Towards the absolute zero

Low-temperature physicists are surpassing nature, using dilution refrigeration, Pomeranchuk cooling and nuclear demagnetization to investigate phenomena hitherto unknown in the universe.

Olli V. Lounasmaa

Low-temperature physicists are constantly trying to extend the temperature range accessible to experimental investigations closer and closer to the absolute zero. While the third law of thermodynamics prevents their ever reaching 0 K, they have long since surpassed Nature herself. The temperature of the cosmic background radiation is 3 K; below 3 K, we are creating new physics, hitherto unknown to the universe. And this has been done without any large expenditures of manpower or equipment—the most sophisticated weapons at the cold frontier of physics are inexpensive indeed.

There are further good reasons for continuing the endeavor. Such fundamentally important properties of matter as superconductivity and superfluidity occur only at low temperatures. It is always possible that new, equally interesting phenomena would be found if we could make measurements at still lower temperatures. A good example is the discovery,[1] in 1972, of the superfluid phases of liquid He^3 below 3 mK. (By contrast, He^4 becomes superfluid at 2.2 K.) Nuclear cooperative phenomena,[2,3] that is, nuclear ferromagnetism and nuclear antiferromagnetism, constitute another example; temperatures less than 1 microkelvin are needed for investigations of this type.

Another, less spectacular but equally important reason for experiments near the absolute zero is our desire to study matter under conditions at which thermal disorder is small. It is then possible to investigate, for example, the angular distribution of gamma rays emitted from a radioactive source of ordered nuclei.

New frontiers

Generally speaking, physics near the absolute zero is the physics of order, and the more subtle the type of order is, the lower is the temperature needed to see it. Anthony Leggett recently discussed[4] the prospects for investigating some interesting phenomena at ultralow temperatures based on these considerations. One example involves looking at very weak couplings. When ordering involves delicate and complicated angular correlations, as it does in superfluid He^3-B, it will be destroyed very easily by incoherent scattering and will appear only when the temperature is so low that the scattering mechanism is sufficiently reduced.

The formation of Cooper pairs in many metals and alloys—the phenomenon of superconductivity—takes place below 23 K. So far only S-wave pairing has been observed; the sophisticated P-wave coupling has not been found yet. This may be due to the fact that in real metals the P-wave interaction is extremely weak and, consequently, that still lower temperatures are needed to see it. Equally well it is possible that P-wave pairing does not exist in metals at all. On the other hand, in superfluid He^3 P-wave coupling does occur.

It is generally believed that P-wave pairing is most likely to be found in strongly paramagnetic metals, in which exchange of long-lived spin fluctuations ("virtual paramagnons") favors triplet over singlet pairing. Of pure metals palladium is the most promising candidate, but recent calculations show that the transition temperature is of the order of 10 microkelvins. Two other systems, Ni–Rh and $ZrZn_2$, in which the degree of paramagnetism can be varied by changing the composition or the externally applied pressure, respectively, appear to be more promising candidates for experimental efforts.

From the theoretical point of view the main interest in P-wave superconductivity is that exchange of virtual paramagnons would probably create an anisotropic phase like superfluid He^3-A. But the currents found in such superconductors would couple directly to the electromagnetic field and would thus be much more easily detected than the electrically neutral currents in He^3. One could thus study the topological properties of the new superfluid phase in great detail.

Another example that Leggett discusses could be called "high-energy physics at ultralow temperature." Current theories of elementary particles predict very weak symmetry-violating effects (see, for example, "Cosmology and elementary-particle physics" by Michael S. Turner and David N. Schramm, PHYSICS TODAY, September, page 42), and one could look for such effects in highly ordered superfluids. For instance, one could study the electron–nucleon interaction due to neutral currents, which is invariant under time reversal but not under spatial inversion, by measuring the electric dipole moment of He^3-B; in this liquid the Cooper pairs have a nonzero expectation value of the vector product of the spin and orbital angular momenta, which is a condition required to make the dipole moment measurable.

Three new methods for cooling to 10 mK and below have rather recently become available. They are He^3–He^4 dilution refrigeration, Pomeranchuk cooling, and adiabatic nuclear demagnetization. A dilution refrigerator can maintain temperatures as low as 2 mK continuously and in the presence of a quite large heat leak. Pomeranchuk's method, cooling by adiabatic compression of a liquid–solid mixture of He^3, is capable of producing temperatures down to 1 mK. Nuclear demagnetization has enabled experimenters to reach a spin temperature of 50 nK and to cool conduction electrons in a metal to a temperature of 50 microkelvins. Figure 1 shows an example of this type of apparatus.

I shall now describe these techniques, emphasizing nuclear refrigeration, and experiments at the lowest presently accessible temperatures, including several examples of recent results. For more details on techniques, refer to my book.[5]

Dilution refrigeration

The operating principle of the dilution refrigerator is based on the peculiar

Cascade nuclear refrigerator constructed and operating in Lounasmaa's laboratory in Finland. The superconducting magnet assembly (blue) and the radiation shields are being installed. The nuclear spin system of copper has been cooled to 50 nanokelvin in this apparatus. For further details, see figure 5 and reference 3. Figure 1

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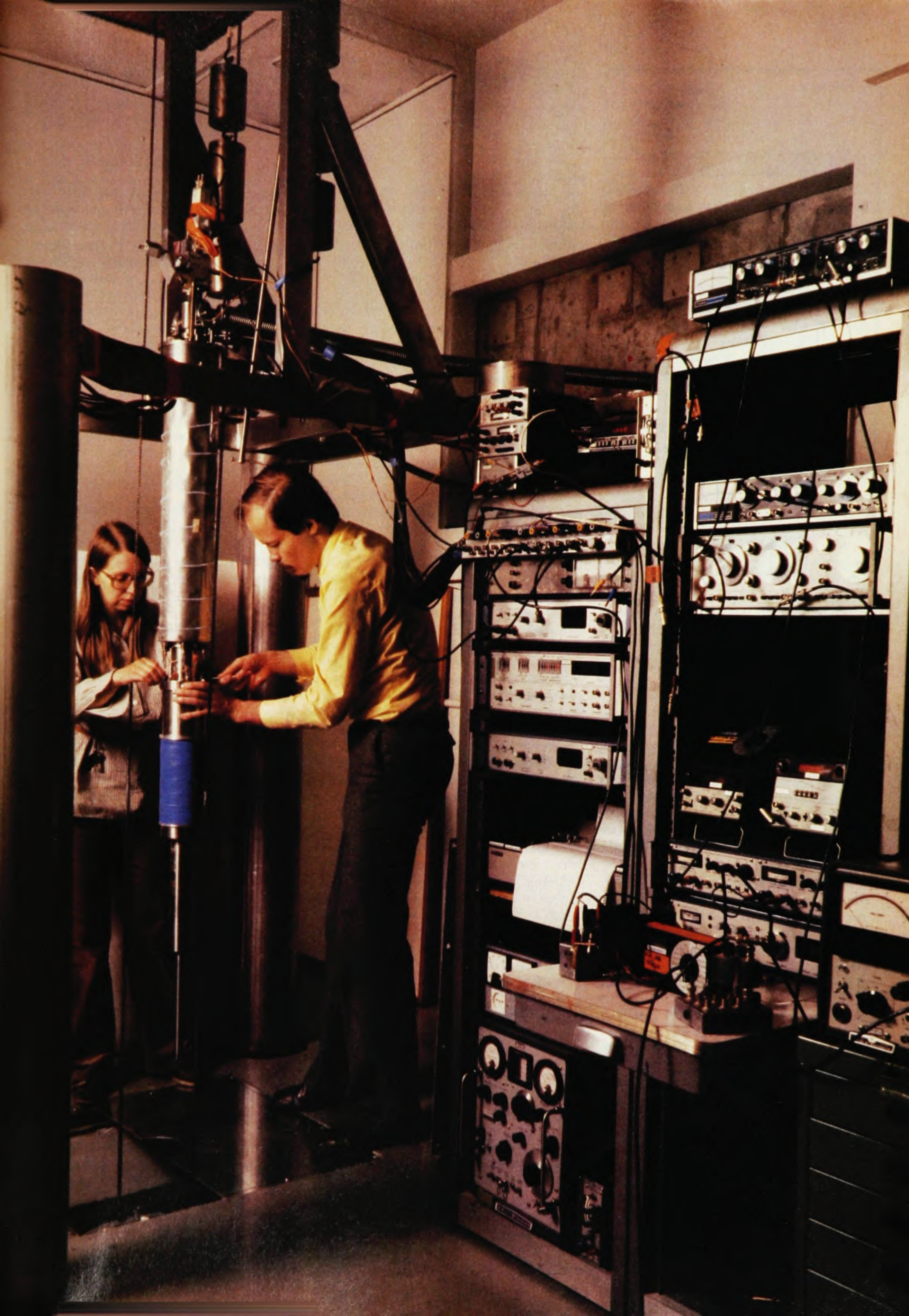
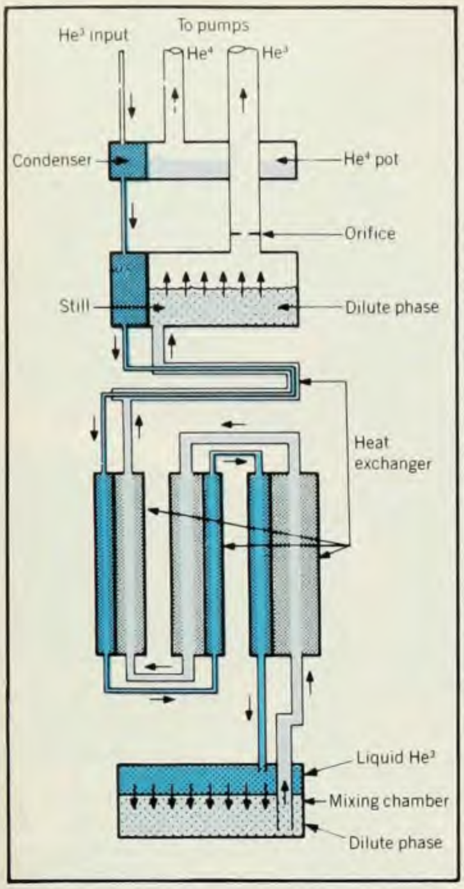




Conventional dilution refrigerator (schematic). To increase the thermal contact area, the main heat exchangers are filled with a sintered copper or silver sponge, with clear holes in the middle for an unimpeded flow of helium-3. Figure 2

properties exhibited by mixtures of He^3 and He^4 at low temperatures. Below about 0.8 K a liquid mixture of these two isotopes spontaneously separates into two components, one of the phases being rich in He^3 and the other rich in He^4. Because of its lower density, the He^3-rich phase floats on top of the He^4-rich phase.

As the temperature is lowered further, the relative amounts of He^3 and He^4 in the two phases change; below about 40 mK the upper, or concentrated phase is practically pure He^3 and the lower, or dilute phase is made of 6.4 atomic percent of He^3 and 93.6% of He^4. The possibility of dissolving a large amount of He^3 in liquid He^4 even at the absolute zero is of paramount importance for the success of the dilution refrigerator.

The other major reason for the success of the dilution refrigerator comes from the remarkably different low-temperature behavior of liquid He^4 and liquid He^3. Because of its zero nuclear spin, an atom of He^4 is a boson; below about 0.5 K, liquid He^4 is, effectively in its quantum-mechanical ground state. Very few phonons are excited and the liquid is a thermally inert superfluid. The light isotope, He^3, has a nuclear spin of $\frac{1}{2}$; it is thus a fermion and does not exhibit the sort of simple Bose condensation He^4 does. Consequently, one may describe the He^4 in the dilute phase as a "supporting medium" or "mechanical vacuum" for the "dilute gas" of active He^3 atoms.

We can now explain the operating principle of the dilution refrigerator by comparing it with ordinary evaporation. The concentrated phase of nearly pure He^3, where the active atoms are close together, corresponds to the liquid phase in the evaporation refrigerator. The dilute phase, in which the He^3 atoms are separated by superfluid He^4, corresponds to the vapor phase in the evaporation refrigerator. In both cases, energy is required to separate the atoms because of attractive forces among them; so moving molecules upward from the liquid to the vapor phase, or atoms of He^3 downward from the concentrated to the dilute phase, lowers the temperature of the liquid or concentrated phase. By constantly pumping molecules from the vapor in the evaporation refrigerator or He^3 atoms from the dilute phase in the dilution refrigerator, one can produce a substantial amount of cooling.

It is now easy to see the importance of the 6.4% solubility of He^3 in He^4. In an ordinary evaporation refrigerator the vapor phase becomes depleted of molecules rather soon, because the vapor pressure decreases exponentially with temperature, whereas in the dilution refrigerator the concentration of He^3 atoms in the dilute phase remains constant as the temperature is lowered below 40 mK. Thus, the number of He^3 atoms that cross the phase boundary per unit time is independent of temperature.

The principal parts of a dilution refrigerator are schematically illustrated in figure 2. Cooling by dilution is achieved in the mixing chamber. The process can be made continuous by circulating He^3 in the system by a pump at room temperature. Incoming gas is first precooled and liquefied in the condenser, which is attached to the He^4 pot maintained at about 1.1 K. The He^3 then enters the still heat-exchanger (where its energy is given to evaporate He^3 on the return path) at about 0.7 K and passes through a set of heat exchangers to cool it further before it enters the mixing chamber.

After crossing the phase boundary there, the He^3 atoms, driven by an osmotic pressure gradient, proceed in reverse order through the heat exchangers, along an unbroken column of the dilute superfluid phase, to the still where the liquid column ends.

Vapor is removed from the still by pumping, which is how He^3 is separated from He^4: Because the vapor pressure of He^4 is almost negligible even at 0.7 K, more than 90% of the outcoming gas is He^3 if a suitable orifice for reducing the flow of the He^4 film is installed.

The analysis of a dilution refrigerator can be put on a more quantitative basis. For instance, the cooling power under ideal conditions is given by the difference between the heat carried by He^3 from the last heat exchanger and the cooling produced by dilution:

$$\dot{Q} = \dot{n}_3(H_D - H_C)$$

where $\dot{n}_3$ is the circulation rate of He^3, H_D is the enthalpy of the dilute phase at the temperature of the mixing chamber, T_M, and H_C is the enthalpy of the concentrated phase at the temperature of the last heat exchanger, T_N. The enthalpies can be determined from the properties of liquid He^3; they are proportional to the squares of the temperatures. Putting in the values for the proportionality constants, we find

$$\dot{Q} = \dot{n}_3[(95\ \mathrm{J/K^2mol})T_M^2 - (13\ \mathrm{J/K^2mol})T_N^2]$$

For a perfect heat exchanger, $T_M = T_N$, so that

$$\dot{Q} = \dot{n}_3(82\ \mathrm{J/K^2mol})T_M^2$$

The cooling power is proportional to the square of the mixing chamber temperature. This is why the external heat leak eventually catches up and further cooling stops, even though the constant 6.4% solubility of He^3 into liquid He^4 ensures that $\dot{n}_3$ is not a function of T_M.

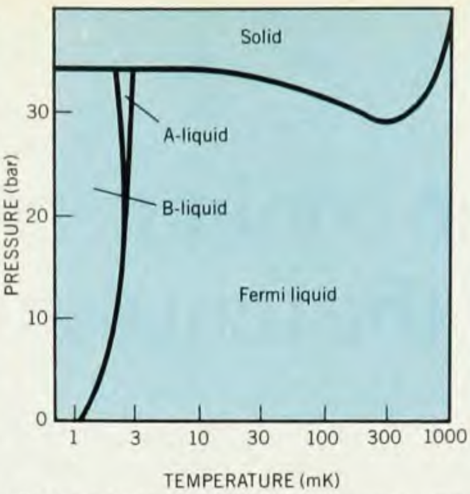
Helium dilution refrigerators have now been in general use for more than a decade. During this time their cooling power has increased by a factor of 100 and the low temperature limit has been pushed from 10 to 2 mK. The dilution refrigerator has already fully established itself as the most important tool for research at ultralow temperatures, replacing adiabatic demagnetization of paramagnetic salts. Many experiments that would have been difficult or impossible below 0.3 K are now performed routinely with dilution refrigerators.

Heinz London first suggested the principle of the dilution refrigerator in 1951; he, together with Geoffrey Clarke and Eric Mendoza elaborated on the suggestion in 1962. Pankej Das, Rudolf de Bruyn Ouboter and Krijn Taconis built the first cryostat of this type at Leiden in 1965; it was able to reach 0.22 K. Within a year Boris Neganov, Nicolai Borisov and Mihail Liburg at Dubna as well as Henry Hall, Peter Ford and Keith Thompson at Manchester built dilution machines capable of reaching much lower temperatures. John Wheatley, among others, has done a considerable amount of development work at the University of California at San Diego. I have recently written a short review on the subject.[6]

Giorgio Frossati and his coworkers at Grenoble have recently built a refrigerator[7] that reaches 2.0 mK in continuous operation; this is the lowest temperature so far produced by the dilution method. The Grenoble group has worked for many years and with good results to improve the performance of dilution cryostats. The success of any dilution refrigerator depends critically on the construction and effectiveness of the heat exchangers, on how close T_N approaches T_M. In the Grenoble refrigerator—which is similar to that shown schematically in figure 2—the heat exchanger contains very fine silver powder to increase the contact area between liquid He^3 and the exchanger body. The temperature of the heat-exchanger stages, as measured from the outcoming dilute mixture, decreases in the following sequence: 25, 6.5, 4.0, 2.7, 2.2, and 2.0 mK. The cooling power, $\dot{Q}$, is 30 nW with a gas circulation, $\dot{n}_3$, of 200 micromol/sec.

Dilution cryostats can easily be adapted to various experimental needs. A good example is the horizontal refrigerator built by Tapio Niinikoski.[8] The unusual construction was dictated by the geometry of magnet pole pieces and by the counting arrangements for experiments on spin-frozen, polarized proton targets. This machine has a very large cooling power—10 mW at 200 mK with $\dot{n}_3$ = 7 mmol/sec for example. More typical values for dilution refrigerators are 20 microwatt at 20 mK, 600 microwatt at 100 mK, and 2 milliwatt at 200 mK.

A very important use of dilution machines is the precooling of Pomeranchuk refrigerators and the various kinds of nuclear refrigerators, which need starting temperatures between 10 and 30 millikelvins.



Phase diagram of helium-3 showing the solid phase, the melting curve minimum at 0.32 K, the normal Fermi liquid phase and the super-fluids helium-3-A and helium-3-B. Note that the transition temperature at zero pressure is 1.1 mK; this is 2000 times less than the λ-point of helium-4. Figure 3

Pomeranchuk cooling

In 1950 Isaac Pomeranchuk proposed a refrigerator based on adiabatically compressing liquid He^3 to a solid. Yuri Anufriev built the first Pomeranchuk refrigerator in 1965 at Moscow.

Figure 3 shows the phase diagram of He^3. Notice that the slope of the melting curve is negative below 0.32 K. A liquid–solid mixture of He^3 therefore cools if it is compressed at temperatures below 0.32 K. During the process, liquid is continuously converted into solid. The main experimental difficulty is that compression cannot be achieved from the outside simply by pumping more He^3 into the low-temperature cell, because the helium in the connecting tube has to pass through states in the solid region of the diagram as it is cooled from higher temperatures; the connecting tube thus becomes blocked with solid He^3. One must therefore compress the cell some other way, for example by squeezing it with a He^4 pressurizer.

The shape of the melting curve of He^3, which forms the basis of the theory of Pomeranchuk cooling, is of course, determined by the Clausius-Clapeyron equation

$$\frac{dP}{dT} = \frac{S_l - S_s}{V_l - V_s}$$

Because $V_l - V_s$, the molar volume difference between liquid and solid remains positive, and because dP/dT is negative below 0.32 K, the entropy of solid He^3 must be larger than the entropy of the liquid at temperatures below the melting-curve minimum. This increase in entropy upon freezing is the basis for Pomeranchuk cooling.

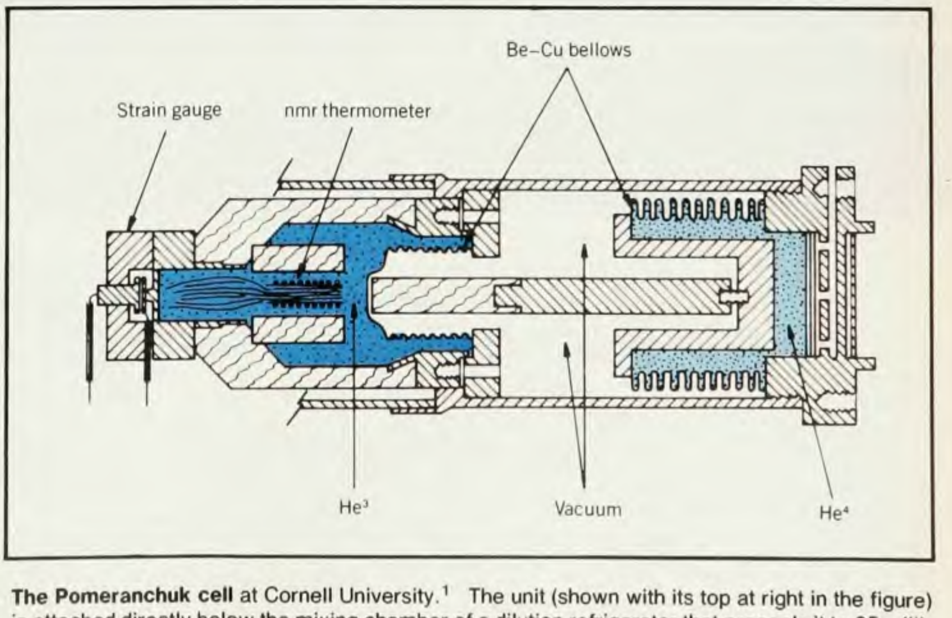
One can easily understand the reason for the entropy difference by looking at the structures of the liquid and the solid. In the liquid, the He^3 atoms are free to roam about and their wave functions overlap extensively; the thermodynamic properties are thus determined by the Fermi statistics. (The situation is analogous to the conduction electrons in a metal.) The entropy of the liquid, S_l, is therefore a linear function of the temperature, vanishing at absolute zero.

In the solid, He^3 atoms are tied to their lattice sites and their wave functions overlap only partially. Every atom is thus fairly independent of its neighbors and the nuclear spin entropy has a value close to $R\ln2$, which corresponds to complete disorder. Thus, at low T, the solid entropy can be larger than the liquid entropy.

Pomeranchuk's technique has been very important for studies of the superfluid properties of He^3. In fact, Douglas Osheroff, Robert Richardson and David Lee used just this method of cooling to discover the new, superfluid, phases. Figure 4 is a schematic diagram of an improved version of their cell. The device incorporates a hydraulic pressure-amplifier that consists of Be–Cu bellows connected by means of a rigid shaft. A moderate He^4 pressure change in the upper chamber (shown to the right in the figure), compresses the lower compartment sufficiently to solidify the entire He^3 charge. At a typical rate of solidification, 20 micromol/sec, cooling proceeds at 4 microkelvin/sec. The low-temperature limit is about 1 mK.

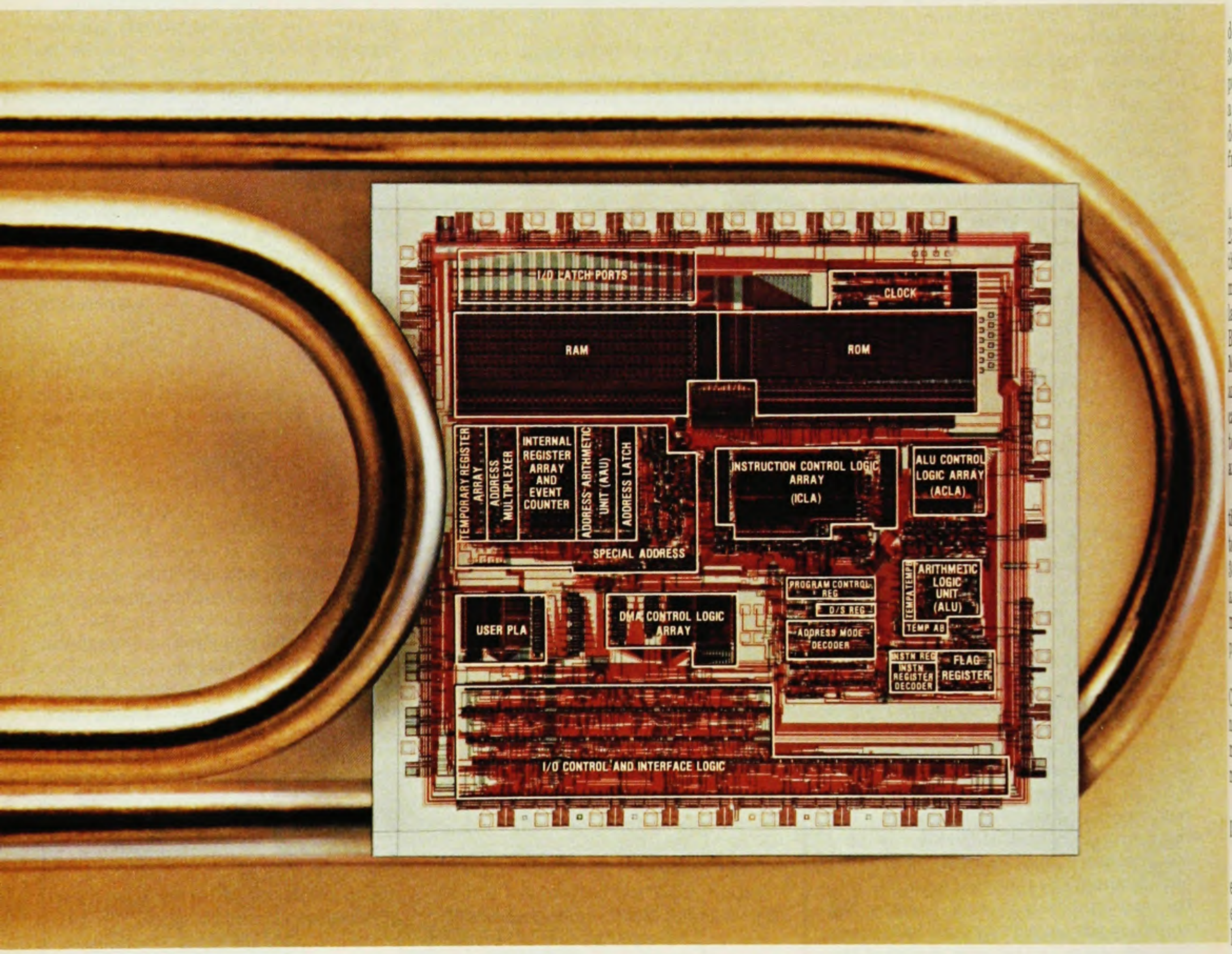
A very recent example of the use of Pomeranchuk cooling is provided by the work of Maurice Chapellier, Frossati, and Finn-Berg Rasmussen[9] on spin-polarized liquid He^3. Owing to the Fermi degeneracy the susceptibility of liquid He^3 is low and significant polarizations cannot be obtained even by applying a high magnetic field near 1 mK. In solid He^3 the susceptibility is much larger. Bernard Castaing and Philippe Nozières suggested that one could produce a polarized liquid by melting solid He^3 that had previously been polarized in a magnetic field at low temperatures.

Chapellier and his colleagues solidified about 35% of a He^3 sample during com-



The Pomeranchuk cell at Cornell University.[1] The unit (shown with its top at right in the figure) is attached directly below the mixing chamber of a dilution refrigerator that precools it to 25 millikelvins. The cell has a Be–Cu capacitative pressure gauge and an nmr thermometer. Figure 4

The one-chip computer: offspring of the transistor



The MAC-4 one-chip computer, developed for a variety of telecommunications applications, is compared to a standard-sized paper clip. The chip's numerous functional areas are labeled.

pression while it cooled from 40 to 2 mK in an external field of 2.6 tesla, thus obtaining a highly polarized solid. They then released the pressure sufficiently so that the solid melted and monitored the polarization via the nuclear magnetic-resonance signal. The results show that the polarization decreases exponentially from the high initial value to the low value characteristic of Fermi liquids; the group recorded initial liquid polarizations as high as about 20% and relaxation times as long as 5 min.

These and similar observations by Gerard Schumacher, Daniel Thoulouze, Castaing, Yves Chabre, Pierre Segransan, and Jacques Joffrin open up the possibility of studies of polarized liquid He^3. Further, it has been predicted that the melting curve is much lowered at high spin polarizations and that the minimum may completely disappear. The melting pressure may even be suppressed below the vapor pressure, in which case one or two triple points would appear in the phase diagram of figure 3.

Nuclear refrigeration

Nicholas Kurti at Oxford University was the first to use, in 1957, the technique of adiabatic nuclear demagnetization. The basic principle of this technique is the same as for paramagnetic salts, but there are very significant differences in practice. Because nuclear magnetic moments are about 2000 times smaller than their electronic counterparts, it is more difficult to produce significant changes in the nuclear-spin entropy by external means. In fact, one must start with temperatures below 20 mK and magnetic fields in excess of 5 T for nuclear cooling, whereas for paramagnetic salts the corresponding numbers are 1 K and 1 T. Fortunately, the starting conditions for nuclear magnetization can be reached rather easily with dilution refrigerators and superconducting magnets. (These are the conditions for the so-called "brute force" nuclear cooling method. I shall also discuss hyperfine-enhanced nuclear refrigeration and dynamic polarization, to which different starting conditions apply.)

The main advantage of nuclear cooling is the very low temperature that one can reach. Nuclei order spontaneously, owing to their mutual dipole-dipole interactions, well below 1 microkelvin. Because spontaneous ordering is the limit of any cooling process, temperatures in the sub-microkelvin region can be reached by nuclear demagnetization. For cerium magnesium nitrate, which is the weakest paramagnetic salt, the ordering temperature is slightly below 2 mK.

The technique of nuclear cooling consists of magnetizing the sample isothermally, raising the magnetic field from 0 to B_i at the (low) initial temperature T_i. One then isolates the sample and reduces the external field to B_f. The adiabatic demagnetization then cools the sample to a final temperature T_f given by

$$T_f = \frac{T_i}{B_i}\sqrt{B_f^2 + b^2}$$

where b is the effective field of the interaction between the nuclei. Thus, for example, if we start with $B_i = 8$ T, $T_i = 16$ mK, and we finish with $B_f = 0$, and the material has $b = 0.3$ mT then the final temperature of the sample is 0.6 microkelvin.

At very low temperatures it makes sense to distinguish between the temperature of the nuclear spins, T_n, and the temperature of the conduction electrons, T_e. The nuclei reach thermal equilibrium among themselves in the spin-spin relaxation time τ_2, and the nuclear spins and conduction electrons equilibrate with respect to each other in the spin-lattice relaxation time, τ_1. At very low temperatures τ_2 is much shorter than τ_1, so that the spins reach thermal equilibrium among themselves very rapidly while only slowly equilibrating with the remainder of the system. Nuclear demagnetization cools T_n, leaving T_e more or less unaffected. One must thus wait for times larger than τ_1 for the cold nuclei to cool the remainder of the system. For most metals τ_1 is the order of seconds at 10 mK; for insulators τ_1 is days or even weeks. It is thus clear that one must use metals for brute-force nuclear refrigeration.

The small τ_1 in metals is due to conduction electrons that act as intermediaries between the nuclear spins and the lattice. Only electrons near the Fermi surface contribute; their number is proportional to T_e, making τ_1 proportional to $1/T_e$, that is,

$$\tau_1 T_e = \kappa$$

where κ is called the Korringa constant. In practice only copper and indium have been used for brute-force nuclear magnetic cooling.

Unavoidably, external heat leaks into the conduction-electron system. This heat flow has an important effect on equilibrium between T_e and T_n. If the rate, $\dot{Q}$, at which the heat flows in is too large, the spin-lattice relaxation process is not sufficiently rapid for cooling the conduction electrons adequately, and the difference between the electron and nuclear temperatures will be large. Quantitatively,

$$\frac{T_e}{T_n} - 1 = \frac{\mu_0 \kappa \dot{Q}}{\Lambda(B^2 + b^2)}$$

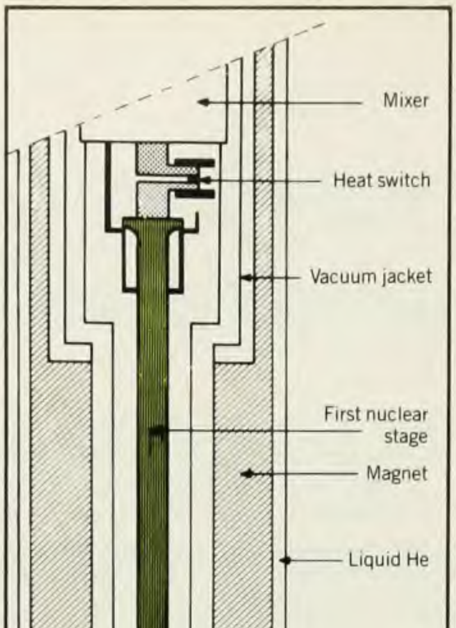
where μ_0 is the permeability of free space and Λ is the nuclear Curie constant.

Numerical calculations show that in many cases—to keep the sample cold for sufficiently long times or to obtain significant refrigeration of conduction electrons or an external specimen—one should not carry the nuclear demagnetization all the way to $B_f = 0$ but should stop at some intermediate field. In particular, one reaches the lowest T_e by demagnetizing to

$$B_f(\mathrm{opt}) = \sqrt{\mu_0 \kappa \dot{Q}/\Lambda}$$

In this case $T_e/T_n = 2$.

The most important use of nuclear refrigerators so far has been the cooling of liquid He^3. At least a dozen such cryostats are now in operation; they have already served for a large number of exciting measurements. One difficulty encountered in these experiments is the Kapitza thermal boundary resistance between liquid He^3 and a solid body. Luckily this difficulty can be partially overcome because the nuclear spins of He^3



The cascade nuclear refrigerator at Helsinki.[3] The first stage is made of 600 grams of insulated 0.5-mm copper wires; the second stage is a 2-gram bundle of 2000 0.04-mm copper wires. A dilution refrigerator precools the wires to 10 mK. The magnets apply fields of 8 and 7T to the first and second stages before demagnetization. (This is the working end of the apparatus shown in figure 1.) Figure 5 are coupled to the magnetic impurities on the copper surfaces, which markedly reduces the thermal resistance. In 1978 Osheroff and Mikko Paalanen at Bell Laboratories were able to use this technique to cool pure liquid He^3 to 0.28 mK, the lowest temperature yet reached for liquid He^3. Several other laboratories both in the US and in Europe, have also been able to cool superfluid He^3 to below 0.5 mK.

With mixtures of He^3 and He^4 the situation is much more difficult because a layer of non-magnetic He^4 on all surfaces prevents magnetic boundary-coupling. One possible approach, which has been tried but so far unsuccessfully, is first to cool pure liquid He^3 by nuclear refrigeration and then dilute it with He^4.

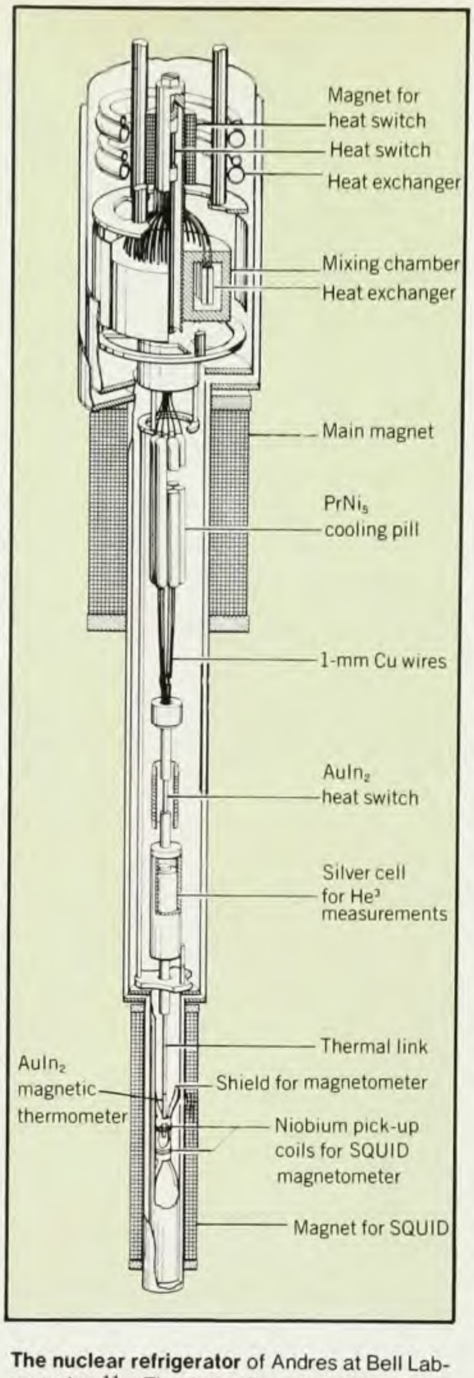
Liquid mixtures of He^3 and He^4 at low temperatures, are extremely interesting. He^3 becomes a superfluid at very low temperatures by a pairing mechanism analogous to that responsible for superconductivity in the Bardeen–Cooper–Schrieffer theory. The comparable theory for helium predicts that for dilute solutions of He^3 in He^4 the pairing will produce S-states, like the Cooper pairs in superconductors; at high concentrations, however, the stable pairs should be in P-states, as they are in pure He^3.

The long-range coherence properties of the superfluid phases can produce large-scale effects from atomic phenomena. Because the atomic phenomena themselves are too small to measure directly, the superfluids are very attractive experimental tools for examining them, as Leggett discusses in his review article.[4]

One simple example is the very small orbital magnetic moment μ_{orb} associated with the rotation of a homonuclear diatomic molecule. The magnetic moment is obviously directed along the orbital angular momentum vector. However, in an ordinary diatomic gas the molecular axes of rotation are oriented completely at random, even in a high field, because μ_{orb} is so small that it is undetectable.

The situation is radically different in the anisotropic superfluid phase He^3-A. In this case we have Cooper pairs, which may be considered as giant diatomic molecules. But, in contrast to an ordinary gas, the pairs automatically condense into the same state ("Bose condensation"), so that the axes of rotation are all in the same direction. The small orbital magnetic moments then add up coherently, we have a macroscopic effect, and the system behaves as a very weak liquid ferromagnet. Douglas Paulson and Wheatley[10] have actually detected μ_{orb} in He^3-A; they measured the anisotropy of zero sound attenuation in a nuclear refrigeration cryostat. This is the first observation of a genuinely chemical effect in helium, the formation of "molecules."

For most substances, as I have mentioned, spontaneous nuclear ordering occurs well below 1 microkelvin. (Solid



The nuclear refrigerator of Andres at Bell Laboratories.[11] The hyperfine structure of the praseodymium–nickel crystal greatly enhances the effective field at the nuclei, thereby also enhancing the cooling due to adiabatic nuclear demagnetization. This apparatus has cooled helium-3 to 1 millikelvin. Figure 6

He^3 is an exception owing to the strong quantum-mechanical exchange force.) Thus, to investigate nuclear cooperative phenomena, such as nuclear ferromagnetism and antiferromagnetism in a metal, one must bring the temperature of the system to the nanokelvin region. Luckily it is not necessary to cool the whole specimen, only the nuclear spins. The conduction electrons may remain at a considerably higher temperature because the spin-lattice relaxation time τ_1, even in a metal, is sufficiently long at 1 mK or below to keep the nuclear spins aligned long enough to perform the experiments.

A group[3] working at the Helsinki University of Technology has recently constructed a cryostat, with two nuclear demagnetization stages operating in series, in which the spins of copper nuclei have been cooled to $T_n = 50$ nK. This is the lowest temperature ever produced. The apparatus is illustrated in figures 1 and 5.

The second nuclear stage, which is also the specimen, is cooled by the first nuclear refrigerator to 0.2 mK in a field of 7 T; under these conditions the equilibrium nuclear-spin polarization is over 99%. There is no heat switch between the two nuclear stages, so one has to demagnetize the specimen and carry out the actual experiments in a time short in comparison with τ_1 to reduce losses of polarization due to relaxation. In zero external field τ_1 is about 20 min. We obtained the experimental information about the nuclear spin system of copper, in fields from 0 to 15 mT, from nmr measurements done with Josephson devices (SQUIDs).

We found the temperature T_n in the second stage after demagnetization by employing directly the second law of thermodynamics as applied to a system in thermal equilbrium: $T = dQ/dS$. We brought a heat input dQ to the nuclear-spin system by applying a small rf pulse at the nmr peak absorption frequency and obtained dS by measuring the polarization of the sample.

After constructing the entropy diagram of copper we found that the measured temperatures were less than those calculated for an interacting paramagnet with a constant local field. For instance, at an entropy of $0.45\,R \ln 4$ the measured temperature is 50 nK instead of the 110 nK that one calculates from a local field 0.34 mT. By analogy with electronic magnetism, a plot of the inverse static susceptibility as a function of temperature suggests that the nuclear spins prefer antiferromagnetic order, with a Weiss temperature of 150 nK.

On the other hand, we found no clear change in the entropy diagram or in the nmr data indicating a transition to the ordered state. This might be due to experimental inaccuracies. It is also possible that the nuclear-spin system was not able to make the transition (owing to the rather rapid demagnetization) or that the actual Néel temperature is below 50 nK. In electronic antiferromagnets the ratio of Weiss to Néel temperatures is often considerably larger than 1. In any case, these experiments show that the nuclear spin system of copper clearly favors antiferromagnetism.

Hyperfine enhancement

Another technique of nuclear cooling, "hyperfine-enhanced nuclear refrigeration" has recently become very successful. This method has been developed since 1967 mainly by Klaus Andres and his coworkers at the Bell Laboratories.[11]

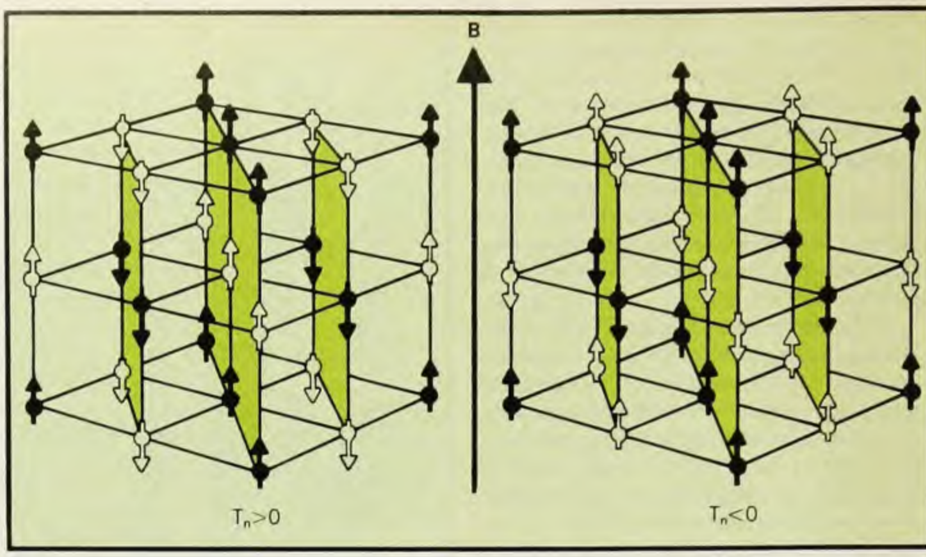
In certain paramagnetic lanthanide alloys in which the rare-earth ion occupies a singlet ground state split by the crystal field ("Van Vleck alloys"), an applied magnetic field mixes excited states with the ground state. The external field thus induces a large electronic polarization, which, in turn, enhances the field at the sites of the rare-earth nuclei.

In the best specimens, which are intermetallic compounds of praseodymium, the field is boosted by a factor varying from 8 to 22.

The enhanced field allows one to remove a comparably enhanced fraction of the spin entropy. With an initial magnetic field of 2 T and an initial temperature of 20 mK, the entropy of $PrNi_5$ is reduced by 25%. The starting conditions for demagnetization are thus excellent. For copper the corresponding entropy reduction is only 0.14%. Compounds such as $PrNi_5$ thus provide a very large cooling capacity during and after demagnetization. Unfortunately, these compounds are not easy to prepare because they must be extremely pure in both composition and structure. Even traces of magnetically ordered impurity phases may lead to irreversibilities that wipe out the entire nuclear-cooling effect. Other drawbacks of these specimens are their often poor thermal conductivity and the fact that spontaneous nuclear ordering, which sets the low temperature limit obtainable by the hyperfine-enhanced nuclear cooling method, occurs somewhere above 0.5 mK, depending on the compound used.

Figure 6 shows the apparatus that Andres is currently using. He has, for example, cooled superfluid He^3 to 1.0 mK. A group working at the University of Minnesota has recently observed[12] nuclear magnetic order in $PrCu_6$ at 2.5 mK.

Jan Huiskamp's group in Leiden has for some time operated a cascade nuclear-cooling cryostat based on the same principle. The first stage is made of $PrCu_6$ and the second of indium. Frank Pobell and his coworkers at the Kernforschungsanlage Jülich in West Germany have recently completed a refrigerator in which the first nuclear stage is made of 1.8 kg of $PrNi_5$ and the second of 0.6 kg of copper. Without a heat load, $PrNi_5$, demagnetized from 6 T, reached 0.48 mK; this is the lowest temperature so far produced by the hyperfine-enhanced nuclear-cooling method. Their second stage produced a record conduction-electron temperature of 50 microkelvins. The group headed by Kazuo Ono at the Institute of Solid State Physics in Tokyo has recently obtained a nuclear-spin temperature of 10 microkelvins in a similar cryostat.



Nuclear antiferromagnetic structures in lithium hydride at positive and negative absolute temperatures when the external field is parallel to a [100] direction of the crystal.[2] Black and open arrows represent lithium-7 nuclei and protons, respectively. Figure 7

Dynamic polarization

A group at Saclay, headed by Anatole Abragam and Maurice Goldman, has for many years studied spontaneous nuclear ordering in insulators, notably in calcium fluoride. Their experimental method of cooling is dynamic polarization by means of the "solid effect," followed by adiabatic demagnetization.

The specimen is a small CaF_2 crystal into which Tm^{2+} ions were introduced as electronic magnetic impurities at a concentration of 10 parts per million. An He^3 refrigerator first cools the specimen to 0.7 K in a field of 2.7 T. Under these conditions the polarization of the fluorine nuclei is almost zero, whereas the electronic moments of Tm^{2+} are almost completely polarized. This is, of course, due to the fact that the electronic magnetic moments are about three orders of magnitude larger than the nuclear moments.

Next, the system is supplied with microwave radiation whose angular frequency $\omega_e - \omega_n$ is equal to the difference in the Larmor frequencies of the Tm^{2+} electronic and the fluorine nuclear moments (the nuclear spin of calcium is zero). The microwaves induce flip-flop transitions, in which an electronic moment originally pointing up (in the direction of the external field) flips down and a neighboring nuclear moment originally pointing down simultaneously turns up. Owing to its short relaxation time with the lattice, the electronic moment will quickly return to its original direction after the flip-flop, whereas the nucleus, because τ_1 is very long in insulators, will retain its new orientation. The Tm^{2+} impurity then performs a new flip-flop process with another fluorine nucleus, and so on. This is the "solid effect."

The end result, in theory, is that the nuclear and electronic polarizations will become equal. In practice there are some losses, but the Saclay group has achieved nuclear polarizations in excess of 90% after 4 hours of microwave irradiation. The microwave power is then cut off and the He^3 refrigerator cools the CaF_2 specimen to 0.3 K. It is important to note that a nuclear polarization of 90% at 2.7 T corresponds to a temperature $T_n = 1$ mK. The starting conditions for nuclear demagnetization, which is the next step, are thus excellent. The end temperature reached in the nuclear spins is about 300 nK. The lattice stays at 0.3 K, but this does not matter.

Another feature of the Saclay experiments is the possibility of producing negative absolute temperatures in the nuclear-spin system. Irradiating the specimen with microwaves at a frequency of $\omega_e + \omega_n$ will induce flip-flip, instead of flip-flop, transitions: the result is that the nuclei become polarized opposite to the external field. This means that after demagnetization the system will reach a negative absolute temperature T_n of -300 nanokelvins.

Calcium fluoride behaves quite differently at positive and negative temperatures. Susceptibility data show that at positive T_n the system remains paramagnetic down to 300 nK, while at negative T_n there is a magnetic transition—apparently to an antiferromagnetic state—with a Néel temperature of -600 nanokelvins.

The Saclay group has recently proven[2] nuclear antiferromagnetism in LiH. Figure 7 shows the ordered structures predicted by the mean-field Weiss theory. The experimental method is the same as that already explained for CaF_2. However, in these experiments, the external field was 6.5 T and a dilution refrigerator precooled the sample to 50 mK. After three days of microwave irradiation and before demagnetization, the Li^7 nuclei were 80% polarized and the proton polarization was 95%. Neutron-diffraction studies both at positive and negative temperatures showed an extra Bragg reflection characteristic of antiferromagnetic structures.

The recent successes of nuclear refrigeration promise further progress in reaching low temperatures. It thus appears that, while Pomeranchuk cooling may remain important in special cases, dilution refrigeration and nuclear demagnetization will be the main tools for future research. Complete dilution cryostats are, in fact, already commercially available.

The examples of experimental results that I have discussed in this review demonstrate, I hope, that there is a great deal of fundamentally important physics research to be done in the milli-, micro- and nanokelvin regions of temperature. Many of the new phenomena to be discovered will probably involve macroscopic quantum coherence. It appears, in fact, both from the experimental and theoretical points of view, that low-temperature physics has once again entered a new and exciting era on the road to absolute zero.

But, of course, we can never reach that goal. This should not discourage us. The third law of thermodynamics is no hindrance to progress. If some new phenomenon occurs at a temperature that has previously not been reached, it is always possible, both in principle and in practice, to employ this phenomenon to step down further and reach this oasis of interest. Only if nothing happens—if there is only a desert between our lowest temperature and the absolute zero—we cannot progress further, but there is no scientific interest for it either.

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