

Initial Exploratory Study of possible in Beam Super-cooled Helium-4 Systems

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ABSTRACT: We explore ways to maximize cooling power while minimizing temperature of a He⁴ bath in an effort to improve the polarization of a DNP target. Decreasing the target bead temperature while receiving beam can significantly improve the over all figure-of-merit of fixed target scattering experiments. We investigate the possibility of a super-cooled He⁴ refrigerator with the cooling power driven by either a He³-He⁴ dilution or a He³ evaporation refrigerator. The target material would then be placed in this super-cooled bath and held below 1 K while receiving beam. It is determined that there maybe the potential to cool to around 0.4 K with a dilution refrigerator for low beam intensity, and to about 0.6 K with a high cooling power He³ evaporation refrigerator for higher beam intensity.

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1. Introduction

The statistical and systematic significance of polarized target experiments strongly depends on how high the average polarization of the target is over the duration of beam-target interactions. The figure-of-merit for such experiments typically scales as the square of the nuclear polarization. In addition, time-dependent drifts are suppressed by the magnitude of the polarization, setting the scale of the observable that the experiment can probe. As a result, increasing the average polarization can extend our physics reach in some cases. Beam heating on a solid polarized target happens from any particle beam of interest if the intensity is great enough to cause significant cascading from pair production in the target. The solid material interfacial thermal resistance along with the over all heat load from the beam limits the beam intensity that can be used on solid state polarized target systems.

We explore the possibility of super-cooled He⁴ refrigerator designs that can maintain target material sub-cooled temperatures, while the material is being heated up by a particle beam and polarized using DNP with microwaves. There have already been many particle/nuclear experiments that use evaporation of He³ to reach material temperature below 1 K. Traditionally these system used 2.5 T magnets and 0.5 K, but a modern optimized system should combined the lowest temperatures and highest magnetic field possible for a specific experimental configuration. Polarization of ~60% have been seen in ND₃ with 2.5 T using a He³ evaporation system, but with very limited cooling potential of the target beam heating. It was shown that mixing He⁴ and He³ in this type of evaporation system significantly improves the observed polarization while in beam. It was suggested [1] that this behavior was a result of the target beads being coated with a superfluid mixture so that the relevant thermal resistance at the bead-fluid interface significantly improved.

The primary challenges of using He³ evaporation systems are the low thermal conductivity (beam cooling properties), its tendency to absorb low energy neutrons, and the difficulty in opening and replacing targets as to not lose any of this expensive gas. The latter can add extensive over-head time in the experiment. In addition there have been studies that have shown that when material beads are in He³, as the bead size decreases the material's polarization decreases as well [1]. Variation of the bead size in He⁴ show little signs of this effect even when the diameter used is much much smaller than 0.5 mm. An optimization would include a closed He³ circuit that cooled a He⁴ reservoir, taking advantage of the low temperatures provided by He³ and the beam cooling characteristics of He⁴, using optimized bead geometry. Such a system would be easy to open and refill with similar maintenance as the standard He⁴ evaporation systems used today in particle/nuclear experiments. The additional goal along with an easy quick target load and maintenance setup is to minimize the cooling time for the refrigerator. Here the goal is to be able to get back to your operating temperature as quick as possible, again trying to minimize the over-head time in the experiment.

Whether using a He³ evaporation or He³-He⁴ dilution system for cooling, the He⁴ reservoir can have a similar design in either case. The He⁴ that fills this reservoir will be precooled before entering the bath as to not heat up the subcooled helium. We also want to take advantage of the Rollin film creep which is expected to be around 2×10^{-7} L/s/cm of perimeter. The film creep provides a slow flow out of the cryostat which is pumped out as vapor with a He⁴ evaporation pump stack. This pumping configuration allows for fast He⁴ evaporation cooling to 1 K in the He⁴ reservoir after loading and unloading the insert. The supercooling system then takes over to proceed to the lower temperatures. As the He⁴ film creep warms up as it moves further away from the bath the evaporation of the film insulates the bath from the up stream end of the refrigerator and insert. Convection flow induced from this He⁴ film evaporation towards the pump-out limiting the heating from high pressure return near the supercooled bath. The precooled He⁴ inflow and the pump-out mouth (film control geometry) of the supercooled reservoir should be optimized for a particular design. Its worth noting that such a design is only applicable for a reservoir of 0.3 K and up.

In order to keep the beads as cold as possible the refrigerator and system thermodynamics must mitigate the heating in the cryostat and the target beads. So, the dilution or He³ evaporation refrigerator must be able to cool down the He⁴ bath as fast as the charged particle beam is heating it up. This means that the cooling power of the refrigerator must be equal or better to the heat load on the beads from the beam. In order to calculate the power being radiated from the beads, how the heat is generated must be considered. While the material is in the beam, lower energy electron positron pairs are produced in the target, the multiple scattering and cascade of this pair production leads to interfacial bead heating as this kinetic energy is absorbed into the target. From this, the heat load in the cryostat due to beam-target interactions can be estimated as,

$$\dot{Q}_B = \rho E I L_T \quad (1.1)$$

where \dot{Q}_B is the change in energy over time due to the charged particle beam, E is the energy per area density from the electron positron pairs productions, I is the charged particle beam current, L_T is the length of the target, and ρ is the density of the material.

The beam currents we focus on here are between 1-100 nA of continuous electron beam. The target material used is ammonia, so the energy per area density generated from the electron-positron pairs is about $2 \text{ MeV}\cdot\text{cm}^2/\text{g}$, but only about 60% of the energy remains in the target, with density of ammonia being $\rho = 0.917 \text{ g/cm}^3$ at $\sim 1 \text{ K}$.

Finally we consider the microwaves from the DNP process. Here we consider trying to attenuate the microwave so that just the level to maintain polarization is used as to minimize the heat load to the target. At lower temperatures, the polarization decays very slowly (ignoring radiation damage) and so lower intensity microwaves can be used to maintain the polarization, but at higher temperatures ($\sim 1 \text{ K}$) high intensity microwaves are preferred. The microwave can add much more than 0.2 W of power to the target material so here was project what is possible by careful minimization of microwave heat load after the initial polarization. The final calculation for the change in energy per unit time of the material is

$$\begin{aligned}\dot{Q} &= \dot{Q}_B + \dot{Q}_M \\ &= EIL_T\rho + \dot{Q}_M\end{aligned}\tag{1.2}$$

where \dot{Q} is the total change in energy per unit time and \dot{Q}_M is the power added from the microwave. This value is the final power that must be matched by the cooling power of the refrigerator. We initially consider a dilution refrigerator.

2. Kapitza Resistance

The target material we focus on in the following examples is ammonia beads. The acoustic mismatch of the target material in liquid helium is just as critical as the over all cryostat temperature with regards to polarization capacity. The heat transferred between the beads to the liquid helium, is via a phonon exchange at the thermal boundary where kinetic energy transport away from the target material occurs. However, the phonons have to be properly aligned and in phase with each other for the energy to be transferred. If the acoustics of the phonons are out of phase, then the energy is blocked from passing the boundary and no heat is transferred in this way. Because phonons are so small, at high temperatures the affect of this Kapitza resistance is practically zero, but as the temperature gets lower, the value of the Kapitza resistance increases. The Kapitza resistance effect at low temperatures is modeled by

$$\frac{\dot{Q}}{N} = AR_K(T_{bead}^4 - T_{mc}^4)\tag{2.1}$$

where N is the number of beads, A is the surface area of a bead, R_K is the Kapitza resistance of the beads, and T_{bead} and T_{mc} are the temperatures of the beads and the mixing chamber, respectively[3]. The total rate of heat change, \dot{Q} , is divided by the number of beads because this equation is only considering a single bead, and so the energy that matters is the energy per bead. At low temperatures, the Kapitza resistance is proportional to T^3 and so the Kapitza resistance of the beads can be written as

$$R_K = KT_{bead}^3\tag{2.2}$$

where K is the Kapitza resistance measured at 1 K. For ammonia, the Kapitza resistance at 1 K has been measured to be about $1.4 \times 10^{-2} \text{ W}/(\text{cm}^2\cdot\text{K}^4)$. Plugging this into the change in heat equation

above results with

$$\frac{\dot{Q}}{N} = AKT_{bead}^3(T_{bead}^4 - T_{mc}^4) \quad (2.3)$$

This equation is the one that is used to calculate the final temperature of the beads.

The above equation is solved numerically for T_{bead} given values for \dot{Q} , N , A , K . Plots of T_{bead} versus T_{mc} were created. \dot{Q} was obtained from Eq. (1.2), and A was calculated as the surface area of a sphere. The number of beads was crudely approximated by dividing the volume of the target cup that holds the beads by the volume of a cube with side length equal to that of the bead diameter. This is a very crude approximation, but for the purposes of theorizing the potential of this method, it is acceptable. The target cup used is a cylinder with a diameter of 3 cm and a length, L_T , of 3 cm as well. The radius of the beads considered was generally 0.5 mm, but the radii of 0.1 mm and 1 mm were considered in extremes to obtain the full potential of this idea.

3. Heat Exchanger Calculations

The cooling power of the dilution refrigerator must be the same as the heat being radiated by the target material, in order to keep the target material at a constant temperature. Therefore, the cooling power of the dilution refrigerator is denoted by \dot{Q} . Now, the change in energy from the dilution refrigerator is equal to the change in enthalpy from the concentrated phase to the dilute phase of helium, and the the rate of change is calculated by multiplying the change in enthalpy by the molar flow rate of He^3 . Therefore, the cooling power is calculated by

$$\dot{Q} = n_3[H(\chi, T_d) - H^c(T_c)] \quad (3.1)$$

where H is the enthalpy of the dilute phase, which is dependent on χ , the molar fraction of He^3 , and T_d , the temperature of the dilute phase[4]. $H^c(T_c)$ is the enthalpy of the He^3 concentrated phase at temperature T_c . Depending on the design of the refrigerator, the concentrated phase may have a different molar fraction of He^3 than pure He^3 because the liquid He^4 can move up the walls of the refrigerator and mix with the concentrated phase. However, the refrigerator considered in this paper is a horizontal refrigerator, and the liquid He^4 will not move horizontally, so the concentrated phase will have very little He^4 . Therefore, the approximation that $H^c(T_c)$ is the same as the enthalpy of pure He^3 at temperature T_c has been made.

The values for the enthalpy of both the dilute and concentrated phases were acquired from tables in the technical note, *Thermodynamic Properties of He^3 - He^4 Solutions with Applications to the He^3 - He^4 Dilution Refrigerator*[5]. A He^3 molar fraction of $\chi = 11\%$ was used in the calculations of this paper. Also, the enthalpy for pure He^3 was used to determine the needed temperature of the concentrated phase, and in order to get more precise temperatures, a computer programed was used to interpolate the enthalpy values from the table.

In order to calculate the length of the heat exchanger, a continuous tube-in-tube heat exchanger was considered. With this type of exchanger, the material being cooled, in this case the pure He^3 , flows down a pipe that resides in a larger pipe. Inside the larger pipe and around the smaller pipe, a cooler material, in this case the dilute He^3 phase, flows up and in the opposite direction of the inner material. Since the phases are in thermal contact with each other via the inner pipe, heat begins to transfer from the pure He^3 , which is warmer, to the colder He^3 - He^4 mixture, and this causes

the pure He³ to cool down. Mathematically, the temperature change over the length of the heat exchanger of either phase can be described by

$$A_i \left[\kappa_i \frac{d^2 T_i}{dx^2} + \frac{d\kappa_i}{dT_i} \left(\frac{dT_i}{dx} \right)^2 \right] - \frac{d\sigma_i}{dx} \int_{T_b}^{T_i} \frac{dT}{\rho_i} + \eta v_3^2 \dot{n}_3^2 \frac{dz}{dx} = \dot{n}_3 c_i \frac{dT_i}{dx} \quad (3.2)$$

In the above equation, A refers to a cross-sectional area within the exchanger, κ is the thermal conductance, σ is the surface area of the inner tube, T_b is the temperature of the body of the heat exchanger itself, ρ is the Kapitza resistance, η is the viscosity, v_3 is the molar volume, z is the impedance, and c is the molar specific heat[4]. The index i is either c if considering the concentrated phase of He³ or d if considering the dilute phase. This calculation is assuming that the heat exchanger is running in a steady state. To understand the equation, it can be unpacked term by term.

The first term on the left hand side is due to the heat conduction down the heat exchanger. Whenever there is a temperature gradient along a material, the temperature along the material will begin to change in order to reach an equilibrium temperature such that the material has a uniform temperature. One can notice that this first term is in fact just $(d/dx)[A_i \kappa_i (dT/dx)]$, which is the derivative with respect to length of the heat flow rate along the material. In an ideal heat exchanger, this term goes to zero because the heat should be flowing from one phase to the other, rather than the heat flowing down a single phase.

The second term is the change in temperature due to the Kapitza conductance (the reciprocal of the Kapitza resistance) of the phases with the heat exchanger. This is known as Kapitza conduction and it is the heat transfer that is desired in the heat exchanger. The temperature of the body of the heat exchanger is only affected by the liquid phases, which implies that any heat from the body of the exchanger that is transferred to one liquid phase must have originated in the other liquid phase.

The third term describes changes in heat as a result of viscous heating effects. Tube-in-tube heat exchangers are generally coiled around in order to slow down the fluid, which allows for a longer amount of time during which the fluids are in thermal contact. However, this can also result in the fluid sticking to the heat exchanger causing some friction which would heat up the fluid. The affect of this term is generally pretty small though and can usually be ignored.

Finally, the term on the right hand side is the change in enthalpy over the length of the heat exchanger. This gives how much the energy of the liquid has changed as it flowed through the heat exchanger.

In this paper, an ideal heat exchanger is considered, and so the heat conductance term and the viscous heating effects are ignored. This also means that the change in enthalpy of one phase should equal the opposite change in enthalpy in the other phase. In mathematical terms,

$$\frac{d\sigma_c}{dx} \int_{T_b}^{T_c} \frac{dT}{\rho_c} = \frac{d\sigma_d}{dx} \int_{T_d}^{T_b} \frac{dT}{\rho_d} \quad (3.3)$$

Given the values for each phase, this equation can be solved numerically for T_b . Since the heat exchanger is assumed to be running in a steady state, T_b is constant throughout the length of the heat exchanger. This will then be used to calculate the length of the heat exchanger.

To continue with the calculation, the values for each liquid phase in the above equation must be stated. First, because the heat exchanger is considered to be a tube-in-tube, the value for the change in surface area per unit length is just equal to the circumference of the inner tube, or $d\sigma_i/dx = 2\pi r_i$. For the calculations in this paper, the values $r_c = 1.5$ mm and $r_d = 2.0$ mm are used.

Next, when the temperature of the concentrated phase is below 0.13 K, the Kapitza resistance for the concentrated side can be described by

$$\rho_c = \left(\frac{20}{T_c^3} \right) \times 10^1 \quad (3.4)$$

If $0.13 \text{ K} < T_c < 0.7\text{K}$, then the Kapitza resistance is calculated by

$$\rho_c = \left(\frac{2.4}{T_c^4} + \frac{1.55}{T_c^3} \right) \times 10^1 \quad (3.5)$$

For the dilute side, there is one equation for the Kapitza resistance for all temperatures in the range of 0.1K to 0.7K. The equation is

$$\rho_d = \left(\frac{7}{T_d^3} \right) \times 10^1 \quad (3.6)$$

Lastly, T_d is equal to the desired temperature of the mixing chamber. This is the temperature that the dilution refrigerator is holding constant throughout an experiment. The value for T_c can then be calculated from Eq. (3.1). The enthalpy of the concentrated phase is calculated, and then the temperature is pulled from the interpolated table of values.

A program was made to conduct all the above calculations given T_d , \dot{n}_3 , and \dot{Q} , which is calculated from Eq. (1.2). The program then was able to construct a temperature versus heat exchanger length graph by using

$$\frac{dT_c}{dx} = - \frac{2\pi r_c}{\dot{n}_3 c_c} \int_{T_b}^{T_c} \frac{dT}{\rho_c} \quad (3.7)$$

iteratively. The molar specific heat of He^3 that was used was $c_c = 3 \text{ J}/(\text{mole}\cdot\text{K})$. The final length of the heat exchanger was chosen to be the length at which the temperature of the pure He^3 went from 0.8 K to the needed T_c temperature.

4. Pumps Needed for Dilution Refrigerator

In order to calculate the pump out rate needed for the dilution refrigerator, the ideal gas law is used to calculate the molar volume of the He^3 gas being pumped out, and then the molar volume is used to convert the molar flow rate to the volume flow rate. To use the ideal gas law, the pressure of the gas and the temperature must be known. Since the pumps are outside of the refrigerator, by the time the He^3 gas is getting pumped out, it is a room temperature, and $T = 300$ K. To obtain the pressure, the fact that the He^3 is coming from the still in the dilution refrigerator is used. Because of this, the pressure of the gas is equal to the vapor pressure of He^3 at the still temperature. Technically, in the still is a He^3 - He^4 mixture and the vapor pressure of He^4 would also have to be considered, but

at the low temperatures considered, the vapor pressure of He⁴ is negligible. Therefore, the volume flow rate, \dot{V} of the pumps needed can be calculated by

$$\dot{V} = \frac{RT_{room}}{P_3(T_{still})} \dot{n}_3 \quad (4.1)$$

where R is the universal gas constant, and $P_3(T_{still})$ is the He³ vapor pressure at the temperature of the still. The still in a dilution refrigerator is usually around $T_{still} = 0.75$ K.

5. He³ Evaporation Refrigerator Calculations

While a He³-He⁴ dilution refrigerator can cool down the He⁴ bath to very low temperatures, at higher temperatures like around 0.5 K, the cooling power of the dilution refrigerator is still pretty low, which makes managing of the heat load difficult. At these temperature, it more advantageous to use a He³ evaporation refrigerator, which has much higher cooling power at these temperatures. In order to examine this possibility, the parameters needed for the refrigerator must be looked at. Since the He³ evaporation refrigerator cools down by simply pumping out He³ vapor in order to make the liquid He³ evaporate faster, the parameters to calculate are the cooling power of the refrigerator and the required volume flow rate that corresponds to the desired He³ holding temperature. Because the cooling is done by evaporation, the cooling power is calculated by

$$\dot{Q} = \dot{n}_3 L_v(T) \quad (5.1)$$

where $L_v(T)$ is the latent heat of vaporization of pure He³ at the temperature T . The values for $L_v(T)$ were acquired from a table of thermodynamic properties of pure He³ in the technical note, *Thermodynamic Properties of He³-He⁴ Solutions with Applications to the He³-He⁴ Dilution Refrigerator*.

Next, the volume flow rate is calculated in the same manner as the volume flow rate for the dilution refrigerator, but instead of the vapor pressure being measured at the temperature of the still, the vapor pressure is taken to be at the temperature that the evaporation refrigerator is holding. Therefore, the volume flow rate for the evaporation refrigerator is

$$\dot{V} = \frac{RT_{room}}{P_3(T)} \dot{n}_3 \quad (5.2)$$

This is the volume flow rate needed to hold the He³ at temperature, T . However, one does not just need pumps that are large enough to pump at a rate of \dot{V} . The refrigerator must also have a tube that is big enough for the vapor to flow through it at a rate of \dot{V} as well. The maximum flow rate of a fluid through a tube is known as the tube's conductance. If the flow through the tube is assumed to be laminar flow, the conductance, L , of a tube is

$$L = 486 \frac{Pd^4}{l} \quad (5.3)$$

where P is the pressure of the gas in mbar, d is the diameter of the tube in cm, and l is the length of the tube given in cm[6]. Therefore, by setting the conductance of the tube equal to the needed volume flow rate, the necessary diameter of the tube can be solved for and calculated by

$$d = \left(\frac{\dot{V}l}{486P_3(T)} \right)^{1/4} \quad (5.4)$$

This equation gives the minimum diameter needed to have the flow rate, \dot{V} . When designing the refrigerator, this parameter must also be considered.

6. Results

Tables 1-11 present the lowest temperature that a dilution refrigerator can hold within reason at each charged particle beam intensity at varying microwave powers. The criteria for being within reason is that the molar flow rate is below 80 mmol/s and the length of the heat exchanger, L_{HE} , is less than 2 m. So, in the tables, T_{dilute} is the lowest mixing chamber temperature that can be held at the required \dot{Q} within reason.

As expected, at lower beam intensities which means lower cooling power needed the dilution refrigerator is able to hold lower temperatures than at higher beam intensities. However, just because the dilution refrigerator can hold a significantly lower temperature does not mean that the beads are held at a significantly lower temperature. Especially with an increase in the microwave power, which would be needed for polarization, the bead temperature stays around 0.6 K at the lower beam intensities, and at the higher beam intensities, the bead temperature increases up to 0.7 K. There is also some error in the calculated bead temperature which comes from the error in the given Kapitza resistance value. This error will be demonstrated and discussed when looking at the bead temperature versus bath temperature graphs. For the tables, the Kapitza resistance value used to calculate the bead temperature was $1.4 \times 10^{-2} \text{ W}/(\text{cm}^2 \cdot \text{K}^4)$ at 1 K.

These tables were generated using a bead radius of 0.5 mm and the number of beads was calculated in the method described previously. However, the only value that is dependent on the size and number of the beads is T_{bead} . All of the other values do not depend on the beads. The microwave power, \dot{Q}_M , is chosen and T_{dilute} , L_{HE} , \dot{n}_3 , depend on the total power \dot{Q} , which is given below each table. \dot{Q} is dependent on the length of the target, L_T , which can be seen in Eq. (1.2), but because it is approximated that the target is completely filled with ammonia, the number and size of the beads in a given L_T does not affect the total cooling power needed. There is some choice in the molar flow rate, and the length of the heat exchanger depends on the choice of molar flow rate. As the molar flow rate increases, the length of the heat exchanger decreases assuming the same cooling power is needed. In these tables, the smallest possible integer molar flow rate was chosen. This means that the lengths of the heat exchanger shown are going to be the longest lengths, and as a result, almost all of the lengths are more than 150 cm. A heat exchanger of this length is well within reason because one can simply wrap the heat exchanger into a coil that condense the amount of space it takes up without shortening it. Theoretically, it could be possible to shorten the heat exchanger by increasing the molar flow rate, but one must recalculate the temperature of the pure He^3 from Eq. (3.1). The problem that could arise is the fact that the final temperature, T_c , cannot be greater than the temperature of the He^3 in the still of the dilution refrigerator. The temperature of the still is generally between 0.7-0.8 K, and so by increasing the molar flow rate, one must be careful that T_c is not increasing past 0.8 K.

Lastly, the size and number of beads does not affect \dot{Q} , but the length of the target, L_T , does affect the number of beads in the target cup and in turn can affect the final temperature of the beads. This is crucial to understand because one cannot simply decrease L_T in order to decrease \dot{Q} . By shortening the target length, the number of beads in the target is decreased as well, and

Table 1. Temperature Capability of Dilution Refrigerator With Various Microwaves at $I = 1$ nA

\dot{Q}_M (W)	T_{dilute} (K)	T_{bead} (K)	L_{HE} (cm)	\dot{n}_3 (mmol/s)
0	0.1	0.3218	186	8
0.05	0.2	0.4802	193.9	38
0.1	0.3	0.5335	154.3	34
0.15	0.3	0.5628	178.2	52
0.2	0.3	0.5849	190.4	71

Table 2. Temperature Capability of Dilution Refrigerator With Various Microwaves at $I = 10$ nA

\dot{Q}_M (W)	T_{dilute} (K)	T_{bead} (K)	L_{HE} (cm)	\dot{n}_3 (mmol/s)
0	0.2	0.4491	192.3	21
0.05	0.2	0.5111	197.1	69
0.1	0.3	0.5520	189.6	44
0.15	0.3	0.5765	190.7	63
0.2	0.4	0.6081	194.1	49

Table 3. Temperature Capability of Dilution Refrigerator With Various Microwaves at $I = 20$ nA

\dot{Q}_M (W)	T_{dilute} (K)	T_{bead} (K)	L_{HE} (cm)	\dot{n}_3 (mmol/s)
0	0.2	0.4949	198.5	50
0.05	0.3	0.5419	181.8	38
0.1	0.3	0.5689	175.8	57
0.15	0.3	0.5898	192.9	76
0.2	0.4	0.6184	165.2	57

since the microwave power is not affected by the decrease in target length, the total power per bead increases. If there is no microwave, then because both the number of beads and the total power would be directly proportional to L_T , the power per bead would actually remain constant. However, the microwave is needed to polarize the beads and so must be considered. Therefore, with an increase in power per bead, the temperature of the beads would also increase, which is counterproductive to what this dilution refrigerator is designed to do. The length of the target used to create these tables was $L_T = 3$ cm.

Table 4. Temperature Capability of Dilution Refrigerator With Various Microwaves at $I = 30$ nA

\dot{Q}_M (W)	T_{dilute} (K)	T_{bead} (K)	L_{HE} (cm)	\dot{n}_3 (mmol/s)
0	0.3	0.5305	176.2	32
0.05	0.3	0.5606	189	50
0.1	0.3	0.5832	195.6	69
0.15	0.4	0.6132	169.7	53
0.2	0.4	0.6277	187.6	64

Table 5. Temperature Capability of Dilution Refrigerator With Various Microwaves at $I = 40$ nA

\dot{Q}_M (W)	T_{dilute} (K)	T_{bead} (K)	L_{HE} (cm)	\dot{n}_3 (mmol/s)
0	0.3	0.5515	176.6	44
0.05	0.3	0.5761	181.9	63
0.1	0.4	0.6078	180.4	49
0.15	0.4	0.6230	194.3	60
0.2	0.4	0.6362	174.6	72

Table 6. Temperature Capability of Dilution Refrigerator With Various Microwaves at $I = 50$ nA

\dot{Q}_M (W)	T_{dilute} (K)	T_{bead} (K)	L_{HE} (cm)	\dot{n}_3 (mmol/s)
0	0.3	0.5685	191.9	56
0.05	0.3	0.5894	186	76
0.1	0.4	0.6181	156.6	57
0.15	0.4	0.6319	175.4	68
0.2	0.4	0.6441	197.5	79

Table 7. Temperature Capability of Dilution Refrigerator With Various Microwaves at $I = 60$ nA

\dot{Q}_M (W)	T_{dilute} (K)	T_{bead} (K)	L_{HE} (cm)	\dot{n}_3 (mmol/s)
0	0.3	0.5828	188.5	69
0.05	0.4	0.6129	160.5	53
0.1	0.4	0.6274	179.6	64
0.15	0.4	0.6401	199.6	75
0.2	0.5	0.6716	165.2	62

Table 8. Temperature Capability of Dilution Refrigerator With Various Microwaves at $I = 70$ nA

\dot{Q}_M (W)	T_{dilute} (K)	T_{bead} (K)	L_{HE} (cm)	\dot{n}_3 (mmol/s)
0	0.4	0.6074	168.1	49
0.05	0.4	0.6227	183.7	60
0.1	0.4	0.6360	167.6	72
0.15	0.5	0.6683	188.2	59
0.2	0.5	0.6777	189.9	67

Table 9. Temperature Capability of Dilution Refrigerator With Various Microwaves at $I = 80$ nA

\dot{Q}_M (W)	T_{dilute} (K)	T_{bead} (K)	L_{HE} (cm)	\dot{n}_3 (mmol/s)
0	0.4	0.6178	192.1	56
0.05	0.4	0.6317	168.3	68
0.1	0.4	0.6439	189.6	79
0.15	0.5	0.6746	140.2	65
0.2	0.5	0.6835	149.5	73

Table 10. Temperature Capability of Dilution Refrigerator With Various Microwaves at $I = 90$ nA

\dot{Q}_M (W)	T_{dilute} (K)	T_{bead} (K)	L_{HE} (cm)	\dot{n}_3 (mmol/s)
0	0.4	0.6272	170.3	64
0.05	0.4	0.6399	191.4	75
0.1	0.5	0.6714	153.4	62
0.15	0.5	0.6806	161.5	70
0.2	0.5	0.6890	170.5	78

Table 11. Temperature Capability of Dilution Refrigerator With Various Microwaves at $I = 100$ nA

\dot{Q}_M (W)	T_{dilute} (K)	T_{bead} (K)	L_{HE} (cm)	\dot{n}_3 (mmol/s)
0	0.4	0.6357	193	71
0.05	0.5	0.6681	172.8	59
0.1	0.5	0.6776	175.8	67
0.15	0.5	0.6863	182.9	75
0.2	0.6	0.7262	88.02	66

Figure 1 displays a graph of the volume flow rate versus molar flow rate for a dilution refrigerator with a still temperature of 0.75 K. The plot was generated from Eq. (4.1) with the vapor

pressure at 0.75 K being 2.0 torr. This pressure was obtained from a table of temperatures given at various vapor pressures of He³ and He⁴. The graphs shows that even for the molar flow rate limit of 80 mmol/s, the volume flow rate needed would be less than 3000 m³/h. This flow rate is easily achievable with some roots pumps. Therefore, a molar flow rate of less than 80 mmol/s is definitely within reason and a good range to use when constructing a dilution refrigerator.

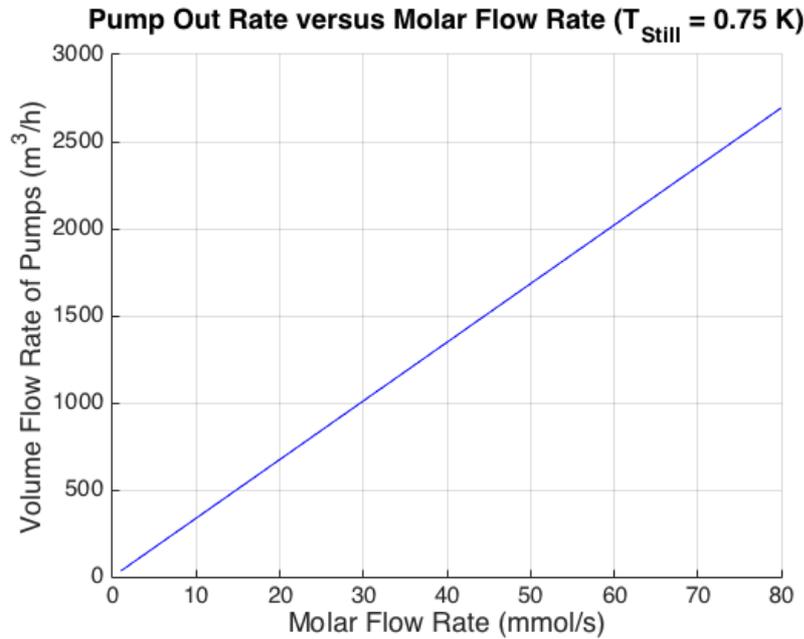


Figure 1.

Figures 2-9 display graphs of bead temperature versus the bath temperature at various beam currents and microwave powers. These plots were generated by Eq. (2.3) with \dot{Q} being calculated by Eq. (1.2). The red line is the data that uses the given Kapitza resistance value of 1.4×10^{-2} W/(cm²·K⁴) at 1 K. However, the Kapitza resistance is a very difficult value to measure and there is about an order of magnitude error in the value. This error then causes an error in the bead temperature which is represented on the graphs by the blue band around the red line.

One thing to note is the fact that the error band gets smaller as the bath temperature increases. This corresponds to the fact that the Kapitza resistance does not have much of an effect at higher temperatures. Especially at low power, the Kapitza resistance effect is small enough to be ignored at higher temperatures, resulting in no temperature mismatch between the beads and the bath. This is what is seen in the graphs below. As the bath temperature decreases, the error in the bead temperature increases because the effect of the Kapitza resistance increases and creates the temperature mismatch. Depending on the exact value of the Kapitza resistance, the temperature mismatch will begin at a higher or lower temperature, which is what is shown by the size of the error band. The power per bead also influences the size of the error. At higher power, the Kapitza resistance has a greater effect, and as just discussed, that means the error in the bead temperature is greater as well.

This actually allows for a way to measure the Kapitza resistance of the beads. By measuring the bath temperature and bead temperature at a given power, the Kapitza resistance can be calculated, which in the future would allow for these graphs to be made more precisely.

The next conclusion to be made from these graphs is just how cold the beads can get with this theoretical setup. Even given the error in the Kapitza resistance, the bead temperature at almost any given power will be below 1 K. Only at the highest powers with the beads having a radius of 1 mm is there the possibility of the bead temperature being around 1 K within the error. However, in most cases, the bead temperature hovers around 0.6 K. If a specialized refrigerator could be built to cool to around 0.6 K for 100 nA, this could have significant impact on future particle/nuclear experiments.

At lower beam intensities, the beads can be cooled to around 0.4 K. In fact, figure 2 shows the extreme case of 0.1 mm beads with a 1 nA charged particle beam and no microwave, and the bead temperature can be decreased to around 0.3 K in this case. However, even in the more realistic case of 1 mm beads with 1 nA beam current and no microwave, figure 8 demonstrates that the coldest temperature of the beads is around 0.4 K. This is important because as the temperature of the beads decreases the polarization of the beads increases. The increase in polarization can be seen by looking at the fact that the polarization in thermal equilibrium is equal to $\tanh(\mu B/kT)$ with μ being the particle's magnetic moment, B the magnetic field, and k the Boltzmann constant. Using this equation, the polarization of the protons in the beads, whose magnetic moment is $\mu_p \approx \mu_e/660$ with μ_e being the magnetic moment of an electron, increases from 0.5% at 1 K in a 5 T magnetic field to 1.3% at 0.4 K in a 5 T magnetic field. So, being able to cool the beads down to 0.4 K could increase the polarization of the protons in the beads by about 2.5 times the polarization from previous experiments that hold the beads at 1 K.

It is worth mentioning that in order to take advantage of the lower temperature DNP system a semi-frozen-spin mode could be employed when polarizing the target. This means that the target is polarized initially at a higher temperature and then cooled down as the microwave power is attenuated down to a manageable heat load, or all the way off. Since the beads are being cooled to lower temperatures, less microwave is needed to hold the polarization.

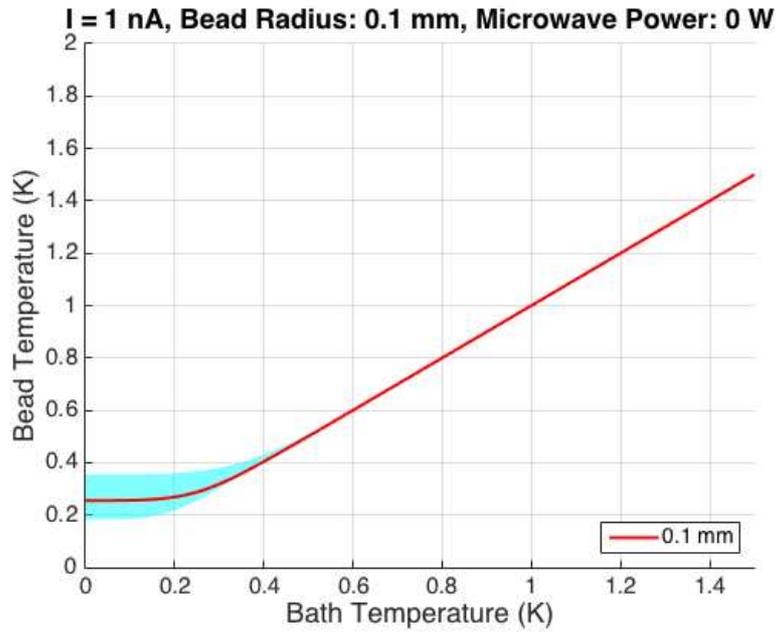


Figure 2.

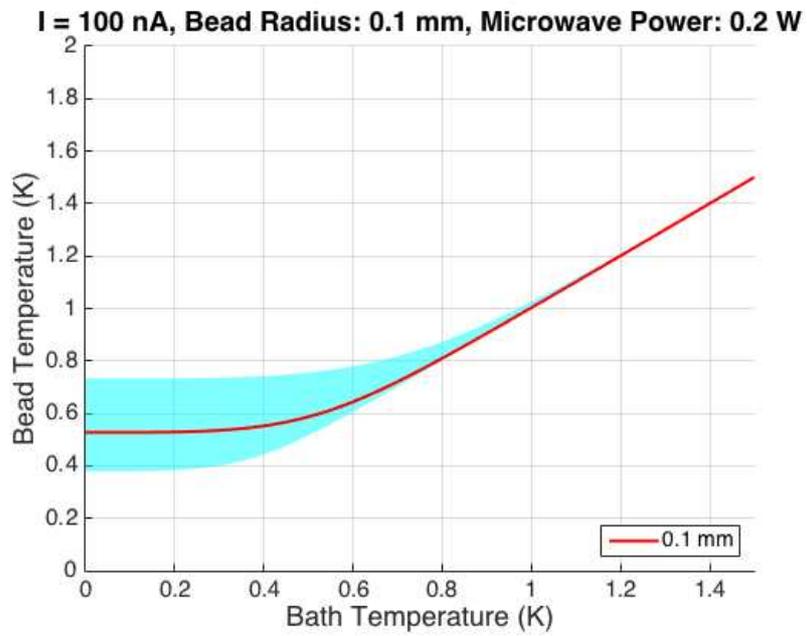


Figure 3.

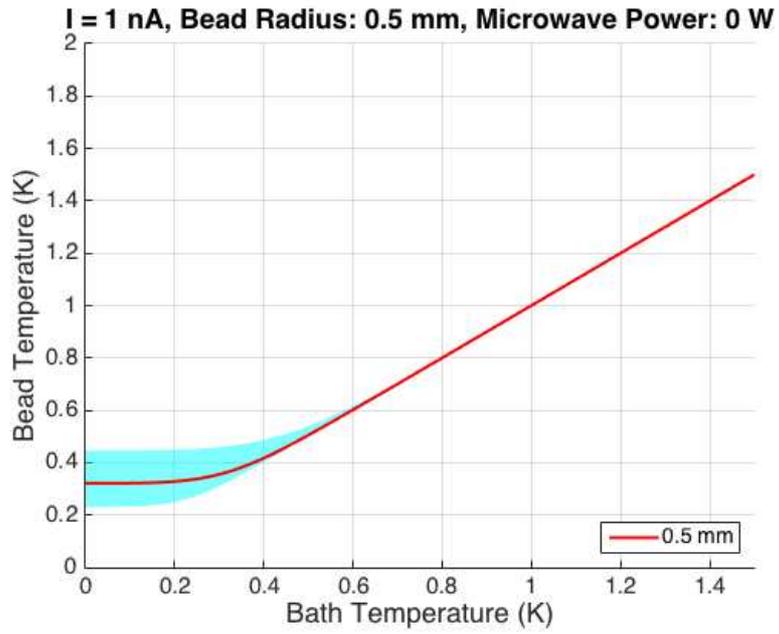


Figure 4.

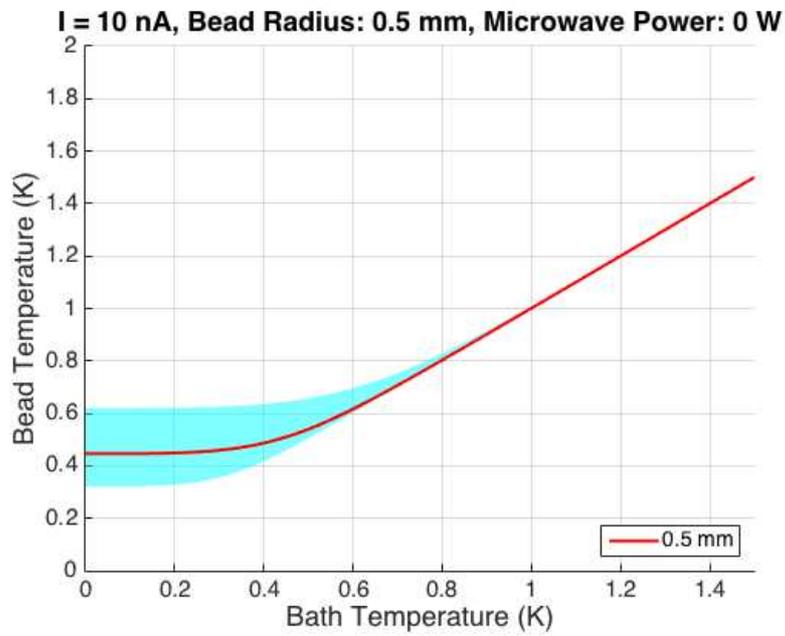


Figure 5.

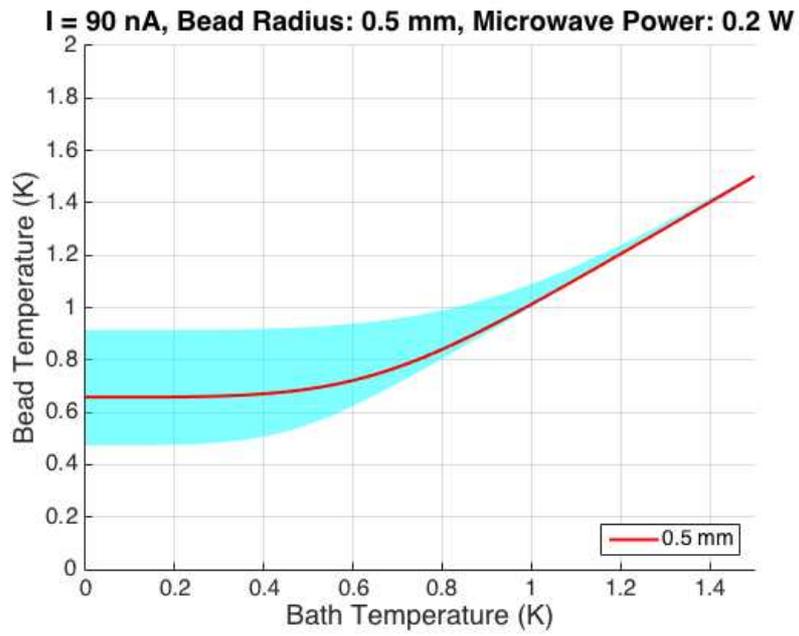


Figure 6.

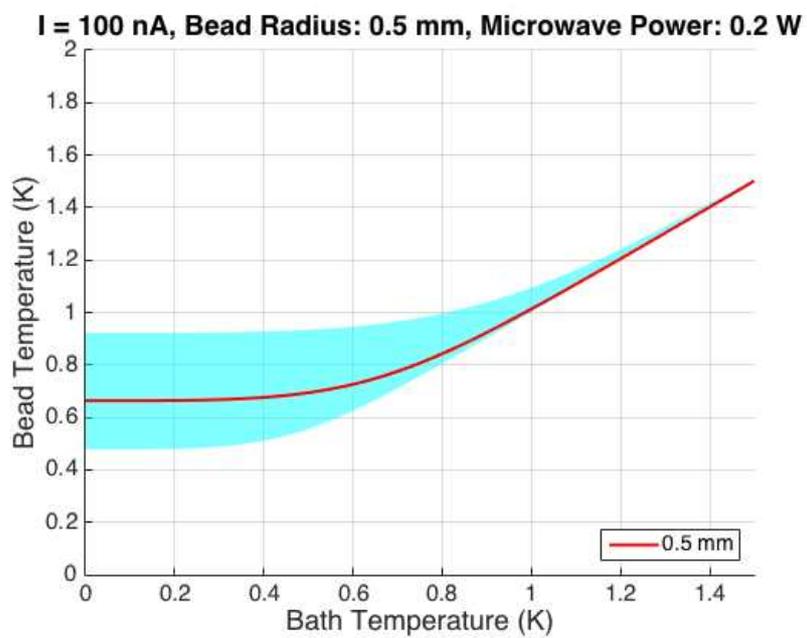


Figure 7.

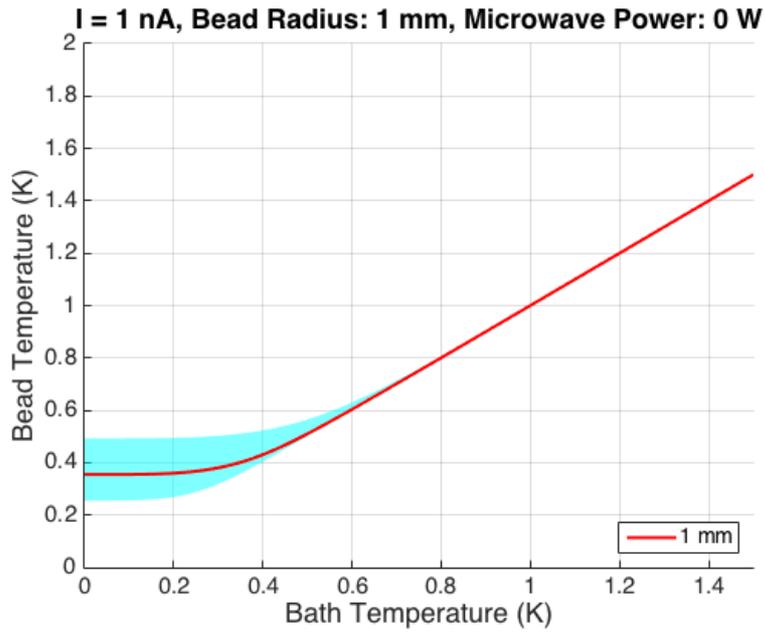


Figure 8.

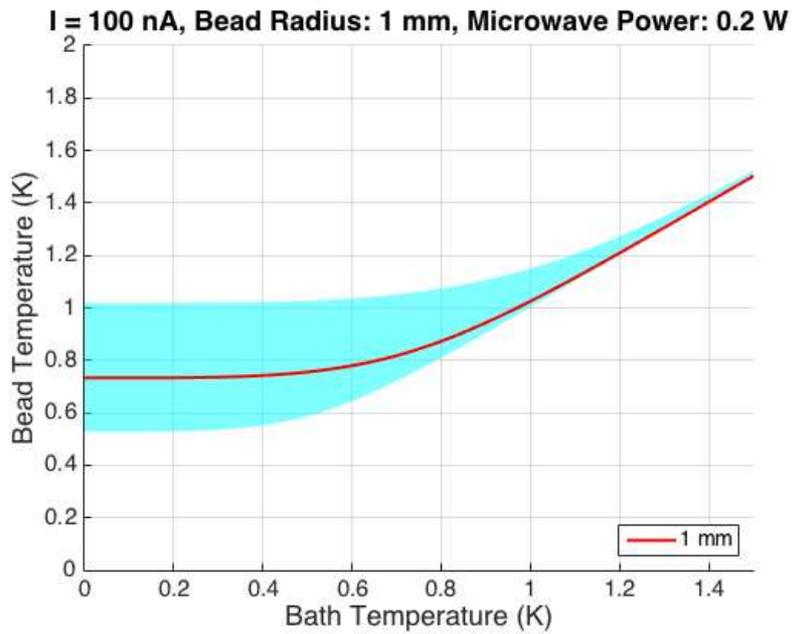


Figure 9.

Figures 10-13 present graphs of various parameters versus temperature for a He³ evaporation refrigerator. The parameters shown are volume flow rate, cooling power, and tube diameter for the pump out. The gray line corresponds in each graph corresponds to a volume flow rate of 7000

m^3/h , which is a flow rate that is financially reachable. Therefore, any point below the green line is reasonably doable, while the points above the gray line would require some pretty expensive pumps to achieve. The black line in each graph corresponds to a volume flow rate of $15000 \text{ m}^3/\text{h}$, which is the highest practical volume flow rate. A higher volume flow rate would require pumps that are so big that they become impractical. Therefore, only points below the black line are practically obtainable.

Figures 10 and 11 display one of the difficulties of using the refrigerator as a He^3 evaporation refrigerator. These graphs display the volume flow rate over the temperature of the He^3 bath. It can be seen that at especially the lower temperatures the volume pump out rate needed is really high making them either impractical or financially difficult to achieve. This indicates that running a dilution refrigerator might be better because the pumps needed to run the dilution refrigerator at the same temperatures would be much smaller and easier to obtain.

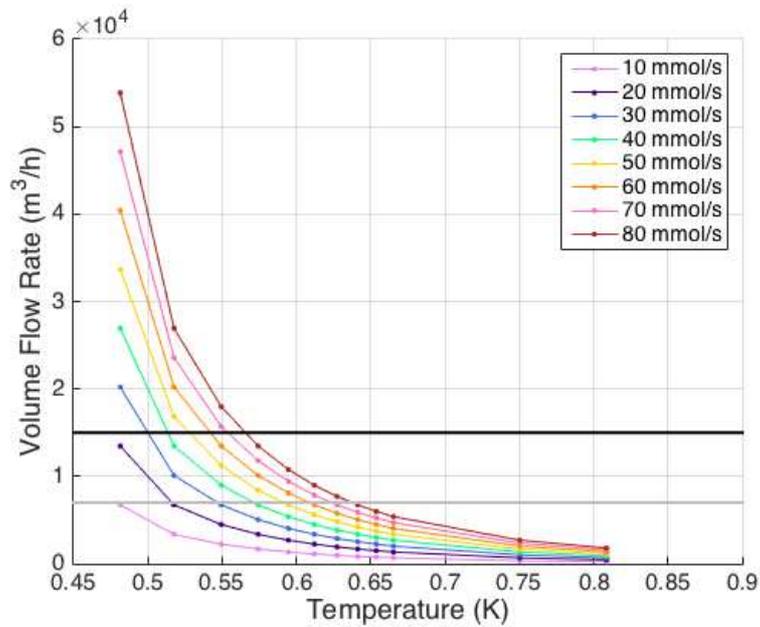


Figure 10.

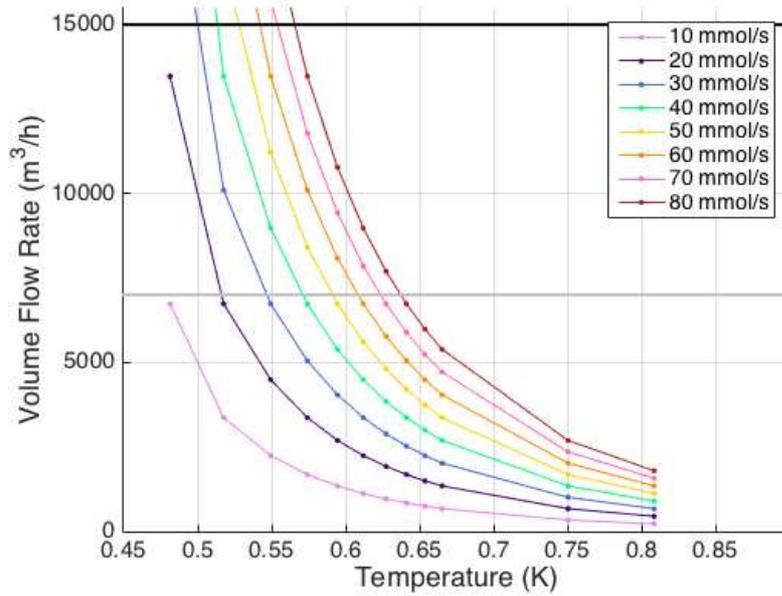


Figure 11.

Figure 12 demonstrates the next difficulty of the He^3 evaporation refrigerator. This is the graph of the tube diameter for the pump out versus the He^3 bath temperature. The plot was constructed from Eq. (5.4) with a length of $l = 70.7$ cm. This length was chosen because the pump out must travel the whole length of the refrigerator, and the refrigerator that was modeled for this paper is 70.7 cm long from the mixing chamber to the He^3 pump out. It can be seen that as the temperature decreases, the tube diameter grows pretty rapidly. It can be noted that to hold a temperature of less than 0.6 K, which is the minimum holding temperature of the dilution refrigerator at high cooling power, the pump out tube would have to be greater than 4 cm in diameter, which is already bigger than the diameter of the target cup. That being said, there are few ways to make the pump out tube more manageable.

The first idea that would require more calculations is to decrease the diameter of the tube towards the mixing chamber. Since the temperature of the gas decreases the further down the refrigerator it is, it can be seen from the ideal gas law that the molar volume also decreases, and because the molar flow rate is independent of temperature and would remain constant, the volume flow rate would decrease with temperature. So, the needed tube diameter would also decrease. While this does make the tube smaller at some places in the refrigerator, it could be cumbersome to make because it would then be a cone shape.

The second idea that this paper shall suggest as the better method is to make the tube a different shape while keeping the same cross-sectional area. Since the flow down the tube is assumed to be laminar, which was an assumption made in the tube's conductance equation, other shapes besides a circle should be able to allow the same volume flow rate. By morphing the shape of the tube, the cross-sectional area can be maintained while decreasing the amount of space that it takes in the refrigerator. For example, one can make the tube into a curved slot that wraps around the

upper part of the refrigerator. It should be noted that because the mathematical theory behind fluid flow through non-circular tubes is not well known, an attempt should be made to make the cross-sectional area larger than needed in order to increase the flow rate through the tube.

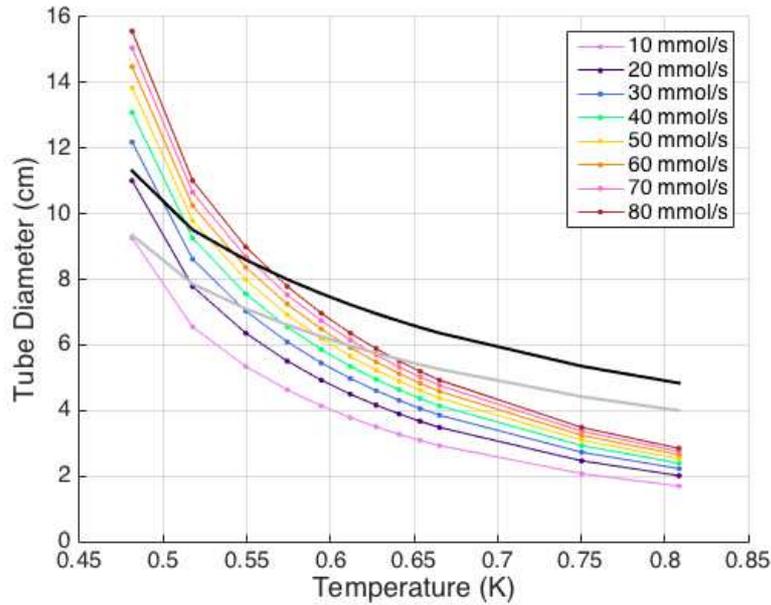


Figure 12.

Finally, figure 13 shows why a He^3 evaporation refrigerator can be better than a dilution refrigerator. The cooling power of a He^3 evaporation refrigerator can be much greater than that of a dilution refrigerator. Even at 0.48 K, the evaporation refrigerator can theoretically get up to a cooling power of more than 0.5 W. Now, in order to get higher cooling power, the flow rate must be increased, but even 0.8 W is financially achievable at 0.55 K, which is much better than the achievable cooling power of the dilution refrigerator at 0.55 K. This higher cooling power would allow for higher intensity beam experiments with continuous microwave polarization. While the temperature can only be held between 0.5 and 0.6 K, this would result in a bead temperature between 0.6 and 0.7 K, which as discussed previously is very good. Therefore, it is definitely still worth while to incorporate the components necessary for the refrigerator to run as a He^3 evaporation refrigerator at higher beam intensities.

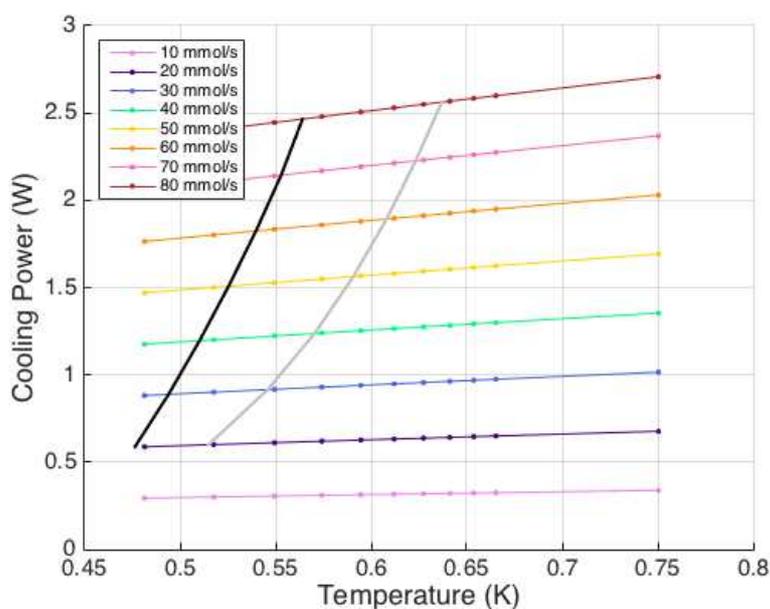


Figure 13.

7. Super Cooled He⁴ Refrigerator Design Concept

The optimal performance of a supercooled system should be specialized with geometry to match the temperature and cooling power goals. From the initial look that we have done here its clear that a higher temperature unit could be made just with a He³ evaporation cooling system for supercooled He⁴ to get the best cooling power. A separate design could be employed for low intensity experiments that could achieve higher polarization at the cost of the cooling power by using a dilution system for cooling. We explore the possibility of combining these but make it clear that this is not the optimal approach for either case.

Figures 14-17 are computer models of the super cooled He⁴ refrigerator concept. All the same components as a dilution refrigerator are present including the He⁴ evaporation part that cools the He³ down to about 1 K. The He³ is then sent through the still, which is about 0.75 K, through the heat exchanger and emptied into the mixing chamber. One of the major additions to the refrigerator is that of the He⁴ bath at the very end. The bath can be seen in Figure 15. Inside the mixing chamber is a nose piece that has a back wall closing off half of the opening to the nose piece. This wall creates a cavity that is then filled with He⁴. This bath is then where the target material is placed in order to be cooled down. In Figure 15, the outlet where the He⁴ is emptied into the bath is seen in red, and two target cups can be seen held by the target stick. The target cups must have a diameter less than half the diameter of the nose piece so that the cups are fully immersed in the He⁴ bath. This also allows for the cups to be inserted and taken out through the opening in the top half of the nose piece. Therefore, the nose piece would have to be greater than 6 cm in diameter because the target cups that have been considered have a diameter of 3 cm. Lastly, note that the stick holding the target cups should be made out of pitch bonded graphite. Since the lower part of

the stick will also be in thermal contact with the target material and the He⁴ bath, the stick needs to have very low thermal conductivity at low temperatures. Pitch bonded graphite has this property. The final note about the addition of the He⁴ bath is the fact that the target is no longer in the center of the refrigerator. Because of this the charged particle beam will also have to be positioned lower such that it aligns with the target cups.

Figure 16 displays the idea for the He³ pump out tube to be used when running the refrigerator as a He³ evaporation refrigerator. The pump out tube is light brown in color and can also be seen running through the entire refrigerator in Figure 14. The tube exemplifies the second idea for dealing with the large cross-sectional area needed that was described above. Instead of being a circle, the tube's shape is a slot that wraps around the top of the nose piece. By wrapping it around the nose piece, it does impinge on the space needed for the stick with the target cups to be placed into the refrigerator. Also, in order to get more area in the tube, the slot can be lengthened rather than adding a lot more diameter to the refrigerator.

Finally, figure 17 demonstrates how the He⁴ will travel down the refrigerator to the He⁴ bath. The He⁴ lines are drawn in red and the He³ lines are drawn in orange. The lines follow each other from the separator to the evaporator into the still and through the heat exchanger. It is important that, especially in the heat exchanger, the lines do not touch each other. If they touch, then the He³ and He⁴ will be in thermal contact and since He⁴ does not cool nearly as fast as He³, the temperature of the He³ will be higher than expected and needed. Note that the length of the heat exchanger is how long the He³ line is if it were to be uncoiled. Therefore, even if a 150 cm or longer heat exchanger is needed, it can be easily condensed by coiling it up.

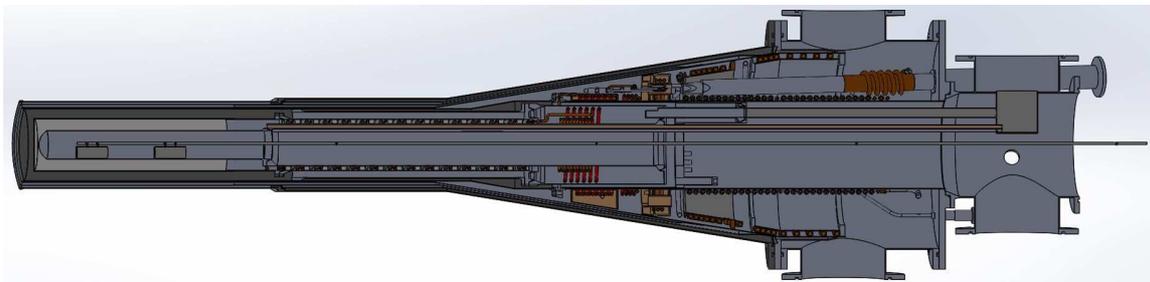


Figure 14. Cross-Section of Super Cooled He⁴ Refrigerator Concept

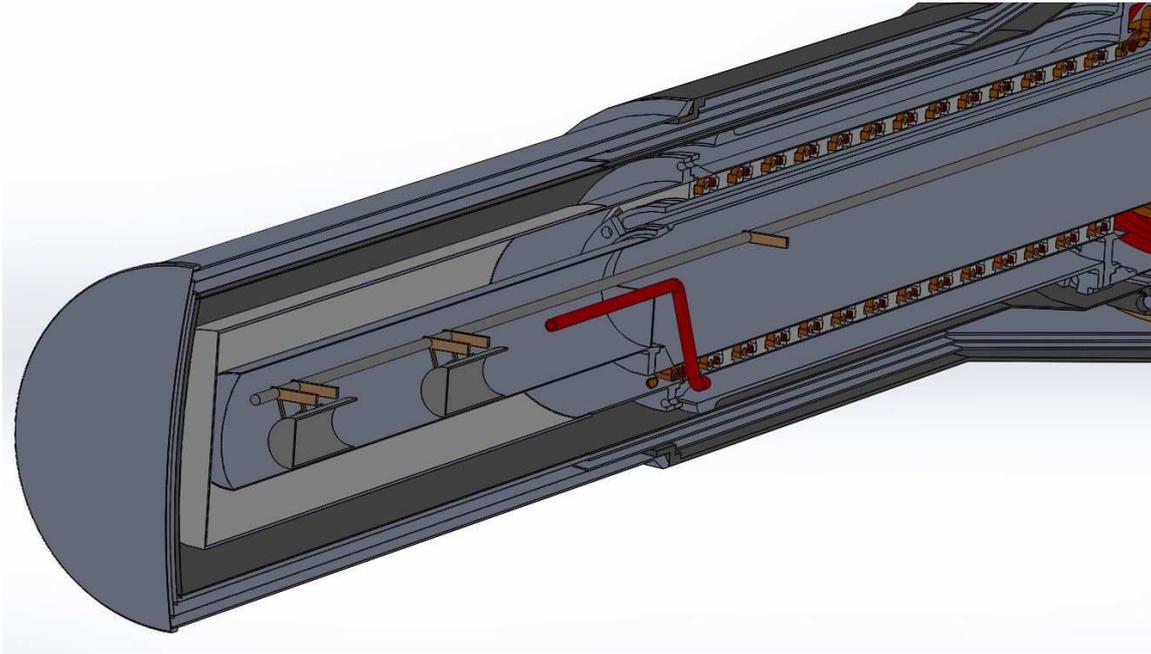


Figure 15. He⁴ Bath

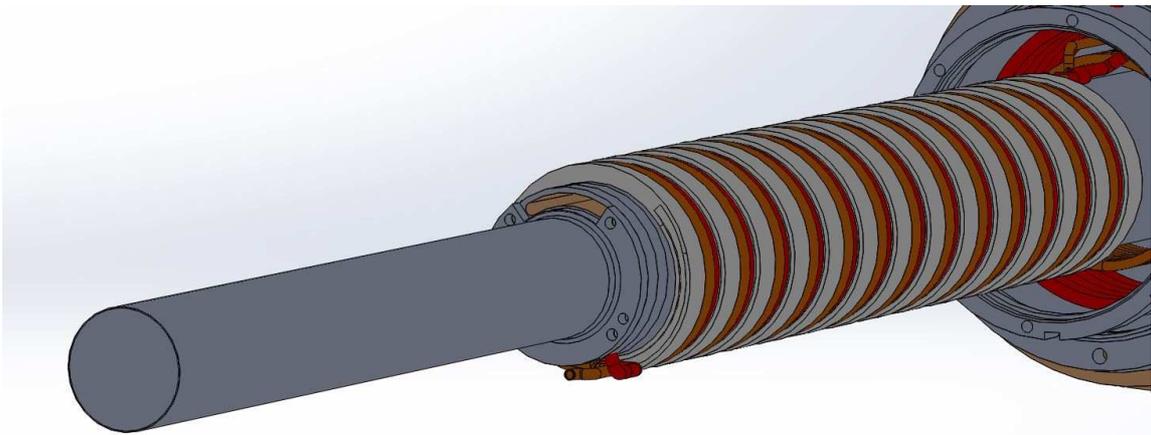


Figure 16. He³ Pump Out for He³ Evaporation Refrigerator

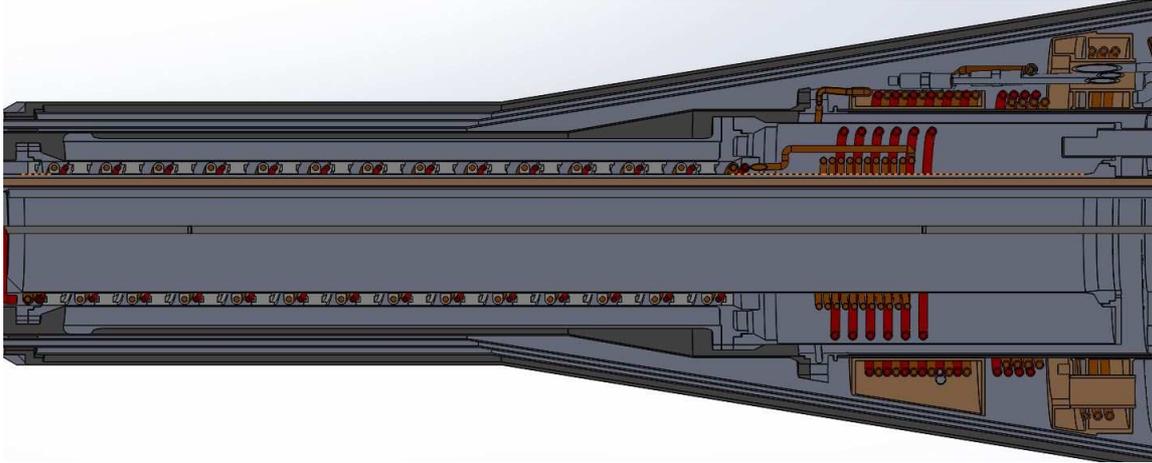


Figure 17. He³ and He⁴ Lines From Separator through Heat Exchanger

8. Conclusion

We conclude that a supercooled He⁴ system is likely possible and very likely to improve the figure-of-merit for specific particle/nuclear experiments. We explore the idea for this type of subcooled helium refrigerator that takes advantage of the low temperature cooling of a He³-He⁴ dilution or He³ evaporation refrigerator while also taking advantage of the better thermal properties of He⁴ over He³. By placing the target material in a bath of He⁴, the material can continuously be cooled while it is hit with a particle beam. So, if the He⁴ bath can be cooled to a low temperature, then the target material can also be continuously cooled to a low temperature as well. The reason for wanting to cool the target material to a lower temperature is that then the material can be polarized to a greater extent.

While the He⁴ bath is cooled to the temperature being held by the refrigerator, the target material will be at a warmer temperature due to the Kapitza resistance between the material and the liquid He⁴. The material considered was ammonia beads. It was determined that at the higher beam intensities 60-100 nA the beads could be held at a temperature of about 0.6 K, and at the lower beam intensities 1-10 nA the beads could be held around 0.4 K. In order to reach the lower temperatures at the lower beam intensities, a frozen spin mode would have to be used. However, with standard He⁴ evaporation refrigerators, the beads are generally held at 1 K, and so the possibility of being able to hold them at near 0.6 K is an excellent motivation to explore this further.

Finally, the concept of a super cooled He⁴ refrigerator was explored by examining the capability of both a dilution refrigerator and a He³ evaporation refrigerator. At beam intensities of 1-50 nA, running the refrigerator as standard He³-He⁴ dilution refrigerator would provide the cooling power needed to keep the beads cold. At beam intensities higher than 50 nA, a He³ evaporation refrigerator would be able to keep the He⁴ bath at a lower temperature with higher cooling power than a dilution refrigerator. The He³ evaporation refrigerator requires much less pumping than the dilution refrigerator for the cooling power that you get.

Moving forward, there is still a lot that could be done as far as modeling the refrigerator with proper dimensions to see just how large it might be. Once the refrigerator (or refrigerators) is

properly modeled, it can be constructed to run polarized target experiments at many different beam intensities and temperatures. In addition to constructing the actual refrigerator, one can look into using different shapes of material as the targets. This paper considered bead shaped material, but one could also consider the shape of a wafer, like a thin disk. With the increase in surface area, one might be able to get the temperature down a bit further.

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