Dilution Refrigerators for Quantum Science

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ICMC Short Course: Quantum Information Science
International Cryogenic Materials Conference 2021

Course outline and objectives

Objective: To understand the physics behind the dilution cooling process, be familiar with the historical development and the design features of modern refrigerators.
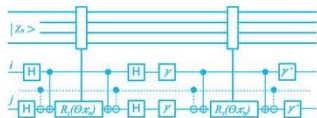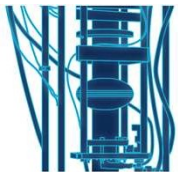
Outline:

- Physics of helium and the behavior of 3He/4He mixtures
- Making a practical refrigerator
- The development of dilution refrigerators from the 1960s to today
- Modern designs and operational matters
- Future developments

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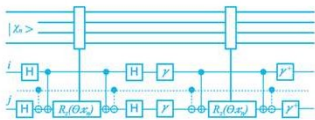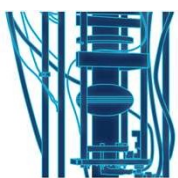
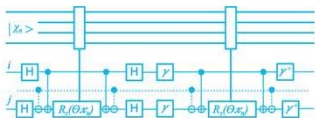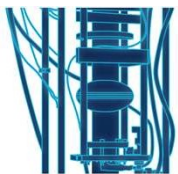# Physics of Helium and Helium Mixtures

Helium mixtures

- Important to note that **Helium-4 obeys Bose statistics**. Superfluid transition occurs at 2.18 K. **Helium-3 obeys Fermi statistics**, obeys the Pauli principle and has a superfluid transition near 1 mK.

- In the following discussions, the respective concentrations of the two helium isotopes are expressed as

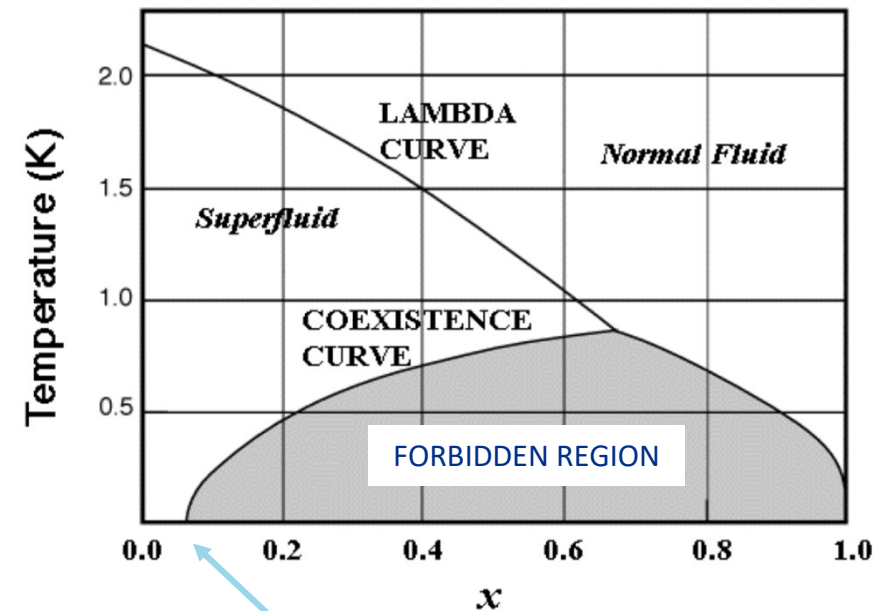$$x = x_3 = \frac{n_3}{n_3 + n_4}$$

$$x_4 = \frac{n_4}{n_3 + n_4}$$

where n_3 and n_4 are the number of atoms or the number of moles of 3He and 4He, respectively.

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Phase diagram and solubility

Temperature (K) vs x phase diagram showing LAMBDA CURVE, Normal Fluid, Superfluid, COEXISTENCE CURVE, and FORBIDDEN REGION.

Diluting 3He into 4He depresses the superfluid transition temperature

Superfluid transition disappears completely for $x > 67.5\%$

As a mixture with $x > 6.6\%$ is cooled, it will separate in to 2 phases.

One phase (the "concentrated phase") will approach x=100%, the other (the "dilute phase") will approach $x = 6.6\%$.
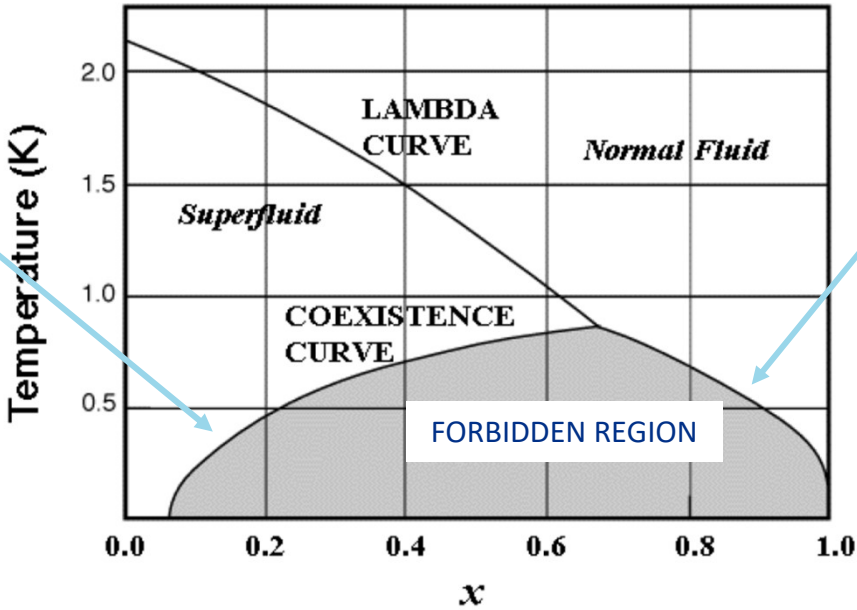
This finite solubility and phase separation is key to the dilution refrigerator process

Finite solubility – on the 4He-rich side, the concentration does not go to zero at T=0

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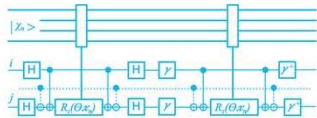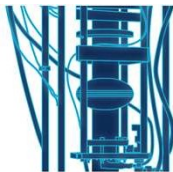MATERIALS & SYSTEMS CENTER
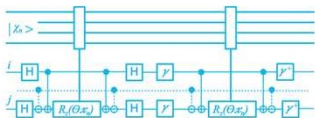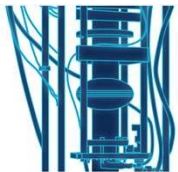
Phase separation at low temperature

4He-rich phase

Higher density

3He-rich phase
(essentially pure 3He)

Lower density, floats
on top of the 4He-rich
phase

LAMBDA CURVE

Normal Fluid

Superfluid

COEXISTENCE CURVE

FORBIDDEN REGION

Temperature (K)

x

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3He-4He mixtures as Fermi liquids

- As mentioned, 4He obeys Bose statistics and will condense into the quantum mechanical ground state at low enough temperature.

- In a helium mixture, the condensed 4He acts as an "inert superfluid background" which contributes to the volume of the liquid and the effective mass of the 3He atoms but has **zero heat capacity**.

- The 3He obeys Fermi statistics and will behave in a similar way to conduction electrons in a metal. However, whereas conduction electrons have a Fermi temperature of ~10000 K, the Fermi temperature of the 3He is ~1 K.

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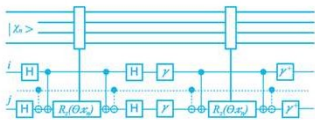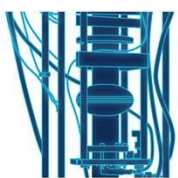
Finite solubility (1)

- Let us consider whether a 3He atom at the phase separation line would "prefer" to be in the 3He phase or be in the 4He-rich phase. The 3He will go into the phase with the **larger binding energy**.

- For **3He atoms in pure 3He**, the binding energy is the latent heat of evaporation.

- For **3He atoms in 4He**, although the van der Waals forces between 4He and 3He are identical, the 3He atoms have larger zero-point motion due to their smaller mass.
  – The 4He atoms occupy a smaller volume than the 3He atoms, so 3He atoms will feel a stronger attraction to a 4He atom.
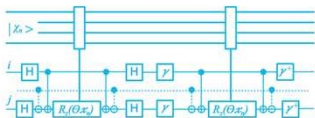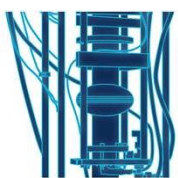
Finite solubility (2)

- As more 3He atoms enter the 4He phase, two effects occur:
  - The larger zero-point motion of the 3He means that the liquid near the 3He atoms is more dilute than near 4He atoms, and the 3He atoms will tend to group. **Binding energy of the 3He atoms will increase with increasing x_3.**
  - The 3He atoms must obey the Pauli principle, so additional 3He atoms must occupy successively higher energy states. **Binding energy of the 3He atoms will decrease with increasing x_3.**

- The first effect is dominant – as x_3 increases, the binding energy of 3He in 4He approaches the binding energy of 3He in 3He, and an equilibrium is reached.

- This is true even at $T = 0$, hence the finite solubility effect.
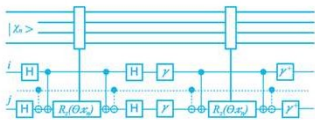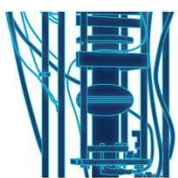
Cooling power of the dilution refrigerator

- Measurements of the specific heat shows that the enthalpy of 3He in the dilute phase is larger than in the concentrated phase. Moving 3He from the concentrated to the dilute phase will result in cooling due to the **heat of mixing**
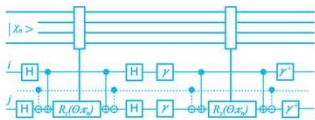
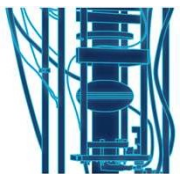$$\dot{Q}(T) = \dot{n}_3 (H_d(T) - H_c(T))$$

- We won't work through the full derivation here, but in summary

$$H_3(T) = H_3(0) + 11T^2$$

$$H_{3,d}(T) = H_3(0) + 95T^2$$

$$\dot{Q}(T) = 84\dot{n}_3 T^2$$

This result is correct to with a few % for $T < 40$ mK

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Cooling power – things to note

- The equation giving the cooling power is for the **liquid in the mixing chamber**. It takes no account of the Kaptiza resistance between the liquid and the chamber itself, or the contact resistance between the chamber and any external equipment such as a cold plate.

- Derivation assumes that the 3He in the concentrated and dilute phases are at the same temperature. In reality, the precooling of the inflowing 3He won't be perfect, so we modify the derviation to account to T_N, the temperature of the 3He leaving the last heat exchanger:

$$\dot{Q} = \dot{n}_3(96T_{mc}^2 - 12T_N^2)$$

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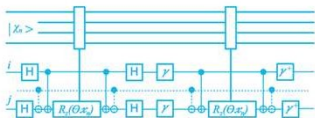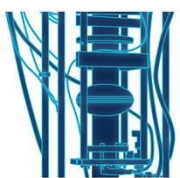
Osmotic pressure

- The osmotic pressure of the Helium mixture is a function of temperature and 3He concentration (see plot right from Betts, 1989).

- We will discuss this in more detail in the next section, but if we pump 3He away from the dilute phase the concentration will be reduced, resulting in a **difference in the osmotic pressure and driving the flow of 3He**.

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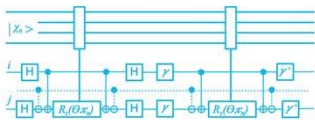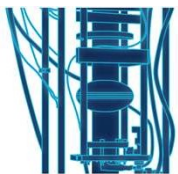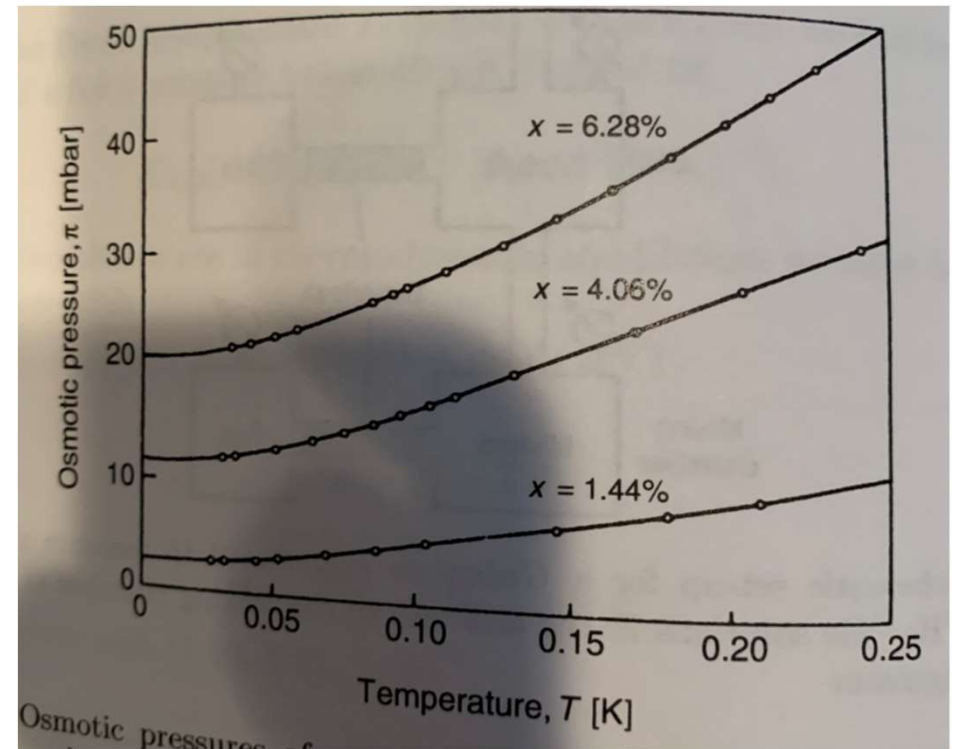
Building a Refrigerator
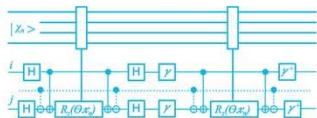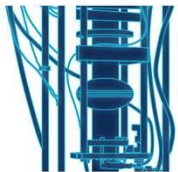
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General layout of a dilution refrigerator

- **Simplest model for the refrigerator is a U-tube**, with the concentrated phase on one side and the dilute side on the other.

- As observed previously, the less dense **concentrated phase floats on top of the dilute phase** since the superfluid has much higher density.

- Removing 3He from the dilute phase surface upsets the equilibrium.
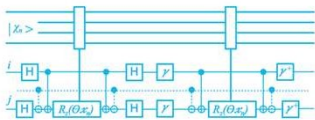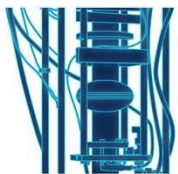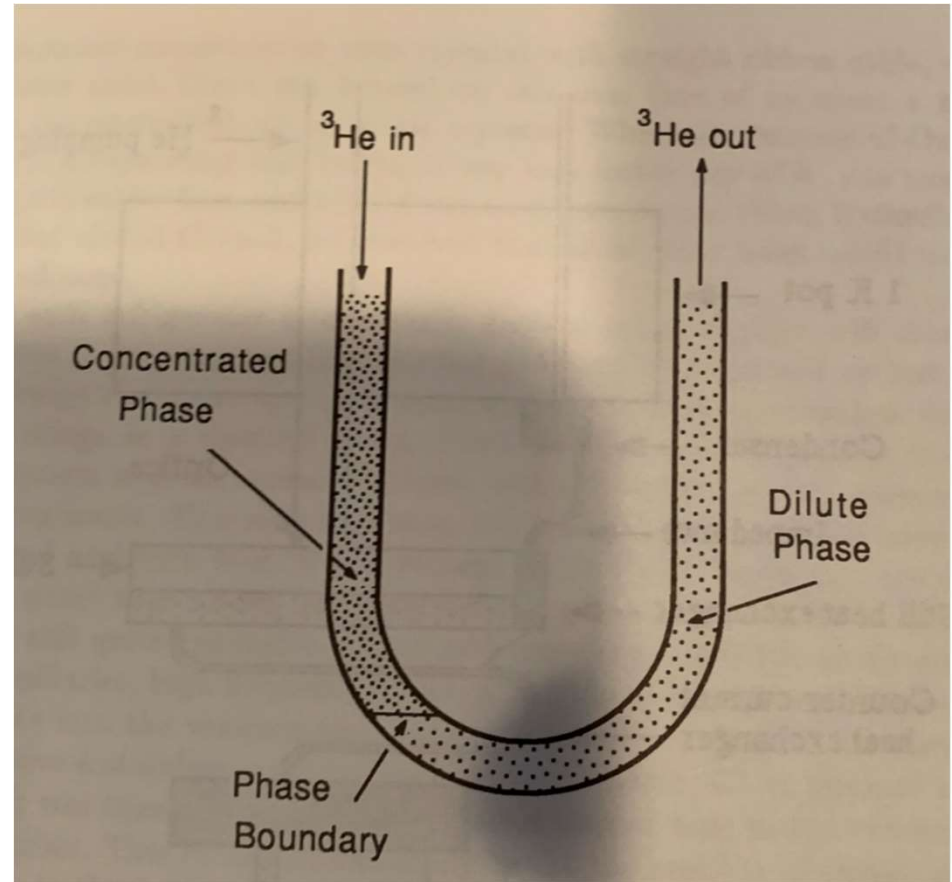
General refrigerator layout

- Major components of a generalized dilution refrigerator shown in the diagram.

- 3He from the room temperature pump system is cooled and liquified at/above the still

- Cooled further on the way to the mixing chamber

- 3He is pumped away from the still

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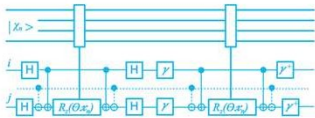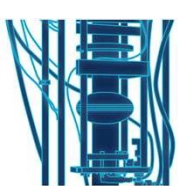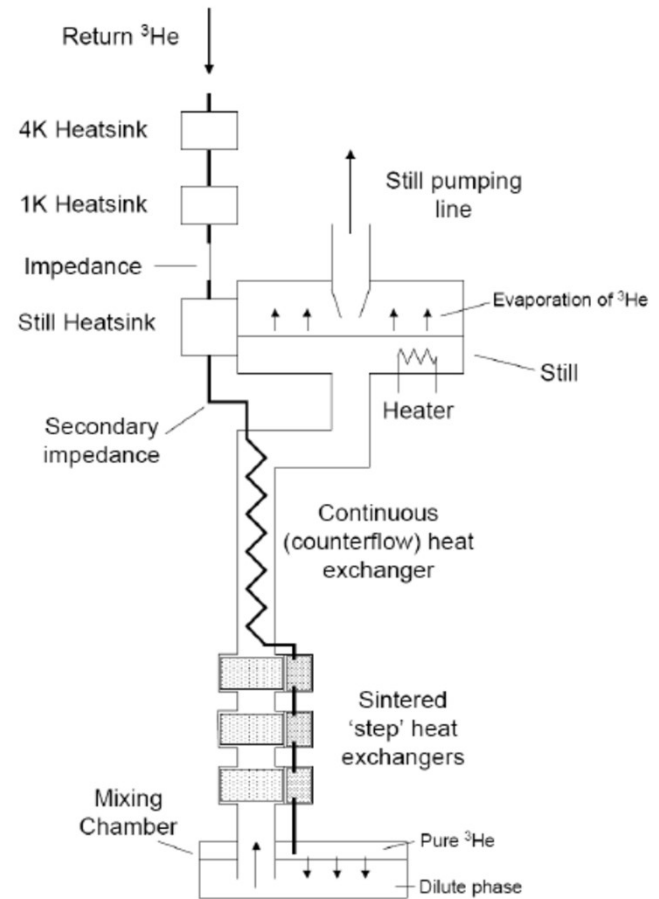
Heat exchanger design above 50 mK

- At higher temperatures, continuous heat exchangers work well. These are typically of the concentric (or coil-in-tube) configuration.
- Heat is transferred from the dilute phase in the annular space and the concentrated phase in the coiled capillary.
- Heat transfer along the fluid and the capillary are negligible.
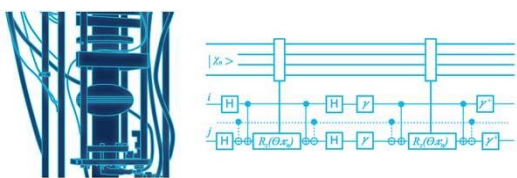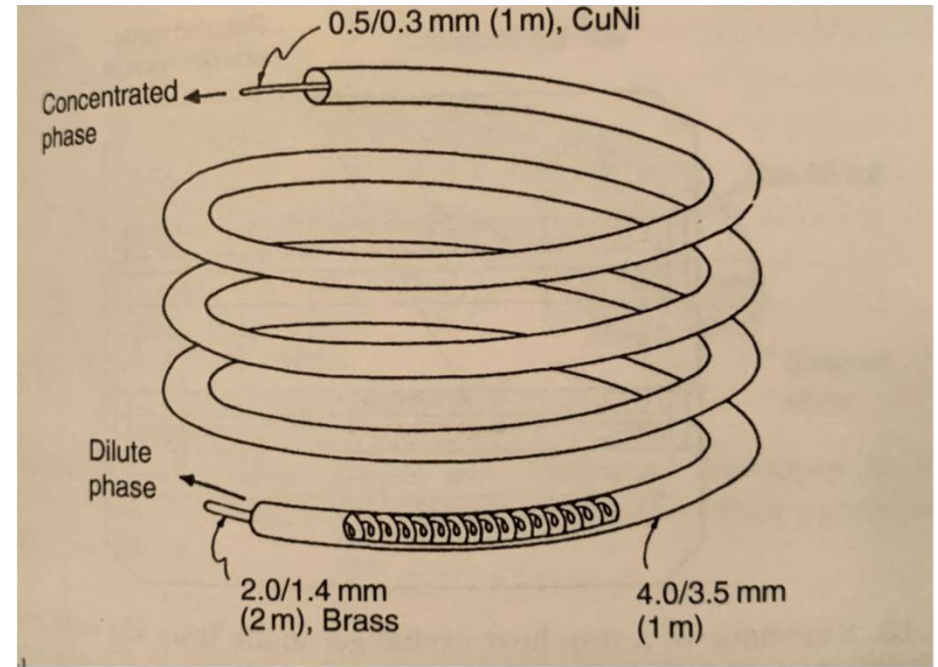
Heat exchanger design below 50 mK

- Designs using continuous heat exchangers can reach ~30 mK.

- For lower temperatures or very high flow rates, larger surface area heat exchangers are needed to overcome the increasing Kaptiza resistance.

- Simple form is a sintered metal block to give large surface area, with a machined channel to reduce flow impedance (and hence viscous heating).
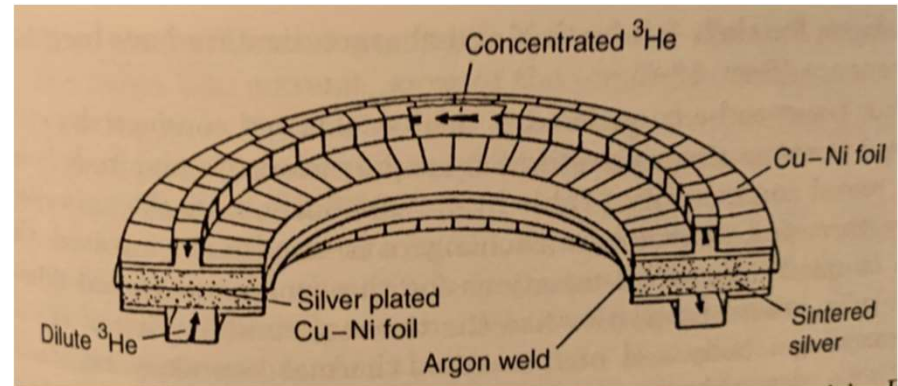
- To minimize axial heat flow, several discrete exchangers are connected in series – "step" heat exchangers.

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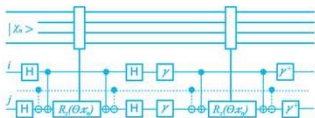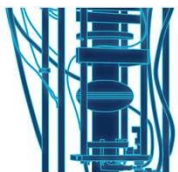
Practical configuration example

- The diameter of the flow channels increases as the temperature is lowered due to the decreasing density.

- Dilute channel has larger diameter than the concentrated side due to the lower density.

Exchanger No	Length	Area	Dilute side channel size	Concentrated channel size
Tubular	9 m coiled ϕ 3 mm tube inside 4 m of ϕ 5 mm tube	0.08 m²	ϕ 2.5 mm (clear bore inside coil)	not well-defined (flattened tube)
0 (above 20 mK flange)	60 mm	24 m² *	3 mm	2 mm
1	60 mm	24 m² *	3 mm	2 mm
2	60 mm	24 m² *	3 mm	2 mm
3	60 mm	24 m² *	4 mm	3 mm
4	60 mm	24 m² *	4 mm	3 mm
5	720 mm	30 m² *	7 mm	4 mm
6	720 mm	60 m² *	9 mm	4 mm
7	720 mm	60 m² *	11 mm	5 mm
8	720 mm	70 m² *	13 mm	5 mm

*—estimate

Table from Bradley et al. (1982). Fridge designed to operate at 3 mK

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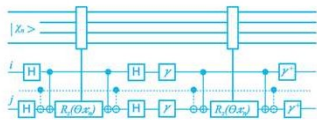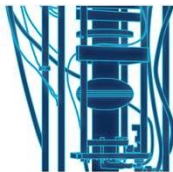
Mixing chamber

- The mixing chamber is a relatively simple. Major requirement is for a large contact area between the helium phases and the metal.

- This is typically achieved through sintered structures, such as the silver sponge example shown right.

- In this design, the diameter of the mixing chamber is approximately 100 mm and each sponge has a surface area of 10 m^2.
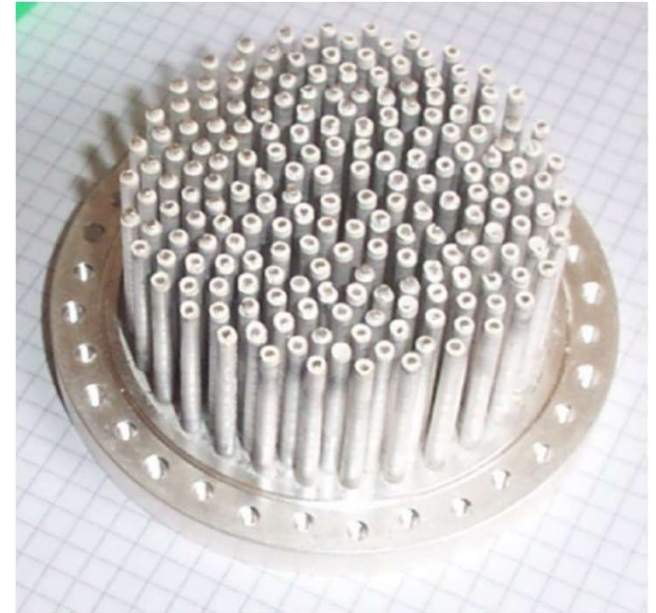
Mixing chamber construction example from Uhlig (2015)

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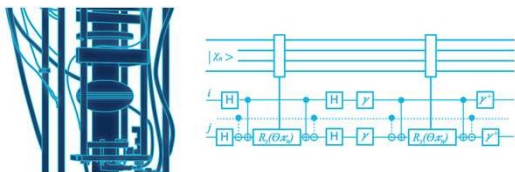
Room temperature components - Pumps

- Choice of pumps depend on circulation rate and desired base temperature.
- Usually consist of a primary pump (scroll or roots).
- Still pumping may be supplemented with one or more turbomolecular pumps for higher-performance systems.
- For efficient condensation, it is often necessary to use a compressor to increase the inlet pressure (and decrease the outlet pressure of the primary pump).
- **Important that pumps are sealed for helium use and do not contaminate the process line**.

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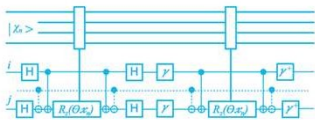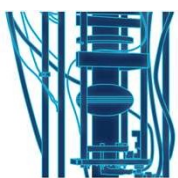
# Room temperature components – Filtering and sealing

- Majority of the gas handling at room temperature will be subatmospheric, so will tend to suck in air that could contaminate the process line.
  - Fridges generally include cold filters with Pt or other metal sinters internally
  - Many will also include an external activated charcoal trap in liquid nitrogen
  - For very long duration runs, an LHe or 4-K trap can also be used.

- Choice of flanges and seals is important. Metal seals generally much better than rubber, but harder to work with.
  - Be wary of parts of the system that will see internal pressure. If using rubber O-rings, it may be necessary to add overpressure rings.
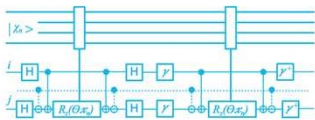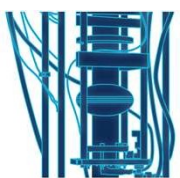
History and Development of the Dilution Refrigerator
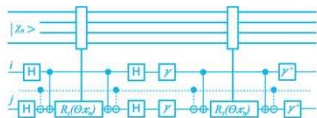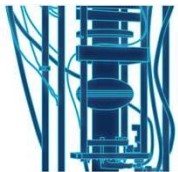
Early development

- The idea of using 3He/4He mixtures as a refrigeration method was first proposed by London in 1951, and further developed in 1962 by London, Clarke and Mendoza.

- First demonstrations:
  - Das, de Bruyn & Taconis (1965) reached **220 mK**
  - Hall, Ford and Thompson (1966) reached **80 mK**
  - Neganov, Borisov & Liburg (1966) reach **25 mK**

- The refrigerator of Hall *et al.* is shown on the right

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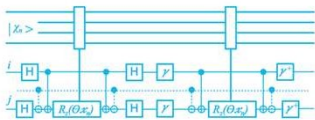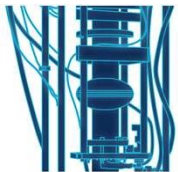
Early commercialization

- The first commercial refrigerators were built in the mid-1960s by Oxford instruments, first in collaboration with Heinz London, and later based on Hall's design licensed from the UK Atomic Energy Authority.
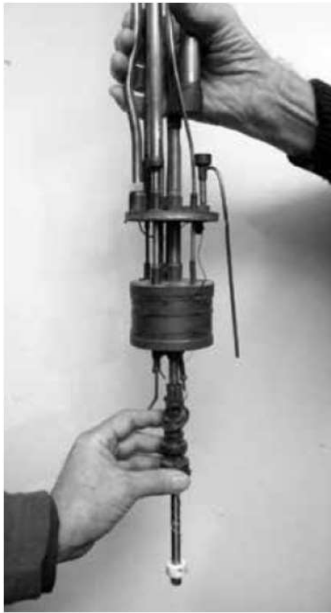

(Left) Early commercial refrigerator insert

(Right) Commercial refrigerator with copper heat exchangers, c.1975

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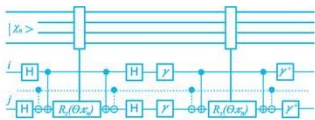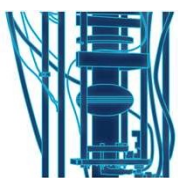
Eliminating the liquid helium

- The primary feature that distinguishes modern refrigerators from the older style is the use of mechanical precoolers in place of the liquid helium bath.

- This was made possible by an earlier innovation, the **Joule-Thomson Heat Exchanger**.

- Described independently by Kraus at ZTB and by de Waele at Eindhoven in 1977, this device eliminates the 1-K Pot in a wet dilution refrigerator by using a long **counterflow exchanger** cool the Helium-3 stream with gas pumped from the still, followed by an **expansion valve**.
  - This stage cools and partially liquifies the Helium-3, with the remaining liquification taken care of by the still.

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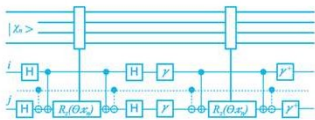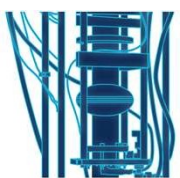
Eliminating the liquid helium (2)

- Removing the 1-K Pot served three important purposes:
    - The pumped pot can be a significant source of vibration in sensitive experiments
    - The pumped pot can dominate the Helium consumption of a system
    - The pot is prone to blockage, making it a common source of fridge failure

- Note that it would not be ideal to just cool and liquify the Helium-3 by running the gas directly to the still since the enthalpy of the gas is larger than that of the gas leaving the still at ~1K.
    - This has been demonstrated, but only for very low flow rate fridges with efficient precooling, so not a very practical approach.
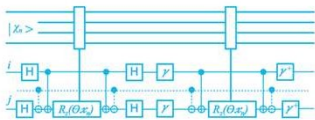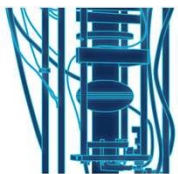
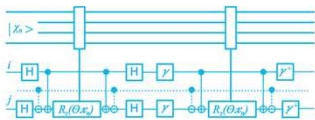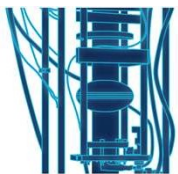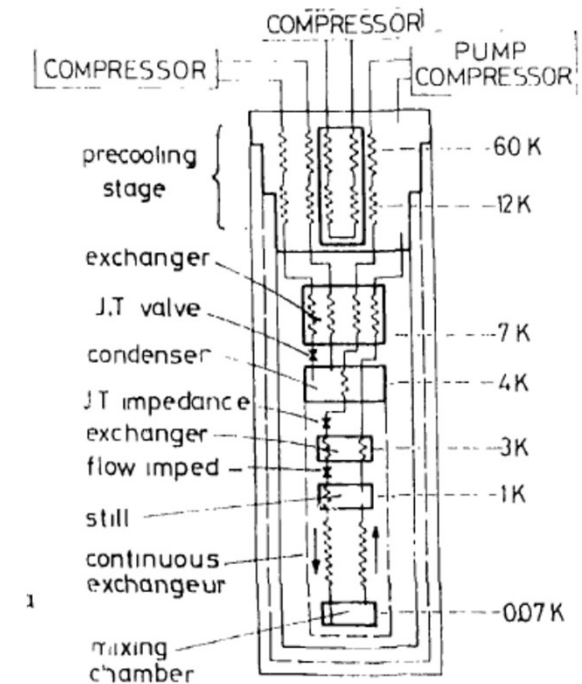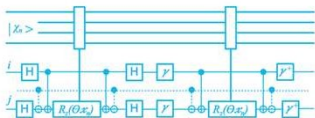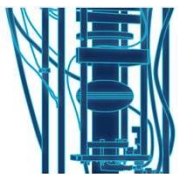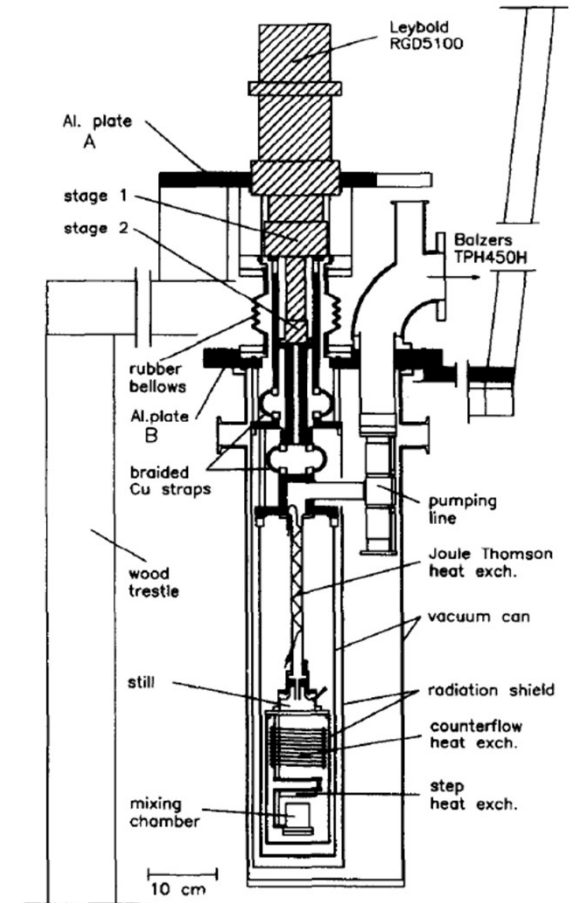# Eliminating the liquid helium (3)

- With improvements in mechanical cryocoolers in the late 1980s, it was a natural step to couple one to a dilution refrigerator.

- First example described by Pari in 1990, followed by the pioneering development work of Uhlig starting in 1993.

- Pari's machine (shown right) used a Stirling cycle cooler operating at 12 K combined with a Helium-4 J-T to produce a temperature at ~4 K.
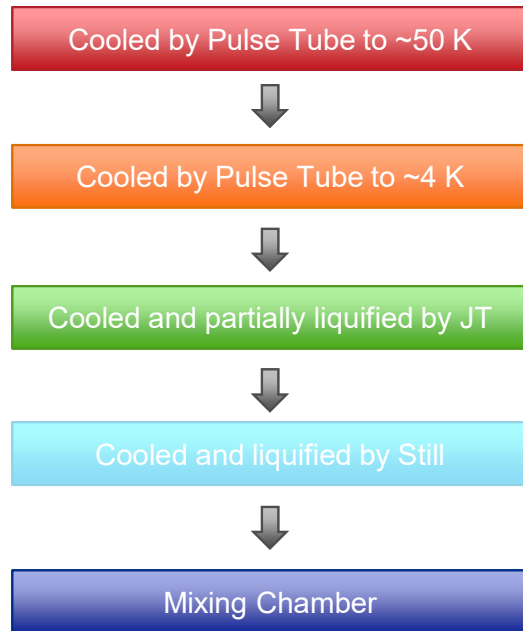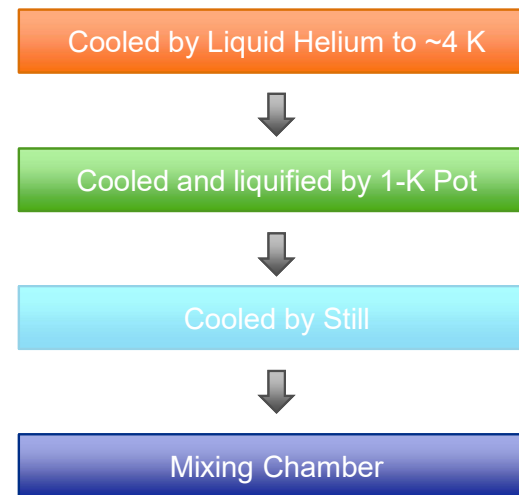
Eliminating the liquid helium (4)

- The development of Uhlig's group eliminated the Helium-4 J-T stage, relying instead on the Joule Thompson Heat Exchangers used previously in wet refrigerators.

- These fridges (example shown right) initially used 10 K Gifford-McMahon coolers, and later small 4 K pulse tube coolers.

- Important to note that 4-K class cryocoolers were not available until 1999-2000. Much of the early work was carried out with 10-K coolers.
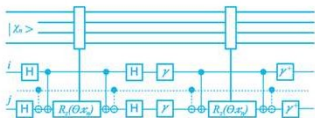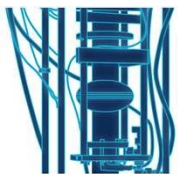
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"Dry" vs "Wet" Refrigerators

"Dry" Process

Cooled by Pulse Tube to ~50 K
⬇
Cooled by Pulse Tube to ~4 K
⬇
Cooled and partially liquified by JT
⬇
Cooled and liquified by Still
⬇
Mixing Chamber

"Wet" Process

Cooled by Liquid Helium to ~4 K
⬇
Cooled and liquified by 1-K Pot
⬇
Cooled by Still
⬇
Mixing Chamber

"Dry" Process

"Wet" Process

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Early commercial dry fridges

- Early examples of commercial cryogen-free fridges came from Leiden Cryogenics and Air Liquide in the early 2000s

- The Leiden refrigerator, built for the SCUBA-2 astronomical camera (right) is still operational at 14000 feet at the James Clerk Maxwell Telescope in Hawaii.

- Limitations of these early designs were often poor heat exchange at 4 K on the second stage of the pulse tube cooler.

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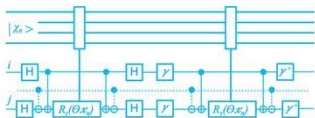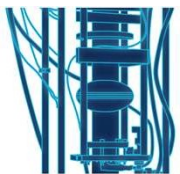
Improvements to the precooler design

- An interesting property of the pulse tube cooler was measured in the early 2000s in that excess cooling capacity on the regenerator between the two stages exists.

- Some heat can be dissipated on the regenerator with little impact on the 4 K cooling performance.

- Adding a coil to the regenerator provides a useful additional heat sink for the inflowing 3He, reducing the cooling requirements at the 4-K stage.
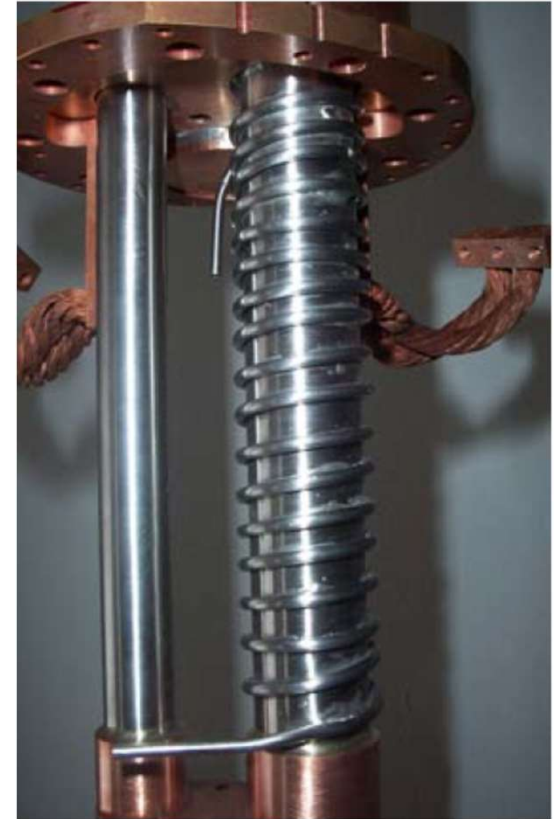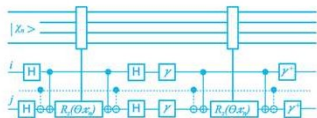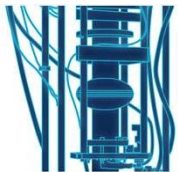

Additional heat exchanger
(Uhlig, personal communication)

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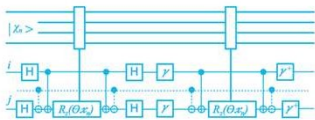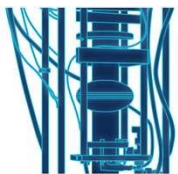
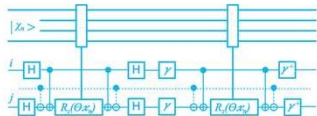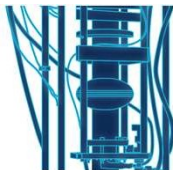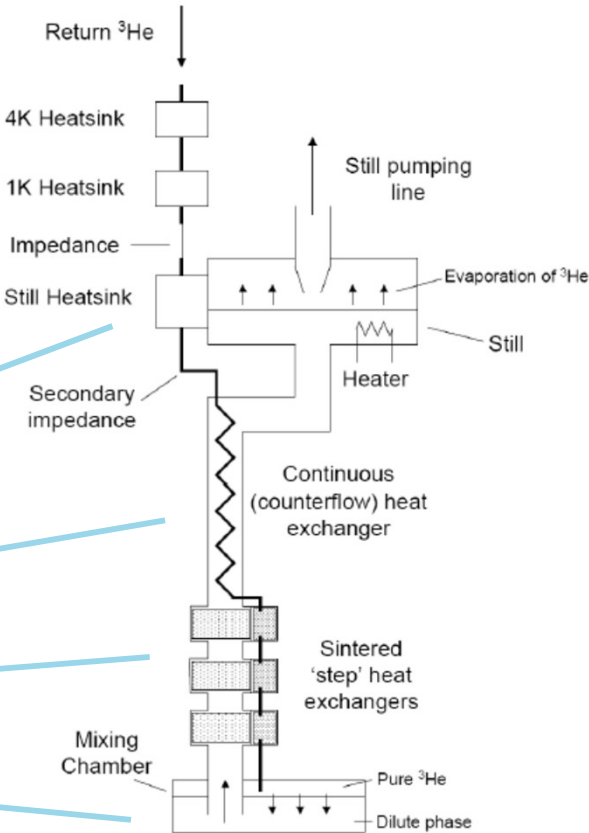# Modern Dilution Refrigerator Design and Operation

Modern refrigerator layout
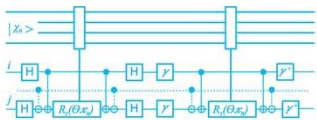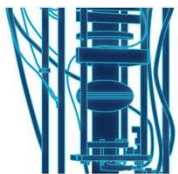
2 stage pulse tube cooler

Additional heat exchanger

Image: Oxford Instruments

Return ³He

4K Heatsink

1K Heatsink

Impedance

Still Heatsink

Secondary impedance

Still pumping line

Evaporation of ³He

Still

Heater

Continuous (counterflow) heat exchanger

Sintered 'step' heat exchangers

Mixing Chamber

Pure ³He

Dilute phase

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Modern refrigerator layout – temperature stages and dilution unit

Still

ICP

Mixing
Chamber

50 K (First stage pulse tube)

4 K (Second stage pulse tube)

1 K (Still)

0.1 K (Intermediate cold plate)

0.01 K (Mixing chamber)
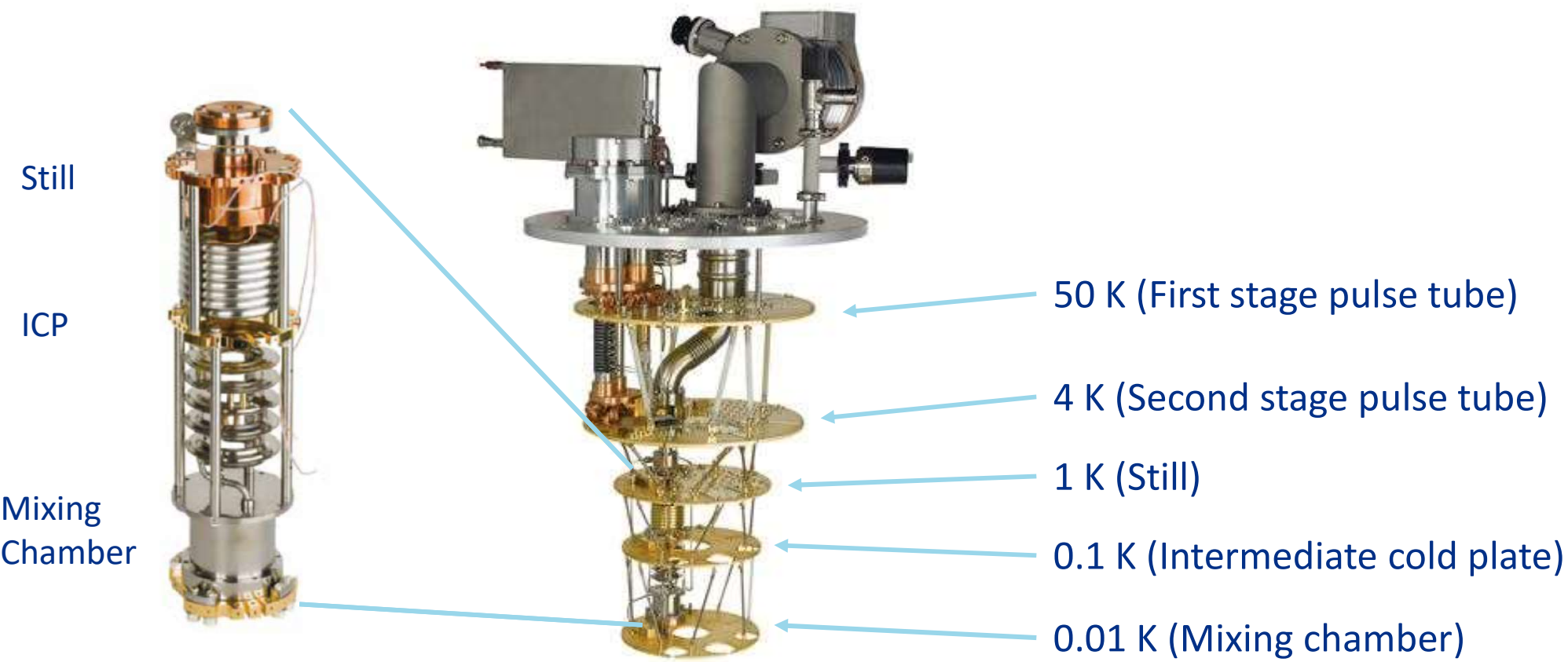
Images: Oxford Instruments

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Commercial fridge examples (1)

Image: Oxford Instruments

Image: Oxford Instruments
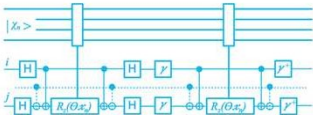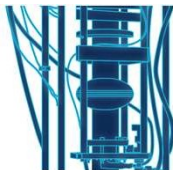
Small fridges
Mixing chamber diameter ~150mm
Minimum temperature ~25 mK

Standard fridges
Mixing chamber diameter ~300mm
Minimum temperature ~10 mK

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Commercial fridge examples (2)

Image: BlueFors

Image: Leiden Cryogenics

Large frame fridges
Mixing chamber diameter ~500mm
Minimum temperature ~10 mK
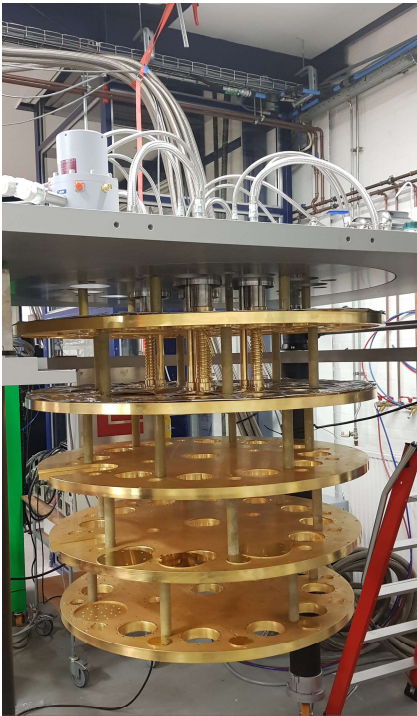
"XXL" fridges
Mixing chamber diameter ~1000mm
Minimum temperature ~10 mK

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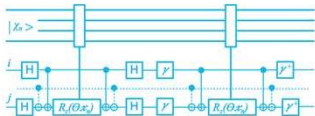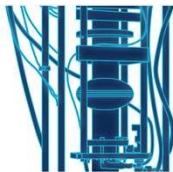
Operational Considerations (1)
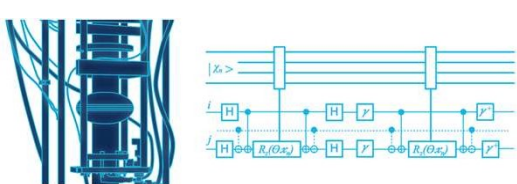
Symptom: High condensing pressure

Possible causes:

1. Blockage in the input lines
2. Blockage in the cold trap
3. High temperature on the condensing stages, either the 4-K stage or the Still

Symptom: Slow cooling, particularly during condensation

Possible causes:

1. Large heat capacity
2. Poor thermal connection

Operational Considerations (2)

Symptom: Low circulation rate/pressure

Possible causes:

1. Low still temperature

2. If combined with high still temperature, possibly too much or too little mixture

Symptom: High mixing chamber temperature

Possible causes:

1. Heat leak or thermal short

2. If combined with weak response to heat input, Quantity of Helium-3 may be too low (phase boundary is not in the mixing chamber).

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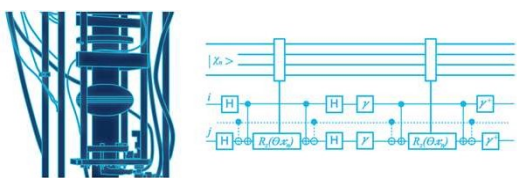
Operational Considerations (3)

- Some general operational comments:

  - Monitor the system behavior run-to-run. Differences in temperatures, pressures and performance can be useful indicators of problems.
  - Monitor how much mixture you are recovering at the end of the cool down, useful to monitor for leaks.
  - Be aware of how much contamination is accumulating in the cold traps between runs since this can indicate the presence of an air leak into the system.
  - Be aware of recommended maintenance internals on pumps. Can be a source of performance issues or leaks.
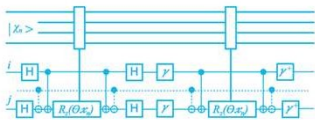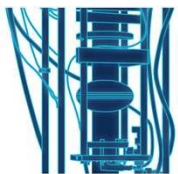
Future Developments

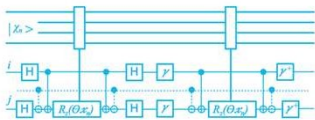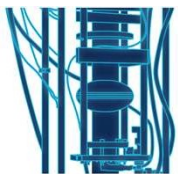Hollister | Dilution Refrigerators for Quantum Science

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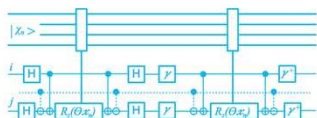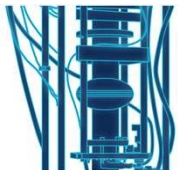
Larger and more powerful fridges

- Drive has been towards larger cold volumes and higher cooling power to accommodate more wires and cryogenic components.
  - Recently, 1-meter class fridges have started to come on the market.
  - Are Fermilab, we are constructing a 2-meter refrigerator.

- Higher cooling power is challenging due to the need to make larger heat exchangers.
  - This can be overcome by making several dilution circuits operating in parallel to increase the available cooling on a common mixing chamber plate.
  - Issues with cooling power at the higher temperature stages could be addressed by using liquid cryogens rather than closed-cycle coolers. Not practical for small labs, but for large centers with multiple platforms having a liquid helium plant becomes viable.

# Summary, Concluding Remarks, References
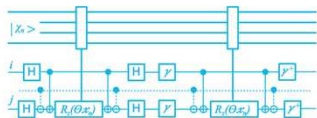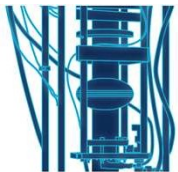
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Summary

- We have reviewed:
  - The physics or Helium-3/Helium-4 mixtures and the dilution process
  - Practical aspects of constructing a refrigerator and the design of key components
  - The history of development of modern, cryogen-free refrigerators
  - Current state-of-the-art
  - Operational considerations and trouble shooting for common problems
 - Possible future developments
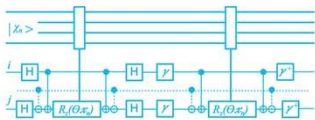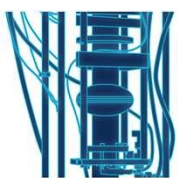
Final Remarks

- Thank you for your attention. Hopefully, you've found this to be useful.

- This is a very in-depth subject, and we've only taken a very brief tour through the processes and engineering associated with these machines.

- Theoretical knowledge is great, but there's no substitute for practical experience!

- If anyone has questions or would like to discuss any of these points further, please reach out to me during the conference via Whova.

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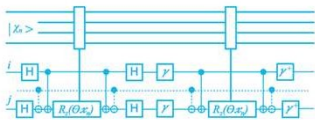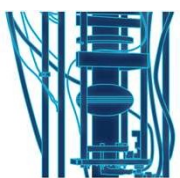
Useful References

- "Matter and Methods at Low Temperatures", Frank Pobell – Very good general textbook for low temperature physics

- "Experimental Principles and Methods Below 1K", O.V. Lounasmaa – Good description of the dilution refrigeration process, although was published in 1974 so is out of date on the technology

- "Experimental Techniques for Low-Temperature Measurements", Jack Ekin – Not so much about the cooling techniques themselves, but has a wealth of cryogenic reference data
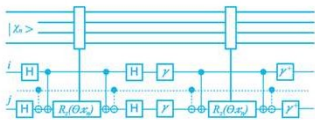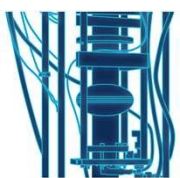
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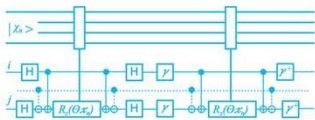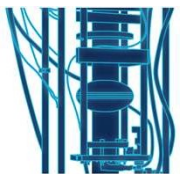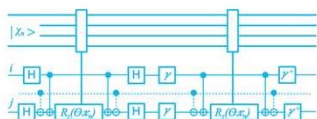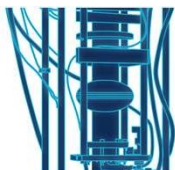
Commercial Manufacturers of Dilution Refrigerators

Provided for reference only. Not necessarily complete, and certainly not an endorsement of products from any one vendor.

- BlueFors Cryogenics (https://bluefors.com)
- Chase Research Cryogenics (https://www.chasecryogenics.com)
- CryoConcepts (https://cryoconcepts.com)
- Entropy Cryogenics (https://www.entropy-cryogenics.com)
- Innovative Cryogenic Engineering (https://www.iceoxford.com)
- Janis ULT (https://www.janult.com)
- Leiden Cryogenics (https://leiden-cryogenics.com)
- Oxford Instruments (https://www.oxinst.com)

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Acknowledgement