}He gas mixture coming into the evaporation refrigerator already running so that the gas is pre-cooled. The insert will be constructed to regulate the temperature of this incoming gas by then passing down the separator region making thermal contact with this area and further condensing. Finally the gas is channeled through the cold helium reservoir and the 1 K plate where the main ^{4}He evaporation takes place. At this stage the 1 K helium has a two fold function in that is works as the ^{3}He-^{4}He gas condenser and a 1 K insulation layer around the dilution unit, making a cryogenic thermal shield. Modular systems with a dilution insert have been explored before [6, 7]. Naturally the challenges of these type of insert designs are dictated by the per-existing geometry and the intended application of the system.

4 Design of the Dilution Insert

In this design the entire dilution insert, shown in Fig. 2, is approximately 1.5 m in length. From the top of the insert down approximately 60 cm is the location of the separator inside the Oxford cryostat. This region of the evaporation refrigerator will be in close proximity to the copper separator cylinder on the insert design. This is meant to be a thermal barrier in both the refrigerator and the insert and is thermally regulated at around 2.5 K. The progression of the temperature gradient in the insert will be similar to the Oxford cryostat until the 1 K region. The dilution unit is expected to have a total weight of about 1 kg, which must be supported by the tensile strength of the insert frame. Below the separator cylinder, there is 50 cm for the cold helium reservoir, the lower 26 cm of which is dedicated to the dilution unit. Between the dilution unit and the separator is a heat exchanger to help pre-cool the ^{3}He as it is pumped towards the dilution unit before reaching the condenser. This pre-cooling heat exchanger ensures an adequate condensing rate of the gaseous ^{3}He when first entering the refrigerator. The design of the non-dilution components are discussed in Sec. 4.1, and the dilution unit is discussed in Sec. 4.2.

It should be noted that the steel used in the design is 316 L stainless steel (unless otherwise specified for specific components). This particular steel has a low magnetic permeability and generally has less interaction with the magnetic field generated by the superconducting solenoid. However, machining

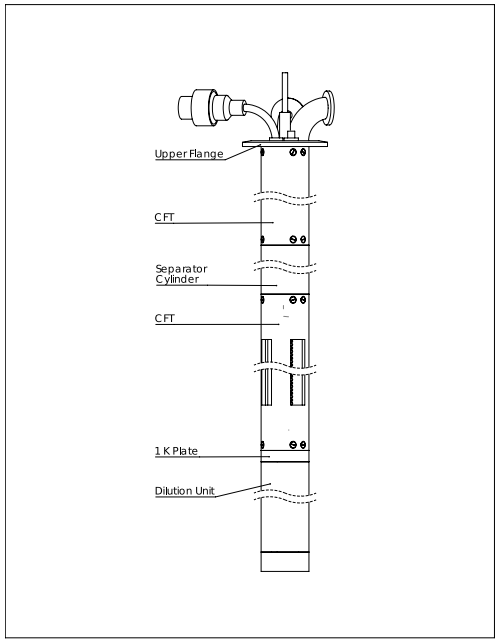


Fig. 2 The assembled outer layer of the dilution refrigerator unit that is inserted into the bore of the Oxford Instruments' evaporation refrigerator.

can change the permeability, some parts of the machined piece can have the relative permeability close to 0.1, which, for instance, in a compact mixing chamber design can have an effect on the NMR line width. In this case the stainless steel pieces should be put through magnetic annealing.

In Fig. 2 the assembled outer layer of the dilution refrigerator unit is shown. The upper flange supports the structure and can hold the rest of the insert's weight. This flange hermetically seals with a KF-50 O-ring and vacuum clamp onto the Oxford cryostat mouth. Carbon fiber tubing (CFT) mechanically connects the flange to the separator cylinder and the dilution unit.

4.1 Upper Dilution refrigerator

The top flange provides feed throughs for a NMR coaxial cable, microwave guide, inner vacuum pump tube, ^{3}He-^{4}He pump tube, the return tube for the ^{3}He-4He mixture, and instrumentation wires.

Below the insert separator cylinder and the liquid level of the ^{4}He reservoir is the 1 K plate, acting as a thermal barrier between the cooler dilution components below and the warmer pre-cooling components above. The CFT mechanically connects the separator cylinder to the upper flange, as well as the 1 K plate to the cylinder. Within this tubing runs all major lines down to the dilution unit. These major lines are thermally anchored to the phase

separator, the 1K plate, and then to the still of the dilution unit to minimize heat transfer along the lines to the low temperature part of the refrigerator unit.

4.1.1 Upper Flange

The upper flange will be made of stainless steel, 7.5 cm in diameter and 5 mm in thickness. The many feed through components in the flange are strategically arrange so that they can all fit with space to connect (Fig. 3).

On the bottom side of the upper flange (Fig. 4) is a 5.2 cm outer-diameter (OD) groove in which the 2 mm thick KF-40 O-ring will sits to make the hermetic seal. Also on the bottom is a 3.88 cm OD metal lip with six M3-threaded holes. Fitted over this lip is a carbon-fiber tube (CFT) that also has six M3-sized countersink screw holes so that machine screws of the same size and type may secure the CFT to the upper flange.

Both the ^{3}He out line and vacuum line connect to their respective pumping systems with a KF 25 vacuum clamp. The ^{3}He in line connects to a swagelok VCR metal fitting from the outside, and reduces in OD from 12 mm to 1.6 mm through the flange on the inside. The ^{3}He out line reduces in OD from 19 mm to 9.6 mm, and the vacuum line and microwave line remain 7.5 mm and 4.75 mm, respectively.

4.1.2 Cylinder/1 K Plate

A CFT connects to both the upper and lower portion of the separator cylinder in a similar fashion as the upper flange (see Fig. 5). Each line through the cylinder remains in a similar location as the upper flange.

The CFT below the separator cylinder has open channels so that the 1 K ^{4}He from the evaporator will submerge the components. In this section, the ^{3}He in line will spiral around the microwave line at the center as it descends to the 1 K plate, over a total length of 2.5 m, maximizing the pre-cooling. The CFT below the separator also connects to the top of the 1 K plate. Fig. 6 shows the features between the separator cylinder and the 1 K plate.

4.2 Dilution Unit

Below the 1K plate are the dilution components of the insert. In Fig. 7 and 8 the dilution unit is shown with certain components removed in order to reveal a more comprehensive assembly. Shown in Fig. 7 is the 1 K Plate, the graphite connection between the 1 K and the still, the main heat exchanger, and the mixing chamber. As can be seen, the main heat exchanger spirals around the microwave line and NMR line that penetrates down the middle of the insert, terminating just above the mixing chamber. Shown in Fig. 8 is the graphite support between still and mixing chamber. The support between the still and mixing chamber connects to the bottom of the still in the same manner as the

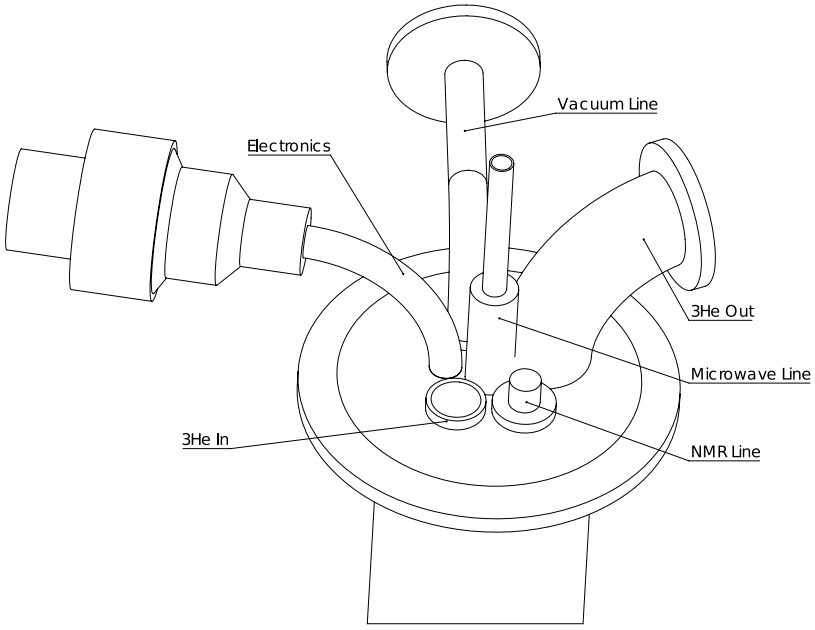


Fig. 3 Top-angle view of the upper flange. This view shows the various components that run through the upper flange.

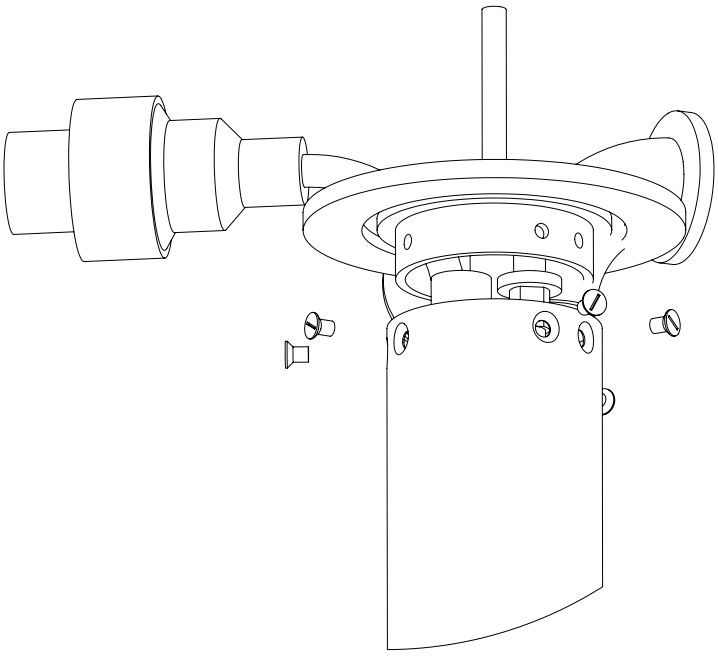


Fig. 4 In this view, components are magnified to show how the insert is assembled. The CFT fits over a lip on the upper flange. Sunken M3-sized machine screws are connected through the holes on the CFT to the threaded holes of the upper flange, securing both components in place.

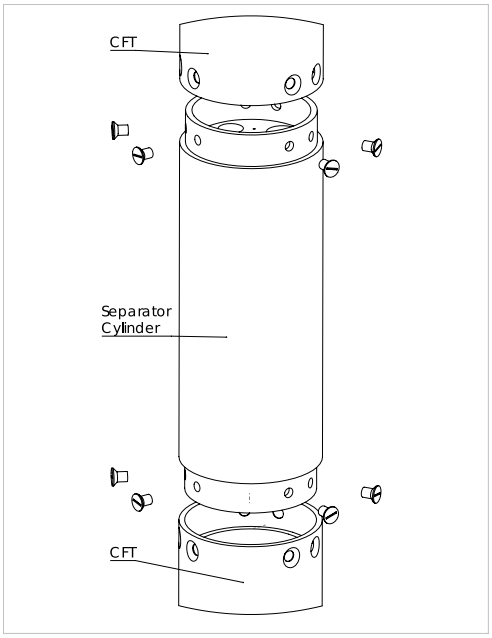


Fig. 5 View of the CFT's fitting to the separator cylinder (inner piping is hidden for clarity).

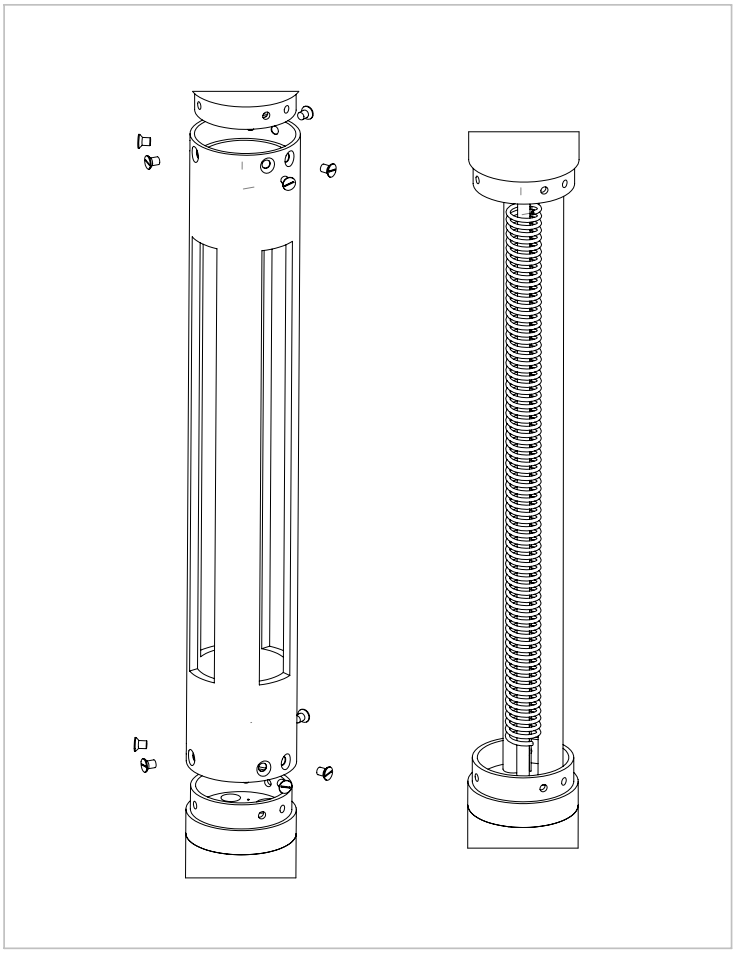


Fig. 6 View of the region between the separator cylinder and the 1 K plate, with and without the CFT (left and right, respectively). In the left image, the open channels of the CFT may be seen. This is to allow 1 K ^{4}He from the evaporator to cool the inner tubing. In the right image, a spiraling ^{3}He in line is shown. This increase in tube length helps pre-cool and condense the incoming ^{3}He. Also visible is the microwave guide running straight down the center inside the spiraling ^{3}He in line.

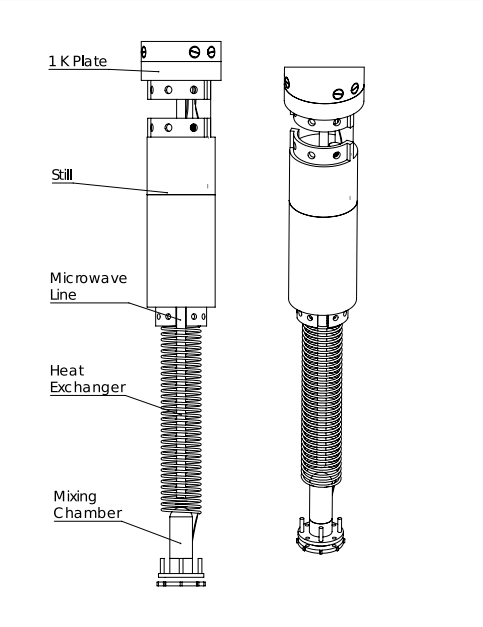


Fig. 7 Dilution unit of the proposed insert. Shown are the 1 K Plate, the graphite connection between the 1 K and the still, the main heat exchangers, and the mixing chamber.

CTF's above the dilution unit (see Sec. 4.1.1). Sections are cut out of the tube so that a vacuum may also thermally isolate the heat exchangers.

A vacuum jacket fits around the inner components of the refrigerator, making no direct contact. The design of this jacket is described in Sec. 4.2.1. Inside the vacuum jacket a vacuum is maintained to thermally isolate the low temperature inner components of the refrigerator, preventing heat leaks from affecting performance. This vacuum is established by the vacuum line that runs down from the upper flange and terminates at the 1 K plate (see Fig. 9).

A graphite support connects the still to the 1 K plate, while the main graphite support connects the still to the mixing chamber. The main heat exchanger is a tube-in-tube, counter-current continuous heat exchanger, the design of which is discussed in Sec. 4.2.6. The microwave line and NMR line penetrates the 1 K plate and still, and travels down toward the mixing chamber. As Fig. 13 shows the upper part of the mixing chamber where the polarized material is, is made from glass. The advantage of using glass is that the microwave guide and NMR coaxial cable do not need feed throughs on mixing chamber, they can be terminated outside since microwaves and NMR RF field penetrate the glass. From the top of the 1 K plate to the bottom of the vacuum jacket, the dilution unit is 26.3 cm in length with a diameter of 4.22 cm (including the vacuum jacket).

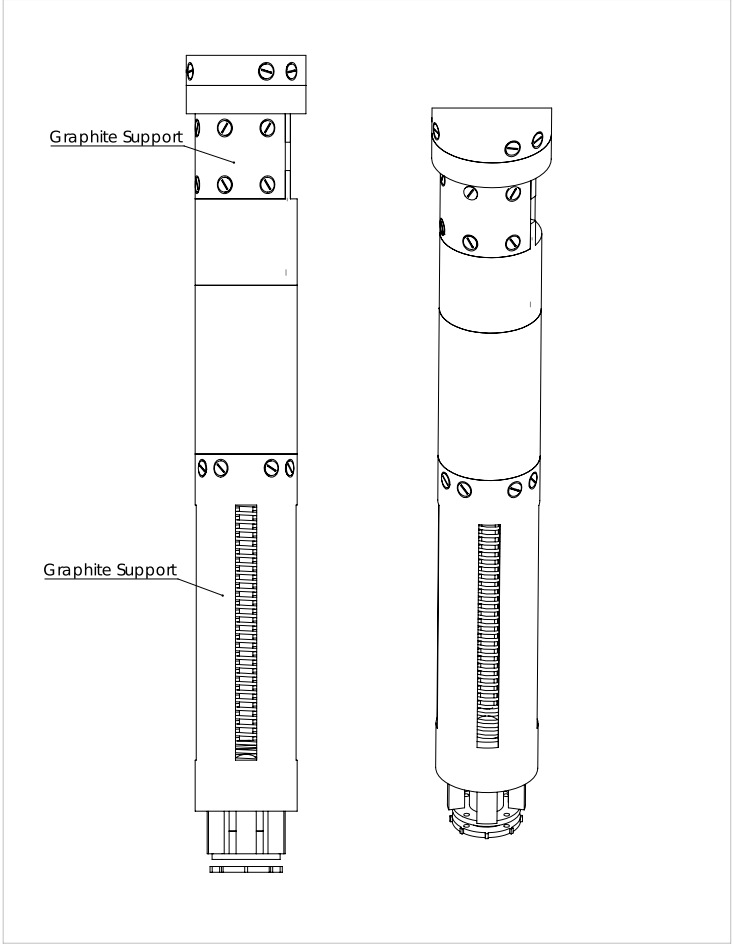


Fig. 8 Dilution unit of the proposed insert, including the graphite support between still and mixing chamber.

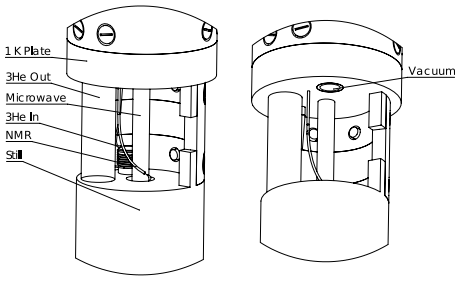


Fig. 9 View of the area between the 1 K plate and the still.

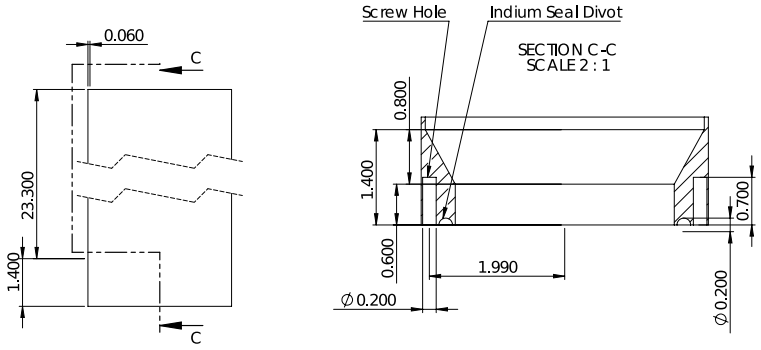


Fig. 10 Main vacuum jacket, surrounding the inner components of the dilution refrigerator.

4.2.1 Vacuum Jacket

The vacuum jacket that encases the components of the dilution unit is stainless steel, and is comprised of two parts. The first is noted as the main jacket, attached at the 1 K plate and surrounding all of the components (see Fig. 10). It is 0.6 mm in thickness (enough to withstand a 2 atm pressure differential without buckling) and is separated from the inner components by at least 2.5 mm at any point along its height. The vacuum jacket is fixed in place by using mounting screws to the 1 K plate and then hermeticly sealed to the surface using a soft solder joint. Using solder here allows for easy removal of the jacket if necessary and makes assembly easier.

The second is the vacuum jacket cap at the bottom of the dilution unit. While the main jacket is attached to the 1 K plate, the vacuum jacket cap is completely detachable, allowing easy access to the mixing chamber when loading/unloading samples.

Where the cap attaches to the main jacket, a groove has been cut for the indium wire seal 10. The indium provides a hermetic seal between the two parts, supporting the high-vacuum environment within the vacuum jacket.

4.2.2 Pre-cooler/Condensor & Main Impedance

As mentioned before, the incoming ^{3}He runs through about 2.5 m of tubing held at 1 K in order to pre-cool/condense the ^{3}He as it approaches the dilution unit (Sec. 4.1.2). In order for ^{3}He to condense, the gas must be held at a high-enough pressure. For a pre-cooler held at about 1 K, ^{3}He can condense in a pressure range of 30-270 mbar [8]. The main impedance immediately follows the pre-cooler on the ^{3}He in line, and is required for both creating the pressure needed to condense and controlling the molar flow rate. The molar flow rate (the rate at which liquid ^{3}He leaves the condenser) must be less than the rate at which the system condenses for functioning continuous circulation [9].

The ^{3}He travels through the series of heat exchangers in which the ^{3}He is cooled by the returning mixture. This returning mixture is the dilute phase between the between mixing chamber and still. The heat exchangers are common tube-in-tube continuous counter-current heat exchangers [9]. Construction simply consists of fitting one smaller tube inside of a larger tube. Liquid ^{3}He is sent downward through the inner tube while the cooler dilute phase fills the outer tube space.

To see the importance of the heat exchanger, we need to look at the enthalpy balance in the mixing chamber where cooling is produced by the transfer of $\dot{n_3}$, number of ^{3}He moles per time unit from the concentrated phase to the diluted phase. The produced cooling power will be used to cooldown the warmer liquid ^{3}He coming from the heat exchanger and to cool an experiment. The enthalpy balance can be written as Eq. 1,

$$\dot{Q} = \dot{n_3}(96T_M^2 - 12T_N^2). \tag{1}$$

Where T_M is the temperature of the mixing chamber and T_N is the temperature of the returning concentrating ^{3}He from heat exchanger.

In the case of no cooling power, or $\dot{Q} = 0$, the condition for *any* cooling to take place is $T_N/T_M \leq 3$. This stresses the importance of effective heat exchange, as a dilution refrigerator whose mixing chamber has a temperature 50 mK must cool its incoming ^{3}He from 0.7 K down to at least 150 mK to have any cooling power left for cooling the experiment.

Exiting the series of heat exchangers above, ^{3}He then enters into the mixing chamber. Here, the incoming ^{3}He is sent to the concentrated phase, sitting atop the dilute phase. By an osmotic pressure gradient created at the still, ^{3}He in the dilute side is moved away from the phase boundary. This in turn lowers the local binding energy of the dilute phase at the phase boundary, inviting more ^{3}He atoms to leave the concentrated phase and transition into the dilute phase, producing the desired dilution cooling [9].

4.2.3 Still

The still is a simple stainless steel cylinder that interfaces with the ^{3}He pump-out and serves as a reservoir for the dilute-phase towards which the circulating ^{3}He travels. The pre-cooled ^{3}He comes into the still in a separate isolated line which passes through a thin tube continuous heat exchanger immersed in the liquid of the still. This He^3 heat exchanger is made [10] of cupronickel tubing of 0.4 mm OD and 0.08 thickness, with a total length of 150 cm submerged in the liquid. This length of tubing can be easily contained within the still space. During operation the still would be held at about 0.7 K. The length of the submerged tube is chosen so that the ^{3}He can cool to this temperature.

The ^{3}He should then leave the still and move into a secondary impedance to maintain a suitable pressure in the still to retard the ^{3}He evaporation. Under normal dilution operation osmotic pressure pushes the ^{3}He from the mixing chamber, through the heat exchangers and eventually back to the still. When

the mixture is optimized and the still holds the minimal required volume of liquid the ^{3}He rests in the dilute phase.

The entire volume of the still is not dedicated to the dilute phase as the volume also contains ^{3}He/^{4}He vapor and the liquid level should remain below the film-suppressor. Connected to the top of the still is a tube that leads directly outside the refrigerator to the sealed ^{3}He pump system used to remove the ^{3}He/^{4}He vapor. As more ^{3}He turns back into vapor, less is left in the dilute phase at the vapor-liquid boundary. This difference in ^{3}He concentration from the mixing chamber to the still is what creates the osmotic pressure difference that drives the ^{3}He throughout the entire dilute phase.

The vapor pressure must be high enough to allow a reasonable flow rate of ^{3}He through the refrigerator, which is achieved at temperatures higher than what the still naturally settles to in steady-state operation. For this reason a still heater is used to generate greater vapor pressure. The still heater will be a set of heating coils fixed inside the still. Increasing the temperature in the still also increases the partial pressure of ^{4}He. In practice, it is beneficial to the performance of the dilution refrigerator to reduce the amount of ^{4}He circulating with the ^{3}He to minimize the temperature gradient instabilities, as well as the heat load to the heat exchangers. The challenge, then, is to heat the still to a temperature leading to an optimized total pressure while at the same time minimizing the partial pressure of the ^{4}He. Generally this temperature has been found to be the aforementioned $\sim$0.7 K [11].

In this design of the still there is a hollowed space down the center of the cylinder, creating an annular space for the dilute ^{3}He (Fig. 11). This allows the microwave and NMR line to run down to the mixing chamber without having to pass in through the still. The still has an OD of 3.6 cm, and inner diameter ID of 8 mm, with 0.5 mm thick walls. Both the ^{3}He out-line and the ^{3}He in-line are connected to the annular space of the cylinder through the top of the still (see Fig. 9).

In designing the still we keep two main points in mind [12]. First, the free surface area of the liquid on the dilute side must be large enough to facilitate an appropriate amount of evaporation. This is especially concerning if the free surface area is not something controllable due to the size constraints. Given a liquid surface of 4 cm in diameter, the resulting surface area would then be 13 cm^2 leading to a molar flow rate greater than 10^{-4} moles/sec. Secondly, the still must also prevent the circulation of ^{4}He, which tends to crawl up the surfaces of the container in the form of a super-fluid film. This is prevented by the inclusion of a super-fluid film suppressor (see Fig. 11), which is simply a sharp edge over which the superfluid passes over in a thin layer, limiting the overall mass flow of ^{4}He up the will walls. Two sharp edges must be included, since the annular space in which the dilute side sits has two inner surfaces.

4.2.4 Secondary Impedance

As the ^{3}He passes through the heat exchangers within the still, the pressure at which it passes should remain considerably above vapor pressure, lest any

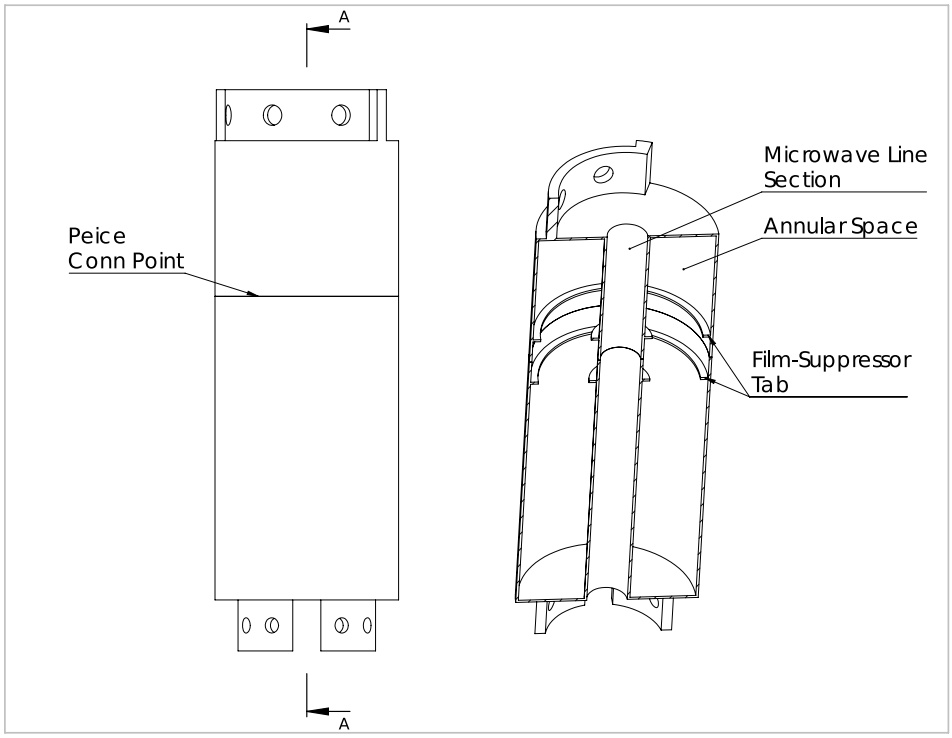


Fig. 11 Cross-section of the still, showing the section for the microwave line and the annular space for the dilute liquid. Also shown are the tabs on which the film suppressors are supported within the still, as well as the location of the connection between the two still pieces.

^{3}He re-evaporate and add a greater heat load to the system. To avoid this, it is common practice to add a secondary impedance immediately below the still, allowing pressure in the still to remain above vapor pressure. It is important to place the secondary impedance in the highest possible temperature ranges to prevent any viscous heating effects.

The pressure drop across the impedance itself should be low, to lessen any heating effects due to the isenthalpic expansion of the liquid ^{3}He. This rise in temperature is approximately proportional to the molar volume of the ^{3}He and inversely proportional to the molar specific heat: $(\partial T/\partial P)_H \approx -v_3/c_p =$ -1 mK/mbar at 0.7 K [9]. This value increases substantially the lower the temperature. With a pressure drop of around 13 mbar, this conservatively allows the temperature in the still to be as high as 1 K before re-evaporation occurs, while raising the temperature of the concentrated phase an approximate 13 mK.

To create this impedance a short length of bare manganin wire can be inserted into the ^{3}He-carrying cupronickel tubing restricting the line. Before

final assembly, the impedance value can be tested and adjusted to allow for the desired pressure drop for the correct molar flow rate.

4.2.5 Main Graphite Support

Connecting the still and the mixing chamber is a tube of length 12.6 cm with 3.6 cm OD and 2.75 cm ID. This tube fits around the bottom of the still and attaches via eight M3 machine screws. This design of the support provides stability and low thermal contact as it both secures the mixing chamber and allows room for the NMR and microwave line at the center of the unit.

The material chosen for this main support is pitch-bonded graphite [13]. This type of graphite is uniquely useful for our purposes as it is a good thermal conductor at higher (room) temperatures and becomes a poor thermal conductor at lower (cryogenic) temperatures, i.e. it acts as passive heat switch. This allows quick-cooling when the dilution process first begins and the innards are still room temperature, and prevents a big heat load to the mixing chamber during steady-state operation.

4.2.6 Heat Exchangers

The main heat exchanger of the proposed dilution insert is a tube-in-tube, counter-current continuous heat exchanger 200 cm in length and comprised of a 70% copper and 30% nickel alloy. The outer tube has an OD of 1.19 mm and ID of 1.03 mm, while the inner tube has an OD of 0.4 mm and ID of 0.24 mm. This type of heat exchanger is both very effective and compact. Despite a length of 200 cm, a coiling of 2 cm diameter with adequate spacing in between each coil reduces this length into about 10 cm in height, which may easily fit into the insert.

4.2.7 Mixing Chamber

The upper part of the mixing chamber, is made from glass as explained above. The chamber is made of 1.8 mm glass wall which is sealed to a copper housing using a low temperature epoxy. As previously described, the glass window is used so that the NMR RF and the microwaves for DNP can pass through. The mixing chamber is secured to the graphite support via fitted pegs that are wedged into peg holes. Fig. 13 shows both the pegs on the mixing chamber as well as the peg holes of the graphite support. In this figure one can see the copper mixing chamber as well as the glass window and the peg holds located on the graphite support. This design for the mixing chamber has an inner volume of 1 cm^3.

Similar to the cap of the vacuum jacket, the mixing chamber cap has a groove for an indium seal, as well as 2 mm diameter holes for M2 screws. Samples are loaded and unloaded by removing this cap, accessing inside the mixing chamber. It is desired that the torque from screwing (unscrewing) these screws in the mixing chamber cap won't damage the graphite support to which

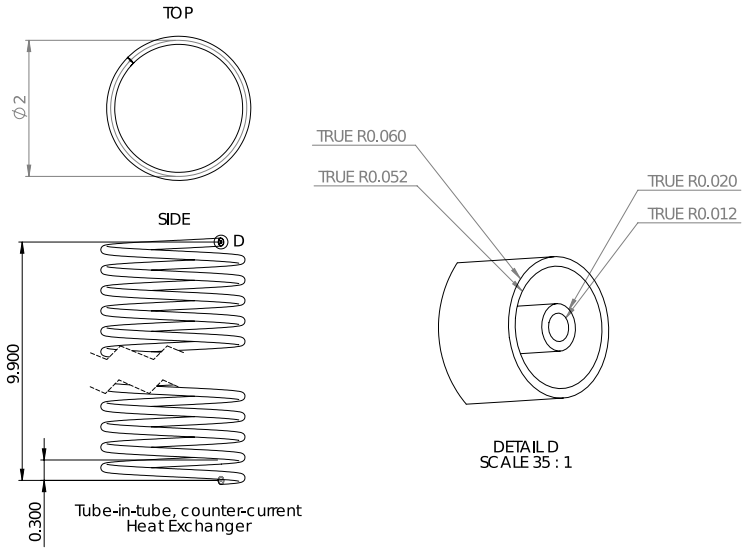


Fig. 12 Main heat exchanger for the proposed dilution unit.

it is attached. Our design allows for the insert itself to be secured upside-down, so that the mixing chamber bottom is pointing up while the end cap is held down by a special "keyed" table so when torque is applied to tighten (loosen) the screws, that torque will be transferred to the table, and not the insert. In Fig. 13 this key is shown in the form of small tabs on the cap, that would fit into a supporting table for assembly (disassembly). Shown in Fig. 14 is the full dilution unit and the support structure that holds the mixing chamber.

5 Design Analysis

Three factors are essential to the dilution unit operation: the heat load of the mixing chamber, heat exchanger analysis, and the pressure profile for the circulating ^{3}He. While some design parameters are adjustable, these specifically must be well understood to confirm the functionality, and aid in the realization, of the dilution unit.

5.1 Heat Load of the Mixing Chamber

The heat load of the mixing chamber is extremely important to the operation of the dilution unit, as it is one of the factors that determine the minimum temperature achievable. This heat load comes in three different varieties. First, the mixing chamber must be initially cooled down from room temperature to 1 K, the temperature of the surrounding liquid Helium. Since the mixing chamber will initially contain a sample that needs to be kept at cryogenic

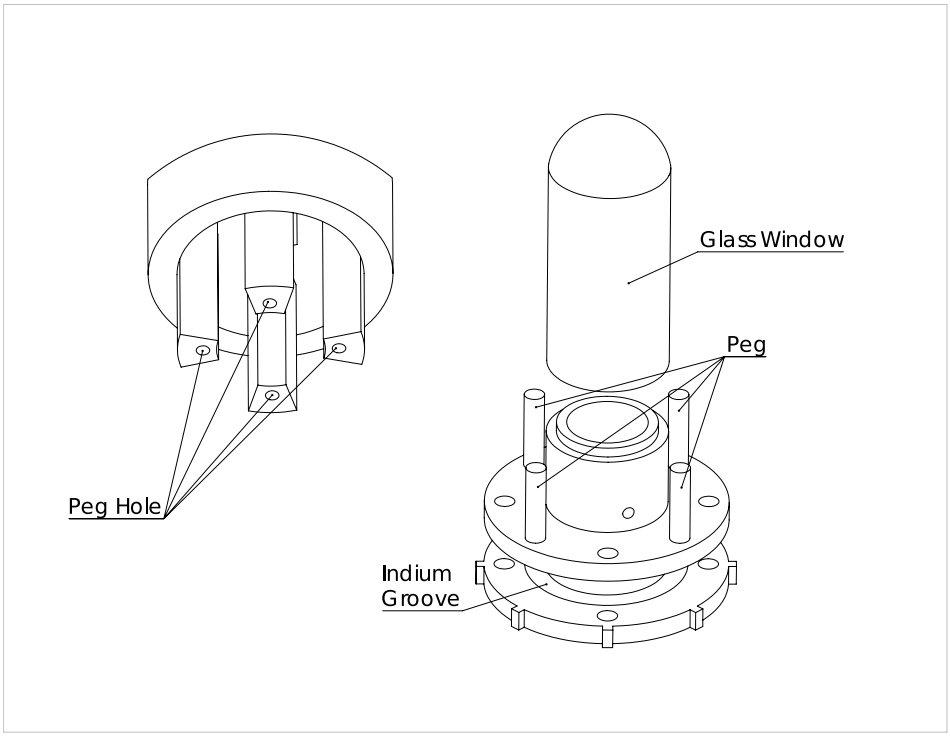


Fig. 13 View of the mixing chamber (right) as well as the peg holds located on the graphite support (left). The mixing chamber will be made of copper and have an inner volume of about 1 cm^3.

temperatures, Sec. 5.1.1 is thus concerned with how much estimated time is need to cool down the metal of the mixing chamber. Sec. 5.1.2 looks at expected heat leaks to the mixing chamber during regular operation of the dilution refrigerator.

5.1.1 Initial Heat Load

When the dilution unit is first inserted into the cryostat, the components of the refrigerator are first cooled down by the surrounding 1 K ^{4}He liquid. More specifically, heat is transferred from the innards of the dilution unit towards the 1 K plate and into the surrounding ^{4}He being pumped on. An approximation of the time needed for this initial cooldown is important since the sample placed within the mixing chamber must remain at cryogenic temperatures. The samples used in solid polarized DNP experiments are usually stored in liquid nitrogen. For the preservation of the paramagnetic centers its necessary to keep this material cryogenically cooled at all times (i.e. lower than 120 K for chemically doped materials, and lower than 105 K for irradiated ammonia). The inner parts of the dilution unit must cool at such a rate so that the mixing chamber stays below this once the sample is loaded.

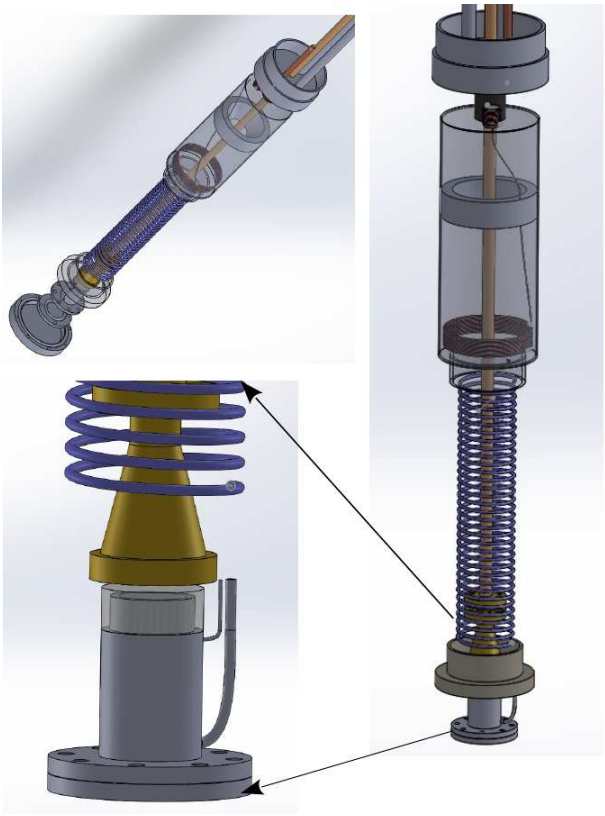


Fig. 14 Full dilution unit and the support structure that hold the mixing chamber as well as the main heat exchanger.

A simulation was developed to predict an approximate cooldown time. The innards of the refrigerator were treated as a one-dimensional bar with several sections of differing material, corresponding to the different components of the refrigerator. A summary of the simulated parts in order from top to bottom is as followed: 1 K plate (aluminum, 1 cm), graphite support (pitch-bonded graphite, 1.5 cm), still (aluminum, 9 cm), main graphite support (pitch-bonded graphite, 12 cm) and mixing chamber (copper, 3.2 cm). The simulated 1D bar was comprised of these subsections, spatially ordered as they were listed, with a total length of 26.7 cm.

The 1D heat equation is used to determine how heat is dynamically transferred. The temperature T of the 1D bar is determined by the thermal conductance k, density ρ and specific heat c, such that,

$$\rho c \frac{\partial T}{dt} = k \frac{\partial^2 T}{\partial x^2} + \frac{dk}{dT} \left(\frac{\partial T}{\partial x} \right)^2 . \tag{2}$$

Numerically solutions to Eq. 2 were generated over both time and space for given starting and boundary conditions. Information was obtained on ther-

mal conductivity, heat capacity and density for copper, aluminum and pitch-bonded graphite to help accurately model the thermal properties of the bar [14] [15] [16] [17] [18] [19]. The entire bar was initially set to a temperature of 300 K with one end in contact with a reservoir of temperature 1 K representing the point at which the 1 K plate touches the 1 K ^{4}He.

Fig. 15 shows the temperature profile of the dilution innards after ten minutes of cooling. One prominent feature that may be noted is that the refrigerator begins cooling very quickly after first contact. Within four minutes the mixing chamber cools below 200 K. The 1 K plate cools very quickly, becoming 1 K almost immediately. The rest of the refrigerator fails to cool this quickly due to the graphite support between the 1 K plate and the still, and prevents these lower components from cooling more than approximately 90 K, even after several hours. The reason for this is due to the graphite support acting as a passive heat switch for the refrigerator. The simulation highlights the fact that when the graphite support connecting the 1 K plate and the still reaches a certain low temperature during the cooldown, further cooling is halted due to this specific feature. For the purposes of thermally isolating the mixing chamber this is very good. But in the interest of preventing this slowing down of the cooling the inner vacuum space can be pressurize with helium gas. This exchange gas at about 40 mbar can be used to more quickly ($\sim$ 30 min.) bring the temperature down by thermally connecting the 1 K plate with the dilution components. After cooling the exchange gas must then be pumped out to re-establish the insulating vacuum. Fortunately, the simulation indicates that passive cooling alone is enough to keep the sample undamaged. In practice, there should be several minutes where the material in the mixing chamber is accompanied by liquid nitrogen, to give time for re-assembly and cooldown initialization.

It is worth noting that in this model it has been assumed that the 1 K helium has already reach steady-state operation temperature in the evaporation refrigerator after the insert has been loaded. There is considerable liquid helium volume (15 L) in the empty evaporation refrigerator when over filled. Over filling the refrigerator with cooled liquid helium gives considerably fast cooling during loading even when loading the insert is done quite slowly. During the insertion process the 1 K plate and dilution unit components under the 1 K plate can be only at a minimum of 77 K when cooled with liquid nitrogen. The insertion of the 77 K unit into the bore of the evaporation refrigerator leads to thermal shock to helium bath leading to a large volume of the overfilled helium reservoir in the refrigerator to be vaporized. However enough liquid helium is estimate to be left in the reservoir that the pumps and refrigerator can start running immediately, reducing the 1 K plate and vacuum jacket to around 1 K within the fist several minutes. From this stage the simulation give an estimate of the cooling time.

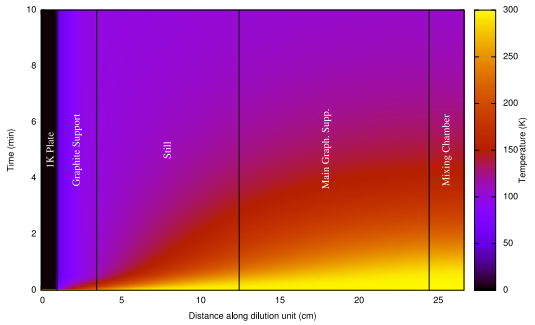


Fig. 15 Temperature profile of the dilution refrigerator sections from our simulation. The left y-axis indicates the time duration and the color scheme indicates the temperature. The diagram show a duration of ten minutes of the cooldown. Each portion of the dilution unit is marked on the figure.

5.1.2 Continuous Heat Leak

During continuous operation, heat leaks to the mixing chamber balance the cooling capacity of the refrigerator as it reaches its minimum temperature. The inner vacuum surrounding the dilution unit is designed to thermally isolate the low temperature components so that the main heat leak present during continuous operation is the heat transferred through the main graphite support. Using recent data on pitch-bonded graphite [18], the graphite tube with dimensions given in Sec. 4.2.5 would transmit approximately 25 nW of conductive heat from a 0.7 K still to a 50 mK mixing chamber. This is the largest continuous load as the design limit the material connecting to the mixing chamber. There would also be small contributions from thermal radiation and thermal leaks from the limitation in vacuum. The estimated heat load is more than manageable for the proposed refrigerator (see next section).

5.2 Heat Exchanger Analysis

Much analysis has been done on continuous heat exchangers, both numerically and experimentally. A "good" continuous heat exchanger [9] is one in which the heat transferred along the heat exchanger is small compared to the total heat transferred. In quantitative terms, a ratio can be defined in terms of the thermal conductance κ, cross-sectional area A, total length L, molar flow rate $\dot{n}_3$ and molar specific heat c:

$$Y_i = \frac{\kappa_i A_i}{L_i \dot{n}_3 c_j}. \tag{3}$$

The subscript i may be c, d or b, representing the column of liquid ^{3}He contained within the inner tube, the column of dilute ^{4}He in the annular space, and the body of the heat exchanger itself.

The total Y is the sum of these three ratios, and in order to be a suitable heat exchanger must be small in magnitude:

$$Y = \sum_i Y_i = \sum_i \frac{\kappa_i A_i}{L_i \dot{n}_3 c_j} \ll 1. \tag{4}$$

Once this is determined it is also possible to find the resulting temperature gradient along the heat exchanger [9]. Given a infinitesimal length of exchanger dx, there will be three main avenues of heat transfer: heat conduction along the heat exchanger by the established temperature gradient between the still and mixing chamber, Kapitza conduction between the dilute phase and concentrated phase, and viscous heat effects. All three of these contribute to the total change in enthalpy occurring in each section of the heat exchanger.

For continuous heat exchanger calculations, in addition to the quantities given above, we have to define a surface area σ between the dilute and concentrated phase, a Kapitza resistance $\rho(T)$, and a bulk temperature T_b of either the dilute or the concentrated liquid and the viscosity η of the concentrated fluid, its molar volumes v_3, and its flow impedances z. The Kapitza resistance is between the fluid and the wall of the copper-nickel tube. The following coupled differential equation then gives the temperature of either the dilute or concentrated side as:

$$\underbrace{\dot{n}_3 c_i \frac{dT_i}{dx}}_{Enthalpy\ change} = \underbrace{A_i \left[\kappa_i \frac{d^2 T_i}{dx^2} + \frac{d\kappa_i}{dT_i} \left(\frac{dT_i}{dx} \right)^2 \right]}_{Heat\ conduction} - \underbrace{\frac{d\sigma_i}{dx} \int_{T_b}^{T_i} \frac{dT}{\rho_i}}_{Kapitza\ conduction} + \underbrace{\eta v_3^2 \dot{n}_3^2 \frac{dz}{dx}}_{Viscous\ Heating} . \tag{5}$$

In Sec. 4.2.6 we detail our continuous heat exchanger for this system. With only about a meter of this type of heat exchanger, a dilution refrigerator could potentially be able to obtain a low temperature of 26 mK [10]. The cupronickel tubing made of 70% copper and 30% nickel, has a significantly lower thermal conductivity than other alloys, or even other cupronickel alloys. However, research has only been done on the transport properties of cupronickel material down to 4K, nowhere near the range desired. This 4 K conductance can still be taken as the *upper limit* on the actual conductance, as thermal conductance generally falls with decreasing temperature. Thus, we use $\kappa_b \leq 0.0072$ (W/cm K) [20].

As for the c_b, this value should be best given as the molar heat capacity of the dilute side [9], which has an upper limit of approximately 75 J mol^{-1} K^{-1}.

Together, these values result in a maximum value for Y_b to be 0.005. This satisfies the conditioned placed by Eq. 4, meaning that this particular heat exchanger is ideal. The term in Eq. 5 due to heat conduction can then be ignored.

The Kapitza resistance of the concentrated side for temperatures between 0.7 K and 0.13 K can be approximated as [21] :

$$\rho_c(T_c) = \left(\frac{2.4}{T_c^4} + \frac{1.55}{T_c^3}\right) \times 10^{-5} \tag{6}$$

While at temperatures between .01 K and .13K, the following approximation is more appropriate:

$$\rho_c(T_c) = \left(\frac{20}{T_c^3}\right) \times 10^{-5} \tag{7}$$

For the dilute side, its Kapitza resistivity can be approximated over the entire temperature range with:

$$\rho_d(T_d) = \left(\frac{7}{T_d^3}\right) \times 10^{-5} \tag{8}$$

Ignoring any viscous heating effects, all change in enthalpy, then, would be the cause of the Kapitza conduction; any heat leaving the concentrated side would (ideally) go directly into the dilute side such that [21],

$$\frac{d\sigma_c}{dx}\int_{T_b}^{T_c}\frac{dT}{\rho_c} = \frac{d\sigma_d}{dx}\int_{T_d}^{T_b}\frac{dT}{\rho_d}. \tag{9}$$

This integral can be solved numerically for T_b in terms of T_c and T_d, given the Kapitza conduction values for the concentrated and dilute side.

The value of $\frac{d\sigma_i}{dx}$ for Wheatley's heat exchanger is simply $2\pi r_i$, where r_i is the radius of the inner tube that touches either the concentrated side ($r_c = 0.32$ mm) or the dilute side ($r_d = 0.4$ mm).

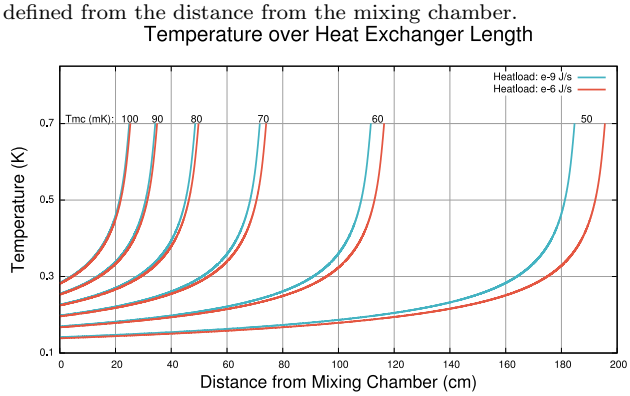
Simplifying Eq. 5 by ignoring the heat conduction and viscous heating terms, one can rearrange and reduce the expression to find the change in temperature per unit length:

$$\frac{dT_i}{dx} = -\frac{2\pi r_i}{\dot{n_3} c_i}\int_{T_b}^{T_i}\frac{dT}{\rho_i} \tag{10}$$

A simple program was made to analyze heat exchanger lengths and heat loads. Taking as input the desired mixing chamber temperature, heat load and molar flow rate, Eq. 1 was used to find the temperature on the concentrated side necessary to match those conditions. These two temperatures T_{mc} and T_c, for the dilute and concentrated side respectively, were the starting point for the analysis. From these values, Eq. 10 could be used by iteration to create a profile of the temperature as a function of distance from the mixing chamber.

Fig. 16 shows the resulting temperature profiles for several of these simulations. Keeping the flow rate of ^{3}He at a constant 10^{-4} mol/s across all simulations, mixing chamber temperatures of 100 mK down to 50 mK (as labeled on the graph) were tested against a heat load of $\dot{Q} = 10^{-9}$ and 10^{-6} W.

Fig. 16 Temperature profile for several different heat exchanger situations. Here we are showing the temperature of the concentrated phase (left y-axis) vs the length of the heat exchanger as defined from the distance from the mixing chamber.



Looking at Fig. 16, it is apparent how the Kapitza resistance becomes more of an issue at lower desired mixing chamber temperatures. For a heat load of 10^{-6} W, a mere 25 cm of continuous heat exchanger is needed to reach 100 mK, while the same heat load requires nearly 2 meters for 50 mK. This could potentially achieve a minimum mixing chamber temperature of 26 mK with a molar flow rate of 10^{-5} mol/s [10].

Fig. 16 also suggests that a heat load of 10^{-6} W or below is more than manageable for a heat exchanger of length 200 cm. Variations in heat load in this range also do not wildly impact the length of exchanger needed. For example, for a mixing chamber of 50 mK, an increase of heat load from 10^{-9} W to 10^{-6} W only requires a 10 cm extension of heat exchanger. Higher heat loads, however, become tricky. The reason for this is due to the limitations on the cooling power for a given molar flow rate. This can be seen by rearranging the change in enthalpy balance in the mixing chamber, Eq.1, finding instead the temperature of the incoming ^{3}He as a function of cooling power:

$$T_N = \sqrt{\frac{1}{12}\left(96T_M^2 - \frac{\dot{Q}}{\dot{n_3}}\right)}. \tag{11}$$

The value of T_N should be real so the sign under the radical should remain positive implying that there is a point at which it is physically impossible to obtain a desired mixing chamber temperature for a given heat load. For a molar flow rate of 10^{-4} mol/s and desired mixing chamber temperatures under 100 mK, this heat load starts to occur around tens of microwatts. Eq.11 suggests that the only solution to a higher heat load is an increase in the circulation of ^{3}He. This comes at a price, however, as the rate of temperature change along the heat exchanger, Eq.10, is lowered by the raising of molar flow rate. Indeed, the effects are not trivial: for a heat load of 1 μW and a desired mixing chamber temperature of 100 mK, a flow rate of 10^{-3} mol/s would need about 250 cm of heat exchanger - nearly ten times the length needed with

a flow rate of 10^{-4} mol/s. The strategy, then, should be to determine the minimum flow rate needed to achieve a desired mixing chamber temperature for the expected heat load, as this would also help determine the minimum length of heat exchanger needed. In high circulation rate dilution refrigerators this problem is solved by using after tube-in tube heat exchangers sintered silver or copper powder step heat exchangers that offer very large surface to volume ratio for heat exchange at lower temperatures [2].

This concern is not too prevalent in our design, as 100 nW of heat to the mixing chamber (while the microwave beam and NMR is turned off) is itself a very conservative estimation. Nonetheless, it is good to keep these things in mind for any future experimentation of the cooling power of the dilution unit.

5.3 Pressure Profile

The last task in the design of the dilution refrigerator insert is to estimate that selected components have the right flow impedances; (flow capacitance) diameter and length to support the designed flow rate. When all the flow capacitances are combined, it gives an overall flow capacity of the concentrated side. The impedance has to be dimensioned so that final flow rate is defined by the main impedance after the still. When the design flow rate is selected and the flow impedances in concentrated side calculated, then the sizing of the pumping tubes, calculation of conductances, after the still can be performed as well as conductance of pumping lines outside the refrigerator and the pumping speed to the pumps. Below is listed results of the pressure profile calculations on the concentrated side for the ^{3}He- ^{4}He mixture flow rate of 1×10^{-4} mols/s and the selected sizing of the impedances is on the order of 10^{11} cm^{-3}.

5.3.1 Pressure Gradient Along a Circular Heat Exchanger

Consider a pipe of diameter D (cm) and length L (cm), through which a fluid of molar mass M (g/mol), density ρ (g/cm^3) and viscosity η (poise) flows at a molar flow rate of $\dot{n}$. The pressure drop ΔP that occurs due to viscous effects in the liquid as it travels across the length of the pipe is then given by the Darcy-Weisbach equation [22]:

$$\Delta P = \frac{1}{2}\psi \frac{LG^2}{D\rho}. \tag{12}$$

Here, we define G as:

$$G = \frac{M\dot{n}}{\pi D^2}, \tag{13}$$

and ψ is a dimensionless factor called the *Darcy Friction Factor*, and is dependent on the physical situation within the pipe, such as the shape and material of the pipe, as well as characteristics of the material flowing through it. It

is usually dependent on the Reynolds number RE of the enclosed fluid (also dimensionless) given as,

$$RE = \frac{GD}{\eta}. \tag{14}$$

A Reynolds number of value less than 2300 corresponds to a regime of fluid flow (*laminar* flow), while RE values above this mark are considered to have *turbulent* flow [22]. Laminar flow is characterized as high momentum diffusion and low momentum convection, i.e. the fluid is more-or-less well behaved as it moves from one end of the tube to another. Turbulent flow, as the name suggests, occurs when the flowing fluid is more sporadic and chaotic in nature, with a low momentum diffusion and high momentum convection. For a circular pipe in the laminar flow regime, the Darcy Friction Factor is:

$$\psi_{lam} = \frac{64}{RE}. \tag{15}$$

The Darcy Friction Factor for turbulent flow, on the other hand, is given [22] as:

$$\psi_{turb} = 0.316(RE)^{-0.25}. \tag{16}$$

In order to use the above equations, it is necessary to determine some properties of liquid ^{3}He as it travels towards the phase boundary; namely viscosity, molar mass and molar density. The viscosity of the concentrated side as a "limiting low-temperature" value is given by [13]:

$$\eta_c = \left(2 \times 10^{-6}\right)/T^2. \tag{17}$$

The molar mass of ^{3}He is 3.02 g/mol, and its molar density can be approximated to be 0.08 g/cm^3 for temperatures of under 1 K [23].

5.3.2 Tracing the Pressure Profile

A zero pressure difference is required between the concentrated and dilute side at the phase boundary, as to keep the boundary from moving above the mixing chamber. A numerical analysis is used, incorporating the above equations to estimate the pressure experienced by the ^{3}He as it travels into the dilution unit and towards the mixing chamber. Care was taken to ensure pressures are high enough for the incoming ^{3}He to condense as it approaches the 1 K plate, as well as stay condensed inside the still (provided by a main and secondary impedance; see Sec. 4.2.2 and 4.2.4). Table 5.3.2 shows this pressure profile in terms of the pressure and pressure difference as well as the impedance value at several location in the still, the mid location within primary and secondary impedance, the heat exchangers and the mixing chamber phase boundary. The phase boundary is non-zero as this is the pressure needed from the incoming ^{3}He to counteract the pressure of the dilution mixture.

Pressure Profile of Incoming ^{3}He

Component	P (mbar)	ΔP (mbar)	Imp. (cm^{-3})
Gas Phase	67	—	—
Pri. Imp.	—	-51	3.8^{11}
Still, Upper	16	—	—
Still HE	—	(negligible)	5.83^{9}
Still, Low	16	—	—
Main. Imp.	—	-14.49	1.1^{11}
Tube-in tube, Upper	1.516	—	—
Tube-in tube	—	-0.001	1.94^{10}
Phase Bou.	1.515	—	—

Table 1 The pressure profile calculations in the concentrated side for the ^{3}He-^{4}He mixture with flow rate of 10^{-4} mol/s. The final sizing of the main impedance is about 4×10^{11} cm^{-3}, and length of the tube-in tube heat exchanger is 200 cm.

6 Conclusion

We have proposed a design for a dilution refrigerator insert to an existing Oxford cryostat to be used in experimental research. The system is designed to polarized materials using DNP and then to switch to a frozen-spin mode where the microwaves are turned off and the ^{3}He circulation begins. This new insert provides the flexibility to change the Oxford configuration from a high power evaporation refrigerator to a low temperature dilution refrigerator and back in a quick straight forward manner.

Borrowing from over fifty years of dilution refrigeration research, this compact design also attempts to maximize overall performance while remaining within the limiting geometric constraints. Two notable design aspects are the continuous heat exchanger that is compact yet reaches temperatures in our goal range, as well as the use of pitch-bonded graphite as a passive heat switch between temperature-sensitive components of the refrigerator.

The cooling power of the refrigerator is discussed, including potential sources of heat leak and heat load. Several numerical analyses are also provided to model the performance of the proposed design. The initial cooldown of the insert is simulated, ensuring that the samples inserted into the mixing chamber preserve their paramagnetic centers needed for DNP. Temperature profiles of the proposed heat exchanger were calculated for different heat loads and mixing chamber temperatures. The expected pressure profile of the dilution unit is also estimated. Our studies suggest that the design proposed can be used effectively as a frozen-spin system in a research or academic lab.

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