Principles of dilution refrigeration

A brief technology guide

The Business of Science®

About the authors

Dr Graham Batey

Chief Technical Engineer
United Kingdom

Graham completed his PhD in Low Temperature Physics at Nottingham University in 1985 and joined Oxford Instruments designing top loading plastic dilution refrigerators to run in pulsed magnets, a rotating dilution refrigerator, and dark matter systems installed deep underground. He was involved in designing Oxford Instruments' **Kelvinox**™ range of dilution refrigerators and the world's leading cryogen free range of dilution refrigerators – **Triton**™, which in 2010 received the Queen's award for innovation. In 2011, Graham received 'The Business & Innovation' award from the Institute of Physics and was elected Fellow of the IOP.

Dr Gustav Teleberg

ULT Product Manager
Sweden

Gustav Teleberg gained his PhD in cryogenics for astronomy at Cardiff University where he developed miniature dilution refrigerators and heat switches for telescope applications. He joined Oxford Instruments in 2007 as a Cryogenic Engineer where he worked on developing the Cryofree® dilution refrigerator that today is known as **Triton**™. He also worked on several patented technologies for rapid sample exchange and heat pipes for accelerated cooling.

Principles of dilution refrigeration

By Graham Batey and Gustav Teleberg

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History

The dilution refrigerator was first proposed by Heinz London in the early 1950s, and was realised experimentally in 1964 at Leiden University. In 1965, Henry Hall built the first ^{3}He-^{4}He dilution refrigerator at University of Manchester. The first commercial dilution refrigerator was developed in collaboration with Heinz London at the Oxford Instruments factory in Osney Mead, Oxford in 1966. The achieved 200 mK base temperature enabled dilution cooling beyond known ^{3}He temperatures.

The dilution refrigerator has evolved over its 50 years with improved performance, reliability and ^{3}He efficiency. Today, temperatures below 4 mK can be achieved with the push of a button and without the use of any external liquid helium cooling.

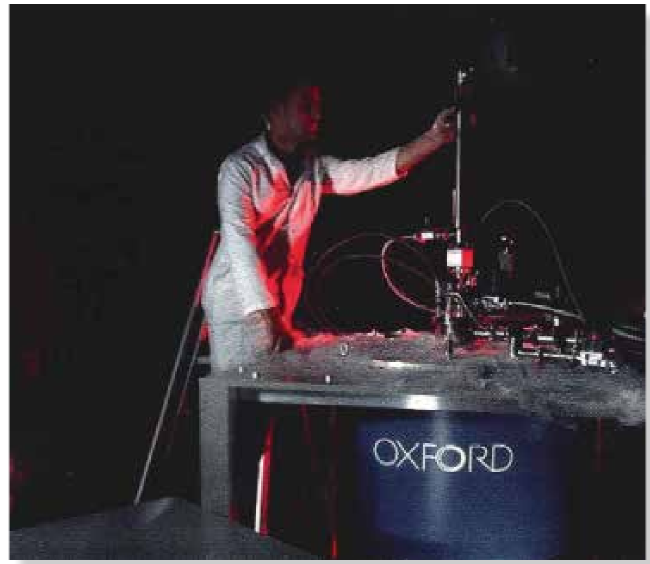
Sir Martin Wood with the mixing chamber of the first commercial dilution refrigerator, built at Oxford Instruments

First commercial dilution refrigerator insert

Early 70's dilution unit with copper heat exchanger

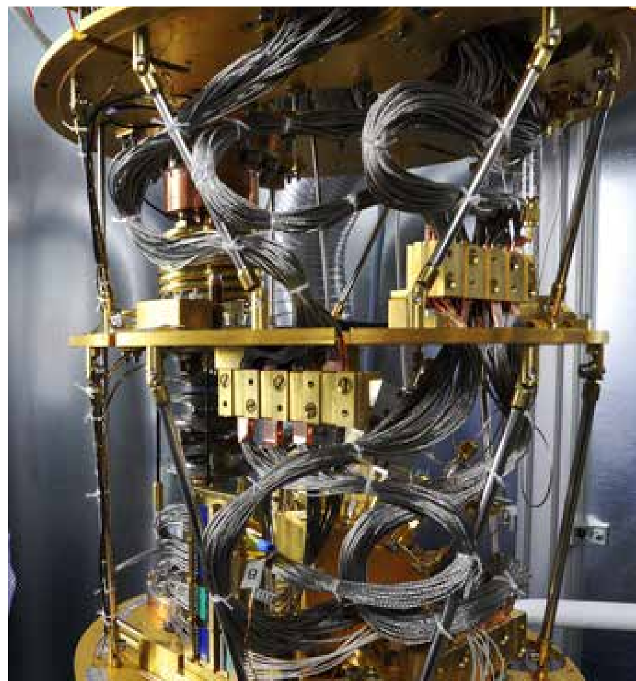
Modern dilution unit with silver heat exchanger

Gravitational wave detector - photo courtesy of Gran Sasso, University of Rome



High magnetic field laboratory - photo courtesy of Bristol University

Centre for quantum devices - photo courtesy of Niels Bohr Institute, Copenhagen



D-Wave quantum processor - photo courtesy of D-Wave Systems

Over the years, dilution refrigerators have been used for cooling anything from millimeter-sized semiconductors to 3 ton gravitational wave detectors, down to milliKelvin (mK) temperatures. Superconducting magnets with field strengths of up to 50 Tesla (T) have been combined with these refrigerators for the study of novel superconductors. Currently, dilution refrigerators are used by scientists trying to realise quantum computers, searching for the hypothesised Majorana particle, and in the study of many other novel quantum properties.

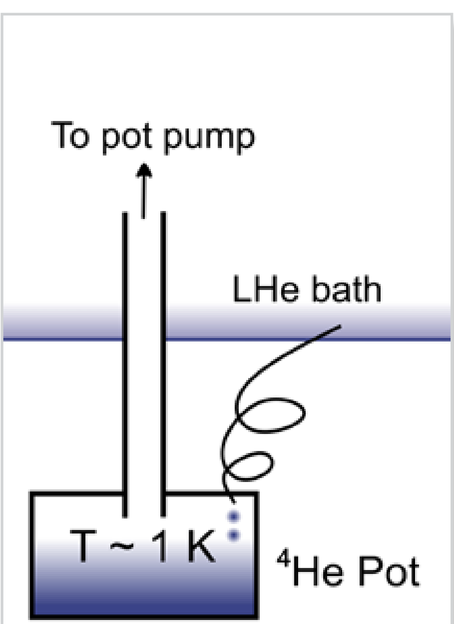
4 K technology

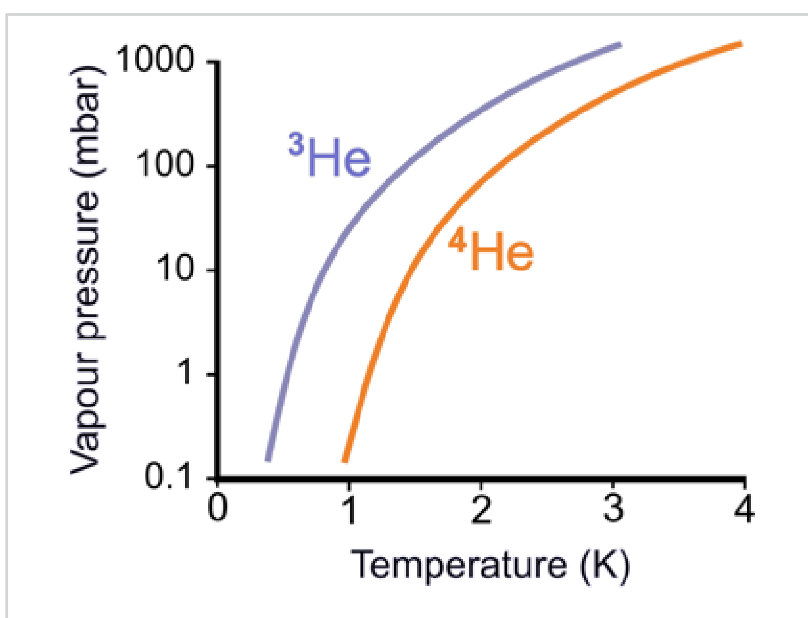
- All dilution refrigerators rely on a stable cold environment at approximately 4 K. Until recently, this has meant a liquid helium bath with a boiling point of 4.2 K at atmospheric pressure
- Now, it is more common to use a pulse tube cooler which greatly simplifies operation and is less costly to operate
- A two-stage pulse tube cooler typically provides 50 W of cooling power at 70 K on its first stage and 1 – 1.5 W at 4 K on its second stage. The power consumption of its compressor is typically 9 kW
- 'Dry' dilution refrigerators based on pulse tube technology now account for 90% of all systems sold. 'Wet' systems are still used but mainly at the extremes of vibration sensitive applications, such as STM

Evaporative cooling of ^4He

We start by describing the principle of evaporative cooling as this is essential also for dilution refrigeration:

- A small amount of liquid helium from the main bath continuously feeds a 'pot' (below left)
- A pump is used to reduce the pressure inside the pot to typically 0.1 mbar
- Cooling is achieved due to the latent heat of the evaporated liquid
- The lowest achievable temperature is limited by the exponentially decreasing vapour pressure. At low enough pressure there are simply not enough molecules to cool the pot further
- Pumping on liquid ^4He is the easiest method to achieve around 1.2 K. This is already colder than outer space (2.7 K)
- Temperatures down to around 0.25 K can be achieved by pumping on liquid ^3He instead, since it has much higher vapour pressure than ^4He





^3He vs ^4He

How is liquid ^3He different to liquid ^4He?

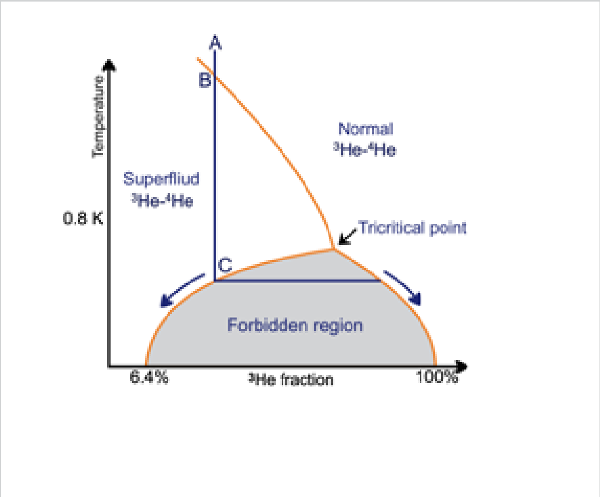
- ^3He is lighter than ^4He which means the binding energy between the atoms in the liquid is weaker. This results in a lower latent heat of evaporation
- The lower latent heat of ^3He results in a higher vapour pressure (p) since: $p \propto \exp(-L/RT)$
- ^3He consists of 3 nucleons (L), which makes it a Fermion while ^4He with 4 nucleons is a Boson. As we shall see, this difference is fundamental for dilution refrigeration

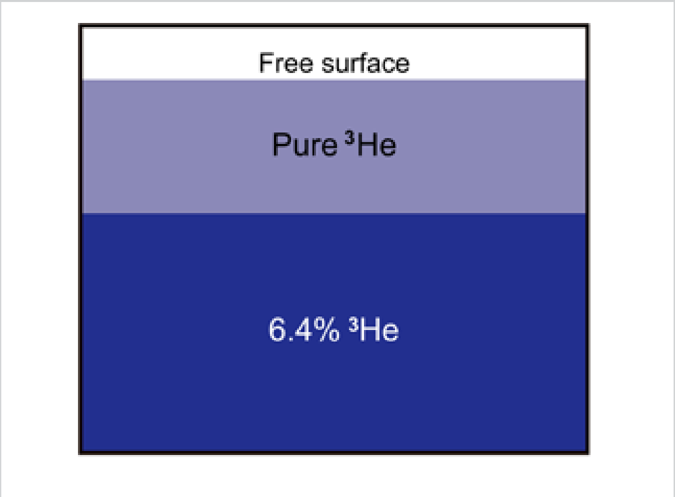
Where does ^3He come from?

- While ^4He can easily be extracted from natural underground reserves, ^3He is a much rarer isotope which is produced as a by-product from nuclear reactions (tritium decay or deuterium-deuterium fusion reaction)
- The majority of all ^3He gas available is used for security detector applications
- Approximately 10% is used for ultra low temperatures applications

^3He - ^4He mixtures

- Pure ^4He, with a nuclear spin of I = 0, obeys Boson statistics and undergoes a transition to superfluid at 2.17 K
- Pure ^3He, with a nuclear spin of I =1/2, obeys Fermi statistics and the Pauli Exclusion principle which prevents ^3He from undergoing a superfluid transition until much lower temperatures at which the spins pair up and then obey Boson statistics
- The superfluid transition temperature of a ^3He - ^4He mixture depends on the ^3He concentration as in the figure below left
- Say we start with a mixture at a concentration and temperature at point A. When this is cooled down to the temperature at point B, it undergoes a superfluid transition. If we cool the mixture further to point C, it seperates into two phases with the ^3He-rich phase floating on top of the heavier ^4He-rich phase (below right)
- The ^4He-rich phase (the 'dilute' phase) contains 6.4% ^3He all the way down to 0 K. This finite solubility of ^3He in ^4He is the key to dilution refrigeration

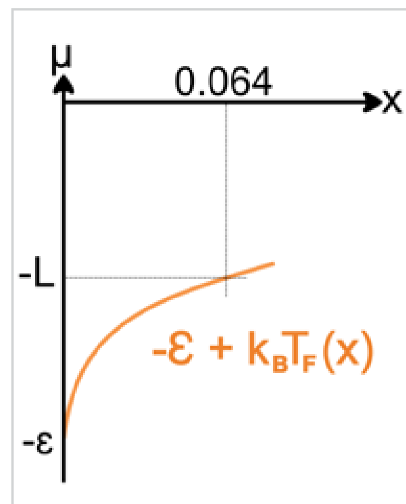




Finite solubility of ^3He in ^4He explained

Imagine we could cool two containers of pure ^3He and pure ^4He separately to 0 K before they are allowed to interact. The first ^3He atom to venture across the boundary will consider whether to stay in the ^4He environment or to go back.

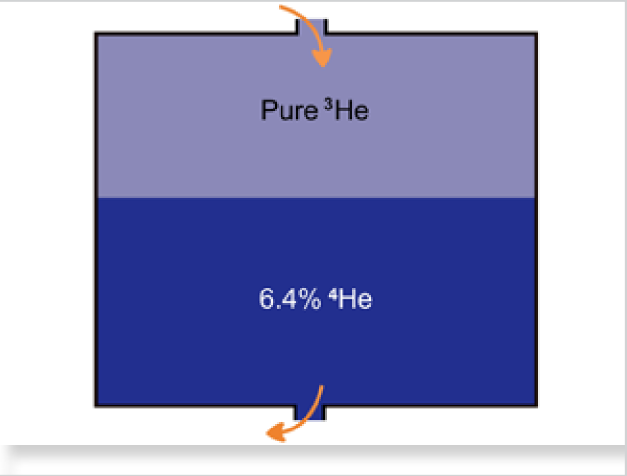
- ^3He is the lighter of the two isotopes. This means it has a larger zero-point motion, in other words it will occupy a larger volume
- The ^3He atom will find itself closer to surrounding ^4He atoms than to surrounding ^3He atoms
- Because the binding between the atoms is due to van der Waals forces, the shorter distance results in ^3He being more strongly bound in ^4He than in pure ^3He
- Since the ^3He atom is more strongly bound in ^4He it will 'prefer' to stay in the ^4He liquid. This is the reason for the finite solubility at 0 K
- To understand why the finite solubility is 6.4%, we need to look in more detail at the binding energy. Consider the chemical potential μ of ^3He in ^4He and let's say the binding energy of a single ^3He atom in ^4He is **ε**
- The binding energy of ^3He in ^3He is equal to the latent heat of evaporation of pure ^3He, L, so **ε** must be greater than L as shown in the figure below right
- The first two ^3He atoms will occupy the lowest energy state – **ε** with anti-parallel spins
- Additional ^3He atoms have to obey the Pauli Exclusion principle and therefore occupy increasingly higher energy states (described by the wave function solutions to the Schrodinger equation of a 'particle in a box')
- The Fermi energy $k_B T_F$ will increase with the He concentration x, as shown in the figure
- At a concentration of 6.4%, the chemical potential equals that of a ^3He atom in pure ^3He. This is therefore the finite solubility of ^3He in ^4He at 0 K



Cooling power of the dilution process

- If we can remove ^3He atoms from the diluted phase, ^3He atoms from the concentrated phase will cross the phase boundary to occupy the vacant energy states
- The associated cooling power is then given by the enthalpy difference ΔH between ^3He in diluted ^4He and pure ^3He multiplied by the ^3He flow rate:

$$\dot{Q} = \dot{n}_3 \Delta H = 84 n_3 T^2$$

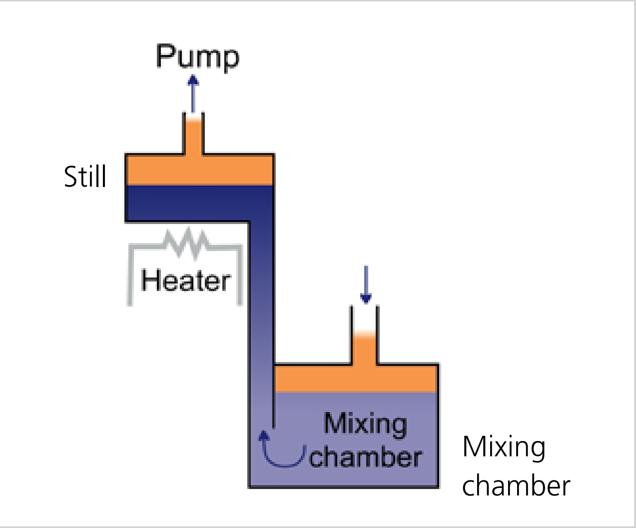


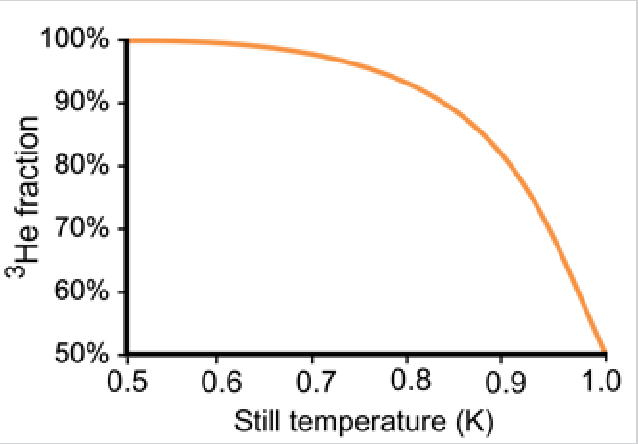
- The enthalpy of ^3He in ^4He is higher than for pure ^3He due to ^3He in ^4He behaving as a Fermi gas. This is analogous to the enthalpy difference between ^3He gas and ^3He liquid that results in cooling power in an evaporation refrigerator
- In reality the returning ^3He is always slightly warmer than the outgoing ^3He due to non-ideal heat exchangers. A more accurate cooling power expression therefore takes into account the temperature of the mixing chamber (T_{mc}) as well as the temperature of the last heat exchanger (T_{ex}):

$$\dot{Q} = \dot{n}_3 (95 T_{mc}^2 - 11 T_{ex}^2)$$

How to remove only ^3He from diluted ^4He

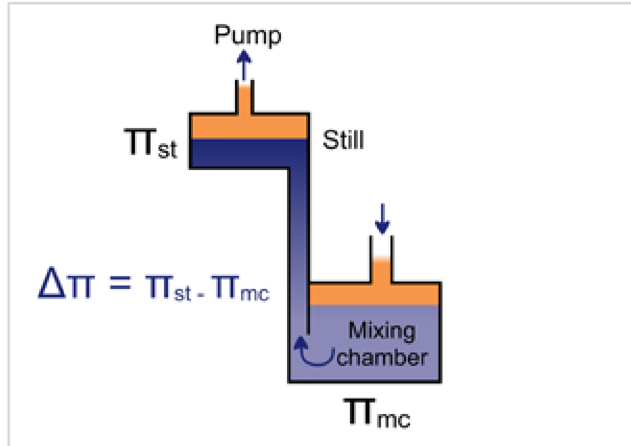
- The mixing chamber connects to a distiller ('still'), which distils the ^3He from the ^4He due to the difference in vapour pressure
- Heat is applied to the still (otherwise it will quickly cool to a temperature where the vapour pressure is so low that the circulation stops)
- More power to the still means a higher circulation rate, which means more cooling power
- On the other hand, if the still temperature is too high, the vapour pressure of ^4He will become significant. Circulating too much ^4He will reduce the dilution process efficiency
- In practice, a ^3He fraction of ~90% in the circulated gas is acceptable, resulting in an optimal still temperature of 0.7 - 0.8 K. The approximate relationship between still temperature and ^3He fraction is shown in the figure (right)





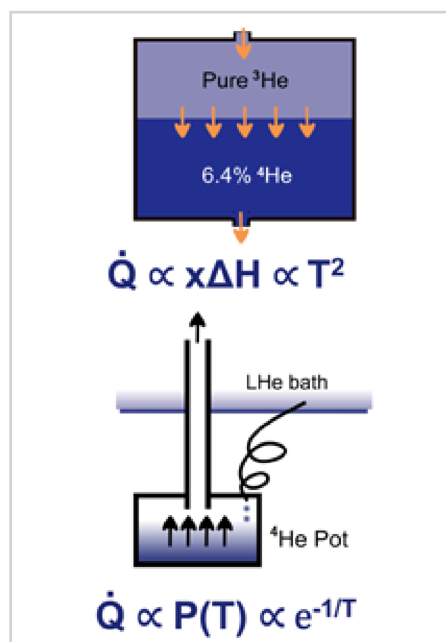
The osmotic pressure

- As we pump ^3He vapour from the liquid inside the still, the ^3He concentration in the liquid will decrease
- The difference in ^3He concentration between the still and the mixing chamber results in an osmotic pressure gradient along the connecting tube
- This osmotic pressure pulls ^3He from the mixing chamber
- Note: Because ^3He in ^4He behaves as a Fermi gas, the osmotic pressure difference $\Delta\pi$ can be understood from the ideal gas law (or more correctly Hoff's law), from which it follows that the maximum osmotic pressure is almost 20 mbar, equivalent to the hydrostatic pressure of 1 meter of liquid helium. This means the osmotic pressure difference is large enough to 'suck' ^3He from the mixing chamber even if they are separated by a vertical difference of 1 meter



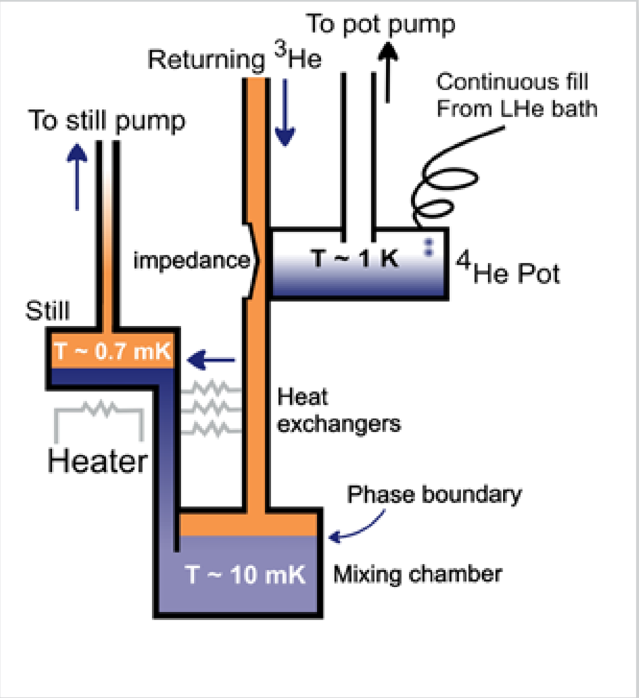
Dilution refrigeration vs evaporative cooling

- The ^4He component of a dilution refrigerator is effectively just a static background which is there to enable the ^3He to transition from its pure liquid state and into a Fermi gas
- The 'dilution process' of ^3He moving across the phase boundary is equivalent to an upside-down evaporator
- The importance of the finite solubility of ^3He in ^4He should now be clear. While the lowest achievable temperature in an evaporator is limited by exponentially decreasing vapour pressure, the equivalent to vapour pressure in a dilution refrigerator (the concentration of ^3He in the Fermi gas) is constant with temperature
- The lowest achievable temperature is limited by the much weaker temperature dependence of the enthalpy of Fermi liquids (T^2)



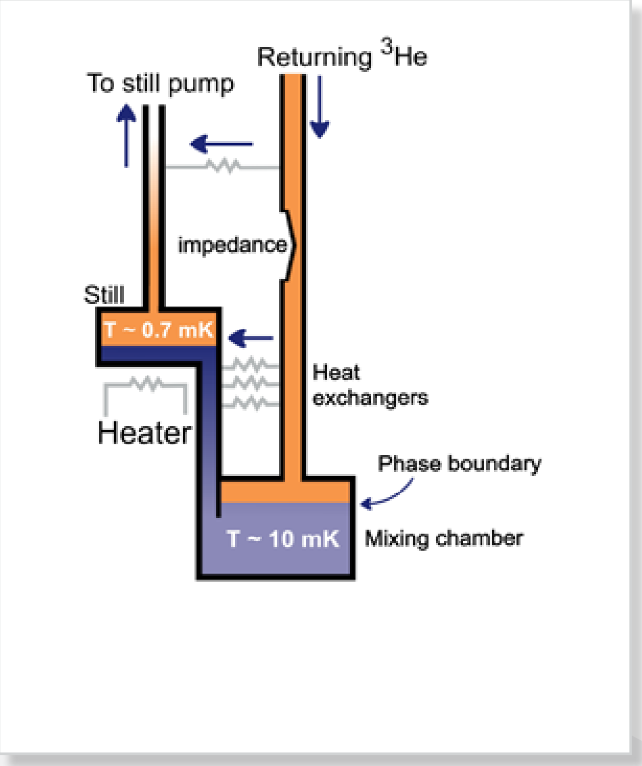
'Wet' dilution refrigerator

- A small amount of liquid helium from the main bath feeds the 1 K pot
- A dedicated pot pump keeps the 1 K pot cold
- An impedance ensures the condensing pressure is high enough (~0.3 bar) for the returning ^{3}He to liquefy
- Several different types of heat exchanger precool the ^{3}He before it reaches the mixing chamber
- Dilution cooling occurs in the mixing chamber
- The osmotic pressure drives the ^{3}He up to the still where it is separated from the ^{4}He
- A room temperature gas handling system and pumps are used to recycle the ^{3}He back into the return line



'Dry' dilution refrigerator

- The 1 K pot has been replaced by an extra heat exchanger located just before the impedance. The returning ^{3}He is cooled by the outgoing ^{3}He vapour from the still
- This heat exchanger in combination with the Joule-Thompson (JT) expansion happening in the impedance is enough to condense the gas
- The JT stage is not as efficient as a 1 K pot, so the condensing pressure is higher: typically 2.5 bar during the initial condensation, which drops to ~0.5 bar near base temperature where the circulation rate is lower
- The higher condensing pressure means an additional high pressure pump (or compressor) is required when circulation is started. It can be switched off when the system has reached base temperature



Helium 3 circulation pumps

Either a turbo pump or a roots pump can be used at the low pressure end. Turbo pumps are oil-free, which means less risk of introducing contamination.

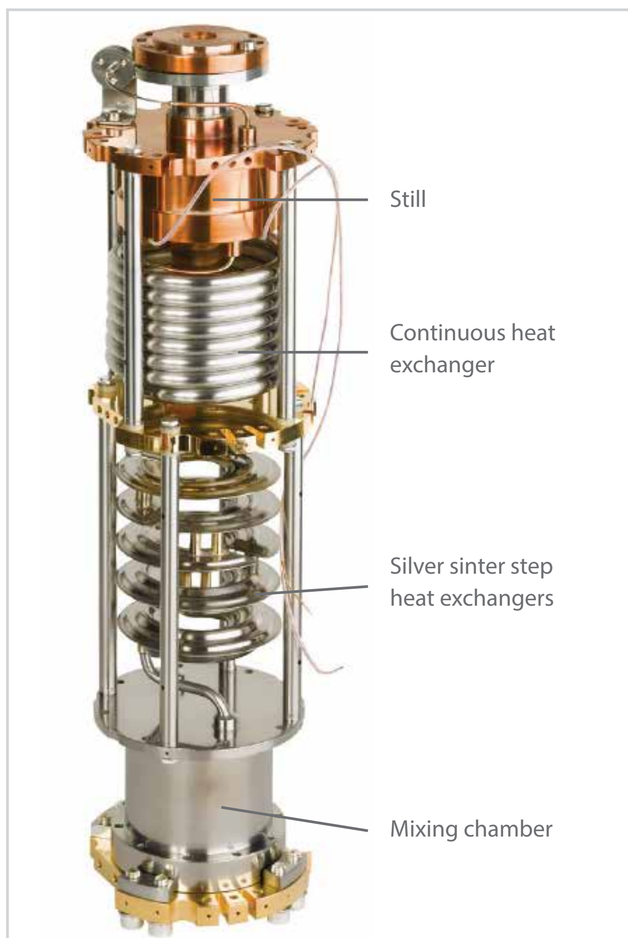
Either a rotary pump or a scroll pump can be used as a backing pump. Rotary pumps are cleaner (scroll pumps generate dust which may cause blockage or damage other pumps).

Either a diaphragm pump or a scroll pump can be used at the high-pressure end. A diaphragm pump is the cleanest option and it can generate a higher outlet pressure (4 bar as oppose to only 1.5 bar). This reduces the condensing time.

The dilution unit

The dilution unit is the heart of the system. It has five main design criteria:

- Minimise the effect of thermal resistance between liquid helium and metals (Kapitza resistance). This enables efficient heat exchangers and therefore a lower base temperature
- Minimise the effect of viscous heating. This enables a high ^{3}He circulation rate and therefore a higher cooling power
- Limit the superfluid film flow in the still. This ensures about 90% pure ^{3}He is being circulated
- Minimise the amount of ^{3}He required for operation reducing cost and preserving this rare and limited resource
- Remain leak tight for many years



Dilution unit for operations below 10 mK

Low temperature properties of helium

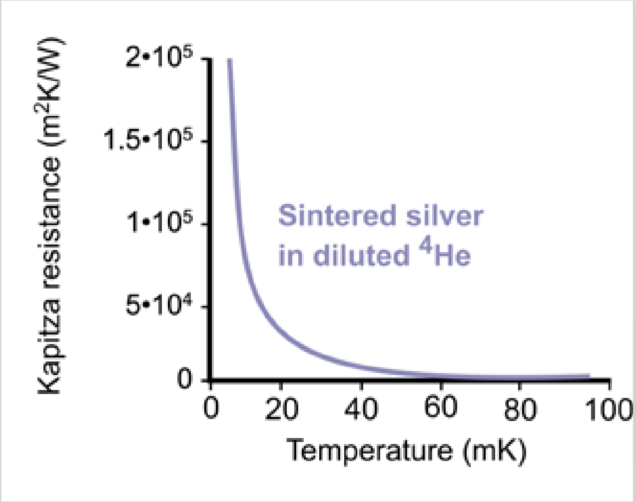
The main engineering challenges when building a refrigerator are due to the following thermal effects:

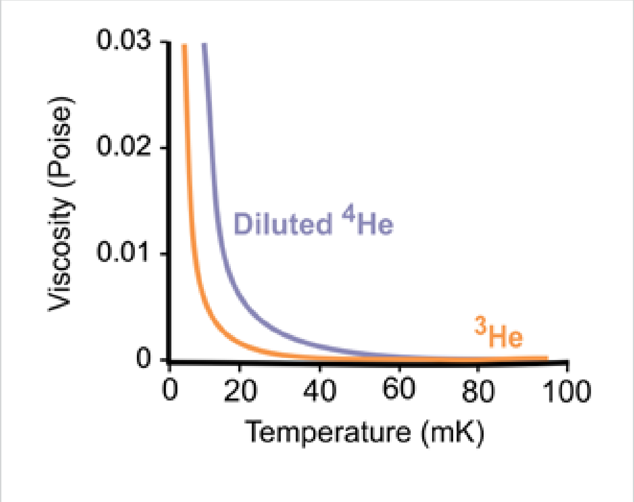
Kapitza resistance

- The thermal resistance is inversely proportional to T^3, so as we approach T= 0 K, designing heat exchangers become more of a challenge
- The solution is to increase the surface area by using sintered silver – typically 1 m^2/g of silver

Viscous heating

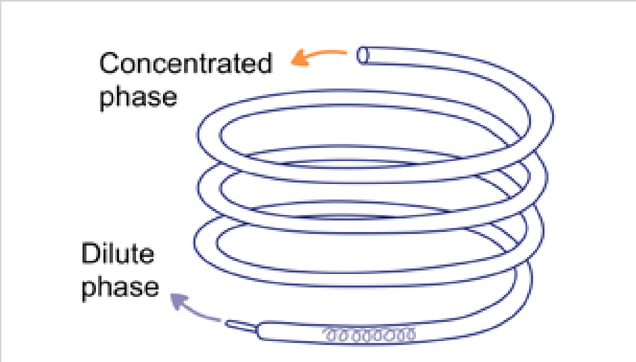
- The viscosity of ^3He increases with lower temperature and the viscosity of ^3He in the diluted phase is 8 times higher than in the concentrated phase
- The friction associated with a viscous fluid flowing through a small channel results in heating, which must be minimised
- The solution is to increase the diameter of the flow channels
- The challenge of course is that larger flow channels conduct more heat and will also need more expensive ^3He



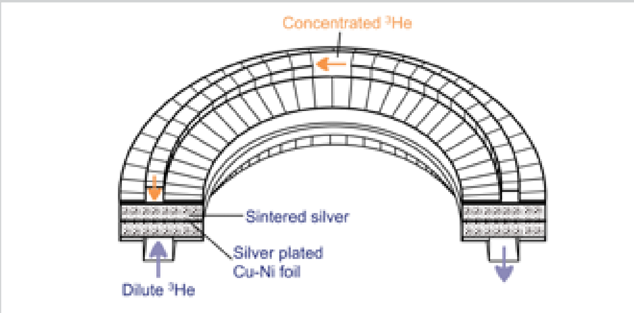


Heat exchangers

- A continuous tube-in-tube heat exchanger is used at the high temperature end where Kaptiza resistance and viscous heating only plays a limited role. It is ^3He efficient and works well down to around 30 mK
- Several step heat exchangers with sintered silver are used at the low temperature end. The size of the flow channels and the amount of silver is increased in each step towards lower temperatures. Most of the ^3He used in a dilution refrigerator is required for the step heat exchangers



Coil heat exchanger (used at higher temperatures)

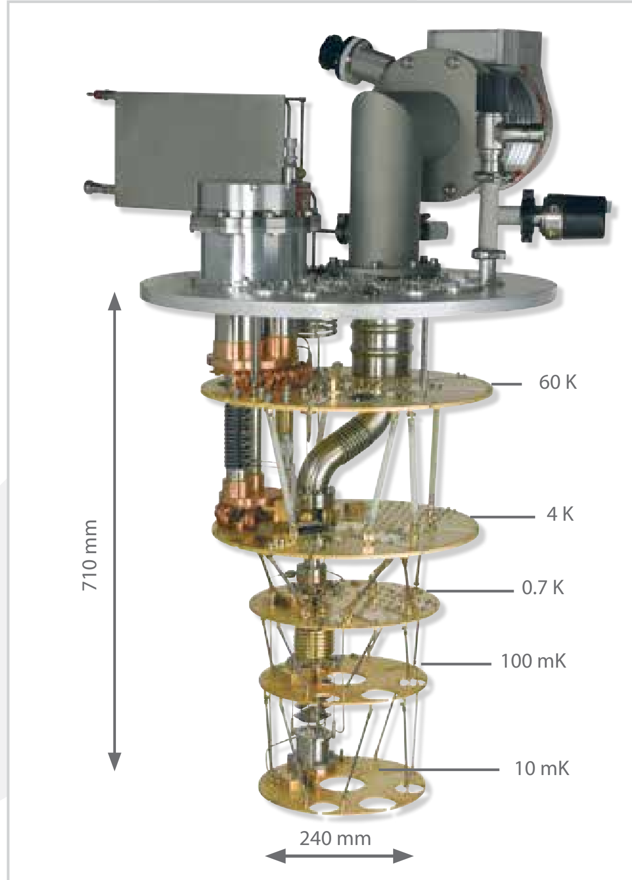


Sintered silver heat exchanger (used at the lowest temperature)

The dilution refrigerator assembly

The photograph shows a typical 'dry' dilution refrigerator assembly. Typical experimental requirements to consider are:

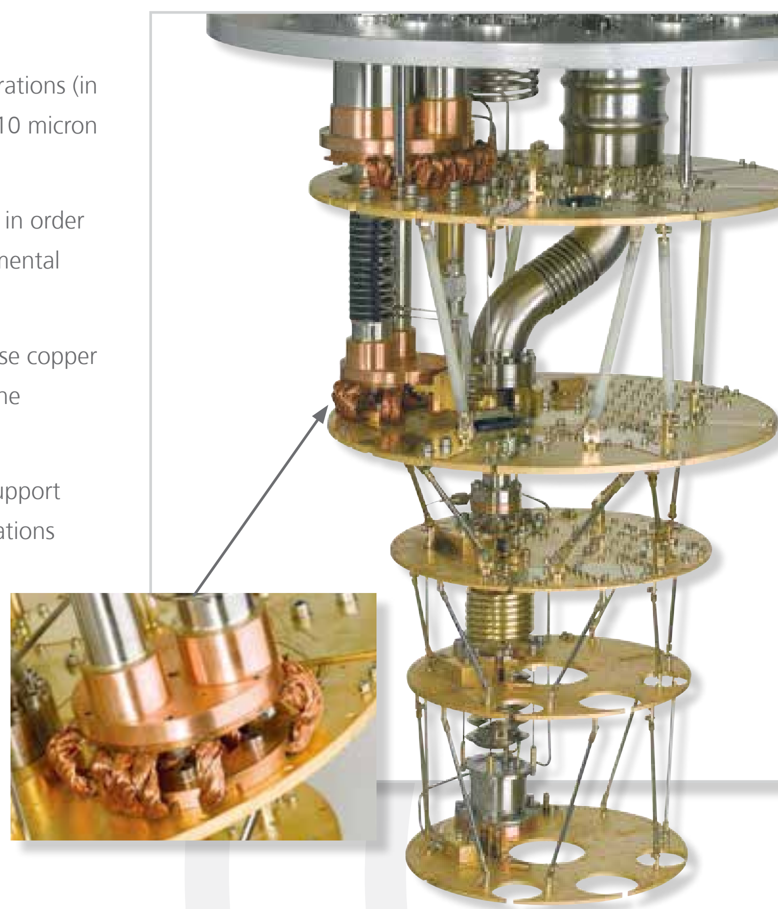
- Vibrations from the pulse-tube cooler reduced to <100 nm on the experimental plates
- Total system cool down time < 24 h
- Gas handling and software for automatic operation
- Integration of semi-rigid coaxes, RF components, DC wiring looms and optical fibres
- Possibility to integrate a superconducting magnet
- Possibility to integrate a sample exchange mechanism to reduce the sample turnaround time



Internal assembly of a Cryofree dilution refrigerator

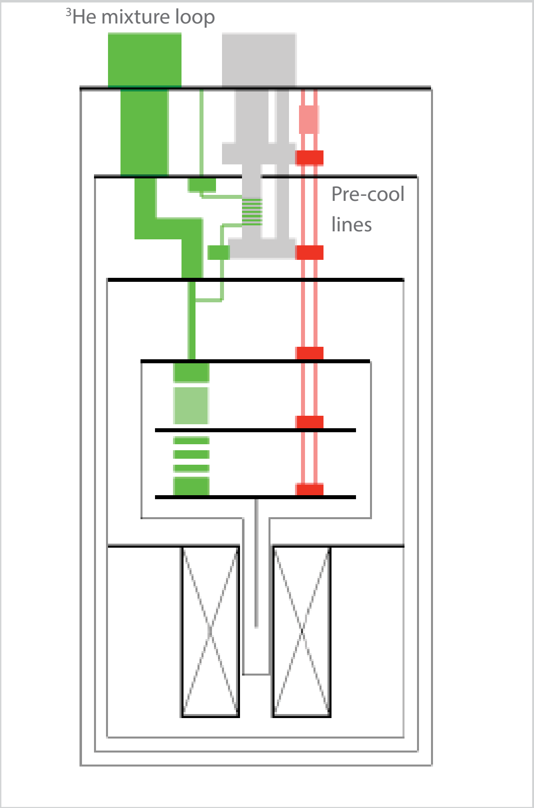
Reducing vibrations

- Pulse tube technology enables relatively low vibrations (in comparison with other cryocoolers): typically 5-10 micron on the cold head stages
- Most experiments require sub-micron vibrations in order to eliminate microphonic noise from the experimental wiring
- The best approach to tackle this problem is to use copper braids to decouple the pulse tube stages from the experimental plates
- Using a combination of braids and a very stiff support structure it is possible to achieve < 100 nm vibrations



Pre-cooling mechanism

- The dilution unit with its three experimental plates need to be cooled from 300 K to ~10 K before mixture condensation can commence
- Wet systems usually enclose the cold stages with an inner vacuum chamber (IVC) which is filled by exchange gas, which is later evacuated
- The pre-cool line (shown in red) removes the need for an IVC. Instead a small amount of mixture is circulated through the pre-cool loop using the membrane pump
- A pre-cool line makes the sample exchange much easier and faster with no need to use indium seals (as on a wet system) and thus reducing risk for developing cold leaks
- Circulating mixture through the low impedance pre-cool line also allows for good temperature control at higher
- Cool down time is reduced by removing the need for the thermal mass of an IVC



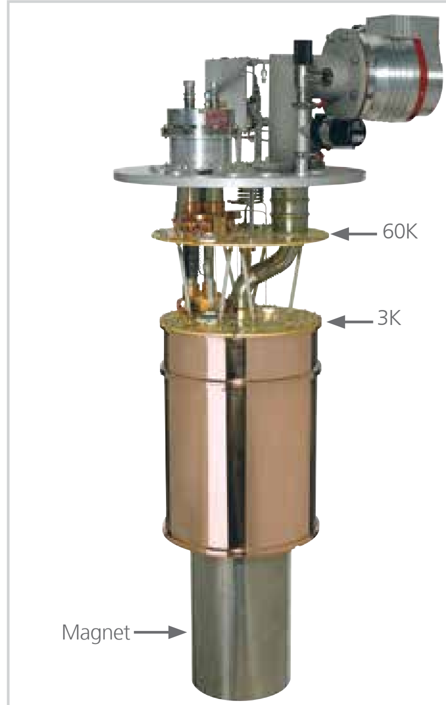
Gas handling and software

Dilution refrigerators need a gas handling system which can automate their operation. Typical requirements to consider are:

- Software for monitoring and logging pressure readings, temperatures and any error messages
- Automatic procedures for cooling and warming the system, condensing and collecting the mixture
- Automatic safety interlocks and pressure-relief valves to protect the system in case of operator error or extended power cuts
- Possible integration of a UPS to allow the software to restore the system in case of a power cut
- A command interface to enable remote system control with, for example, LabVIEW and easy integration with other control languages such as Python, Matlab, C++ and many more

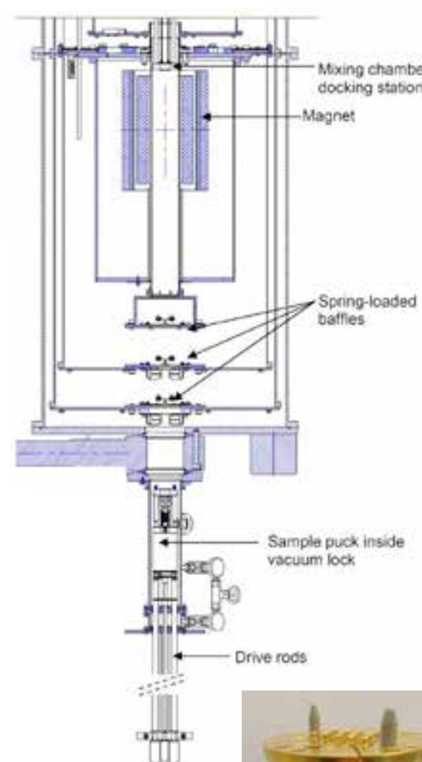
Integrated superconducting magnets

- In order to avoid eddy current heating effects from vibrations in magnetic fields, the superconducting magnet should be cooled by the same pulse tube cooler as that of the dilution refrigerator
- Mounting the magnet and the dilution refrigerator on the same 4 K stage as shown in the photo on the right ensures that any motion will be common to both
- Integrated cryogen free solenoid magnets up to 18 T, split pair magnets with optical access and a variety of vector magnets to tilt or rotate the field with respect to the sample are now commercially available



Sample exchange mechanism

- Large magnets add significantly to the overall system cool down time. A system with an integrated 14 T solenoid typically takes 45 hours to reach base temperature unaided
- A top or bottom loading mechanism allows a new sample to be cooled overnight and also greatly simplifies the manual work involved
- The figure shows the principle of a bottom loading mechanism where a demountable ‘puck’ loads though a series of baffles and attaches to the mixing chamber plate with a bolted contact
- The sample puck in the photograph has provision for up to 14 RF connections plus 50 DC connections
- The wires connecting the sample to room temperature are permanently installed on the fridge platform (and remain cold when changing the sample). This reduces the heat load to the sample and therefore offers best performance
- Some designs use a wiring ‘stick’ with clamps creating thermal contacts to higher temperature stages
- The sample puck method has two significant advantages over the sample stick concept: firstly, having the wiring on the fridge platform means there is much more space available for installling filters or additional RF components. Secondly, because the heat load from wires is dissipated directly on the fridge instead of on the sample holder, the achievable sample temperature will always be lower with a demountable puck than with a wiring stick. The demountable puck is currently the only method able to achieve <10 mK sample temperatures



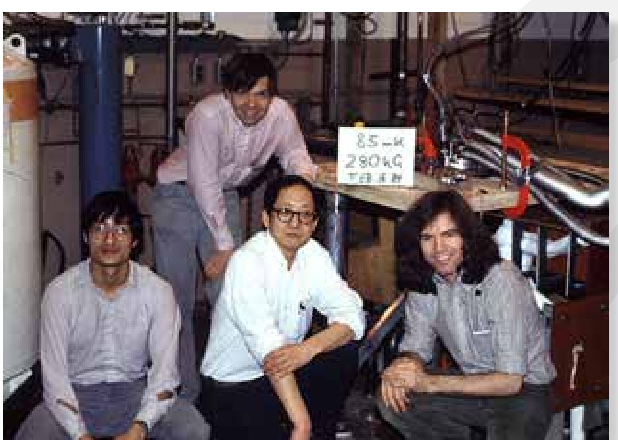
Dilution refrigerators at the leading edge of physics

Dilution refrigerators have been used for a large number of significant scientific advancements in, for example, graphene, topological insulators, quantum dots and superconducting circuits, and quantum computing/quantum information processing (QIP).

Two Nobel Prizes for Physics have been based on experiments using dilution refrigeration.

Superfluidity in ^{3}He was discovered in 1971 by Lee, Osheroff and Richardson using a dilution refrigerator, equipped with a pomeranchuk cooling stage. Their results laid the groundwork for the study of quantum materials that display macroscopic quantum effects. They were awarded the Nobel Prize for Physics in 1996.

The discovery of the fractional quantum Hall effect in 1981 by Daniel Tsui, Horst Störmer and Robert Laughlin was done using a top loading into mixture (TLM) dilution refrigerator. This work, which was awarded the Nobel Prize for Physics in 1998, led to a breakthrough in our understanding of quantum physics and to the development of new theoretical concepts of significance in many branches of modern physics.



Albert Cheng, Peter Berglund, Daniel Tsui and Horst Störmer (from the left)

Robert C. Richardson - © Cornell University photography

Further reading:

The following reference articles and publications contain useful background information about the fundamentals of cryogenics and low temperature systems.

Integration of superconducting magnets with cryogen free dilution refrigerator systems
(doi: 10.1016/j.cryogenics.2009.09.008)
Download the article here

A micro Kelvin Cryofree platform with noise thermometry
(doi: 10.1088/1367-2630/15/11/113034)
Download the article here

A rapid sample-exchange mechanism for cryogen free dilution refrigerators compatible with multiple high frequency signal connections
(doi: 10.1016/j.cryogenics.2014.01.007)
Download the article here

A new ultra low temperature cryogen free experimental platform
(doi: 10.1088/1742-6596/568/3/032014)
Download the article here

Helium Cryogenics (2nd edition): by Steven W. Van Sciver, Springer Press, 2012, ISBN 978-1-4419-9979-5

Matter and Methods at Low Temperatures (3rd edition): by Frank Pobell, Springer-Verlag Berlin Heidelberg 2007, ISBN 978-3-540-46360-3

Magnetic Venture, The Story of Oxford Instruments: by Audrey Wood, Oxford University Press, 2001, ISBN 978-0-19-924108-8

History and Origins of Cryogenics: by Ralph G. Scurlock, Oxford University Press, 1992

An Introduction to Milli-Kelvin Technology: by David S Betts, Cambridge University Press, 1989, ISBN 0-521-34456-5

Experimental Techniques in Condensed Matter Physics at Low Temperatures: by Robert C Richardson and Eric N Smith, Addison Wesley Publishing Company Inc, 1988, ISBN 0-201-15002-6

Experimental Techniques in Low Temperature Physics (3rd edition): by G K White, Oxford University Press, 1987, ISBN 0-19-851381-X (Pbk), ISBN 0-19-851359-3

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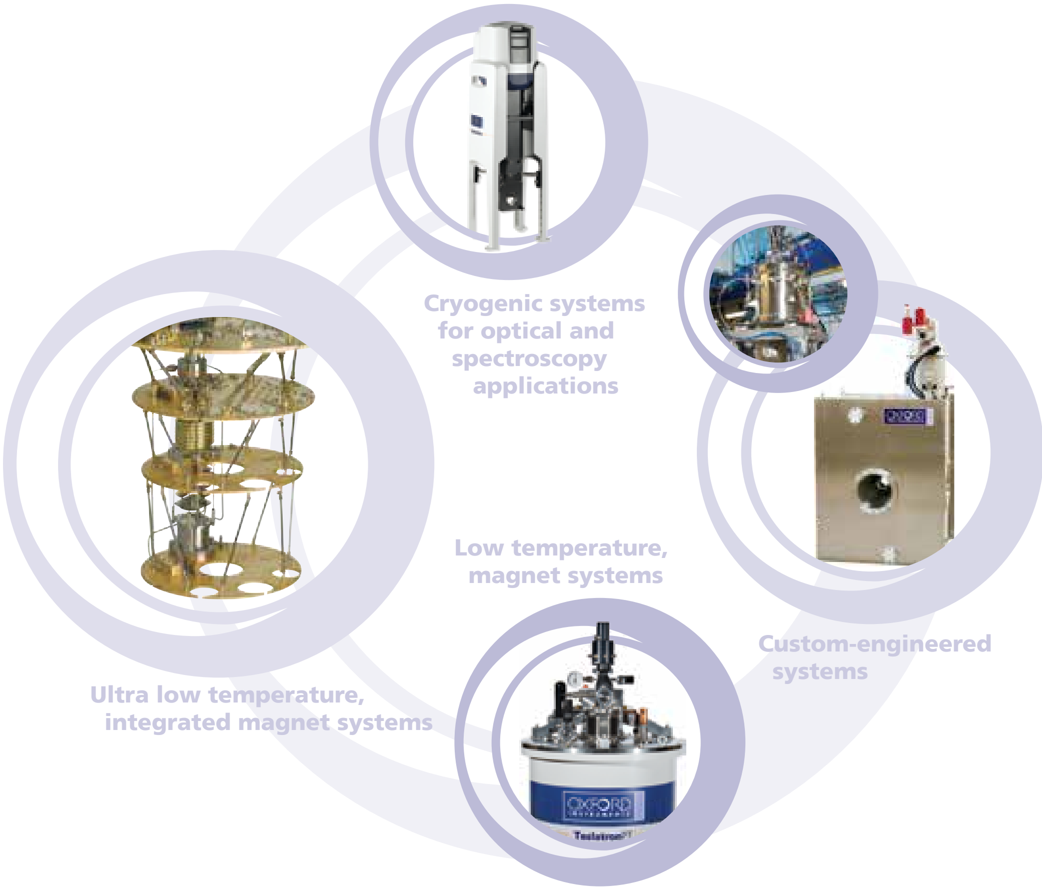

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