Pictured above (left to right) are: J. R. Dillinger, chairman of the low-temperature conference steering committee; J. G. Daunt, member of the steering committee and chairman of the awards committee; F. G. Brickwedde, member of the steering committee and a representative of the United States on Commission I for very low temperatures of the International Union of Pure and Applied Physics; C. J. Gorter, secretary of Commission I of IUPAP and director of the Kamerlingh Onnes Laboratory, Leiden; and N. Kurti.

The first Fritz London Address:

Nuclear Orientation and Nuclear Cooling

By Nicholas Kurti

IN this talk I propose to give a general survey of experiments with oriented nuclear systems produced by cryogenic methods. In order to give a reasonably comprehensive picture I shall not restrict myself to work with which I personally was associated but shall also refer to work by others both in the Clarendon Laboratory and elsewhere.

Let me first of all state that the expression "oriented nuclei" is generally used to denote a system whose nuclear spins instead of pointing at random are preferentially oriented along one or several particular directions. In other words, in an oriented nuclear system the orientational disorder of the spins is not complete; their entropy, S, is somewhat or appreciably smaller than that corresponding to complete degeneracy of the magnetic substates. We can define nuclear orientation by

$$\frac{R \ln (2I+1) - S}{R \ln (2I+1)} \text{ not} \ll 1,$$

where I is the nuclear spin and R the gas constant. I should mention, by the way, that even if this condition is fulfilled it does not follow necessarily that the oriented nuclear system will show anisotropic properties.

Strictly speaking this definition only applies to equilibrium or static methods of nuclear orientation, that is, to cases in which the whole system comprising nuclear spins, electron spins, lattice vibrations, etc., remains in the same state as long as it does not exchange energy with its surroundings. But I chose this definition, somewhat lacking in generality, on purpose, because I do not intend to discuss the large number of nonequilibrium or dynamic orientation methods that have been put forward ever since Overhauser's [1] proposal. This omission is at least partially justified by the fact that Professor C. D. Jeffries will, in a later session, describe his method of polarization and the experiments,[2] in which for the first time sizeable nuclear polarization produced by a dynamic method was demonstrated by radioactive means.

For our condition of nuclear orientation to be fulfilled, that is, for the nuclear spin degeneracy to be at least partially lifted, it is necessary that the energy differences between various spin orientations should be of

Table 1

The Equilibrium (or Static) Methods for Nuclear Orientation

	Method	*Parameter Defining the Axis of Orientation*		*Nuclear Spins*
I	External field polarization	H ↑ 50 koersted .01°K	(direct coupling)	↑↑↑↑↑↑↑↑
II	Magnetic hfs polarization	H ↑ 1 koersted .05°K	(through electron spin)	↑↑↑↑↑↑↑↑
III	Electric hfs alignment	crystalline electric field ↑ .05°K	(through electric quadrupole moment)	↑↓↑↑↓↓↑↓
IV	Magnetic hfs alignment	crystalline electric field ↑ .05°K	(through electron spin)	↑↓↑↑↓↓↑↓

the order of, or at least not small compared with, kT. It turns out that for this condition to be satisfied the temperatures must be at least of the order of 1°K, or, more often, smaller or even very much smaller than 1°K. Hence the essential connection between nuclear orientation and cryogenics.

Table 1 summarizes the four basic equilibrium methods for nuclear orientation.

Method I, the simplest in principle, but experimentally difficult to realize, relies on the action of an external magnetic field on the nuclear magnetic moment (μ). Because of the smallness of μ, large values of H/T are required for the condition $\mu H/kT \approx$ or $\not< 1$ to be satisfied; thus something of the order of 50 koersted would be required at .01°K in order to produce a sizeable nuclear polarization. (For the illustration I assumed, for simplicity's sake, complete orientation; i.e., in this case all nuclear spins point in the same direction and in the same sense.)

Method II, proposed independently by Gorter [3] and by Rose,[4] relies on the strong local magnetic field due to unbalanced electron spins and orbits. These fields, which are responsible for the hyperfine structure (hfs) of spectral lines, are known to reach values of 500 koersteds, and even more; hence at a temperature of a few hundredths of a degree absolute the nuclear spins will orient themselves with respect to the electron spins, and, if the latter are polarized, by application of a quite modest external magnetic field, nuclear polarization will follow. This method only applies to nuclei subject to the action of unpaired electrons, that is mainly, though not exclusively, to nuclei in paramagnetic ions.

While in these first two methods the axis of orientation is provided by an external magnetic field, in the other two methods the nuclear spins align themselves with respect to a crystal axis, thanks to the action of the crystalline electric field. In method III, proposed by Pound,[5] we have the straightforward action of the electric field gradient on the nuclear electric quadrupole moment. In method IV, proposed by Bleaney,[6] the interaction is with the nuclear magnetic moment through the intermediary of the electron spin and electron orbit. In both III and IV the resulting orientational order is one of direction only and not of sense; hence the nomenclature "alignment", as distinct from "polarization". Both III and IV require single-crystal specimens. Method IV, like method II, is confined mainly to paramagnetic ions, while method III is of practical use only for a small group of compounds exhibiting large electric hfs coupling.

ALL these methods have been realized experimentally. The first method to be used was the magnetic hfs alignment method. It was with this that the first conclusive nuclear orientation experiments were carried out in September 1951 in Oxford by Daniels, Grace, and Robinson.[7] They found appreciable anisotropies in the γ-ray emission from radioactive Co^{60} nuclei contained in a single crystal of copper rubidium sulphate cooled by adiabatic demagnetization to a few hundredths of a degree. Similar experiments were carried out about the same time in Leiden by Gorter, Poppema, Steenland, and Beun.[8]

The magnetic hfs polarization method was first performed about a year later in Oxford by a rather long list of coauthors, namely: Ambler, Grace, Halban, Durand, Johnson, Lemmer, and myself.[9] Here again the γ-ray anisotropy from Co^{60} incorporated in magnetically cooled cerium magnesium nitrate was the indicator.

The external field polarization method was first demonstrated in 1955 by Dabbs, Roberts, and Bernstein [10] in Oak Ridge. They magnetized In nuclei in the metal by a field of about 12 koersted at .05°K and detected the nuclear polarization by the difference in the absorption of a polarized neutron beam according to whether the neutrons were polarized parallel or antiparallel to the In^{115} spins.

And, finally, it was in Oak Ridge again, that the electric hfs alignment method was first demonstrated about 18 months ago by Dabbs, Roberts, and Parker.[11] In

these experiments the anisotropy of the α-emission from U^{233} contained in a single crystal of uranyl nitrate served as a detector.

Hitherto most nuclear orientation experiments have been carried out by the two magnitude hfs methods, and had as their chief aim the study of the anisotropy of the γ-ray emission in a variety of radioactive decay schemes. Without going into details I shall sketch briefly the type of information that this sort of experiment can provide.

The angular distribution of the intensity of γ-rays emitted can be written, in most cases of practical interest:

$$W(\theta) = 1 - a(I_0, I_1, I_2, i_\beta, i_\gamma, A/kT)\cos^2\theta - b(I_0, \cdots, A/kT)\cos^4\theta$$

Here θ is the angle between the direction of observation and axis of quantization; I_0, I_1, I_2 are the nuclear spins associated with the various levels figuring in the decay scheme; i_γ and i_β are the angular moments carried away during the γ-decay and the preceding β-decay, respectively; and, finally, A is a coupling constant which, through the Boltzmann factor $\exp(-A/kT)$, determines the degree of nuclear orientation. a and b are rather complicated functions of these various parameters. The γ-ray anisotropy defined as

$$\epsilon = \frac{W(0) - W(\pi/2)}{W(0)}$$

is obtained by measuring the intensity parallel and perpendicular to the axis of orientation. The determination of ϵ as a function of temperature together with additional measurements at other values of θ gives one more or less detailed information about the functions a and b.

Fig. 1 shows typical experimental results obtained with Mn^{54} oriented by means of the magnetic hfs polarization method. The line shows the temperature (right-hand side ordinate scale) as a function of time after demagnetization. The dots and circles represent the counting rates (left-hand ordinate) as measured by scintillation counters oriented respectively parallel and perpendicularly to the polarizing magnetic field. (A slight asymmetry in the counters is responsible for the difference in counting rates at high temperatures, where the γ-emission is isotropic.)

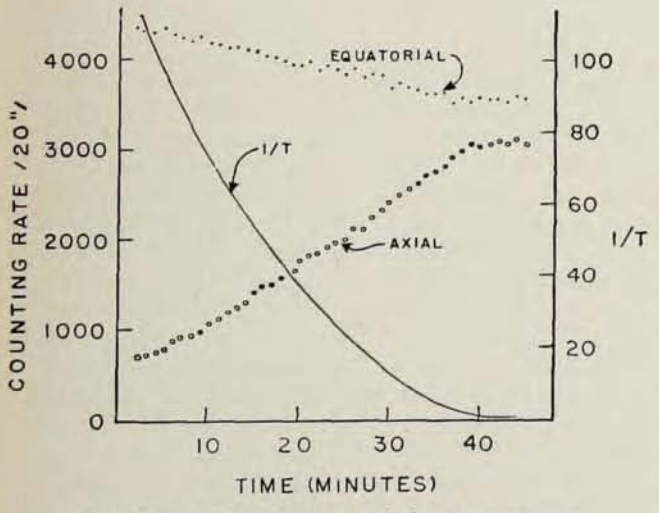

Fig. 1. Counting rates and inverse absolute temperature as a function of time. (γ-rays from oriented Mn^{54} nuclei.)

Fig. 2 shows the γ-ray anisotropy as a function of $1/T$, derived from these measurements. In this particular experiment a maximum γ anisotropy of 90% was obtained, corresponding to a 95% polarization of the Mn nuclei.

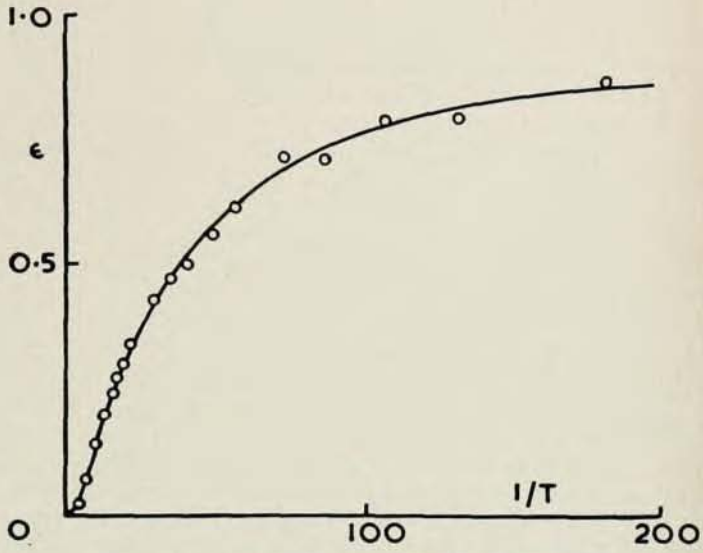

Fig. 2. Anisotropy of the γ-rays emitted by oriented Mn^{54} nuclei as function of inverse absolute temperature.

Such results on the γ-ray anisotropy are often supplemented by measurements of the sign and degree of the linear or of the circular polarization of the emitted γ rays. The former have mainly been used in the Oxford experiments and I want to mention Drs. Bishop, Goldschmidt, Knipper, and Perez y Jorba as mainly responsible for the development of the techniques.[12] The trickier problem of the measurement of the circular polarization in nuclear orientation work was solved in the Leiden experiments of Wheatley, Huiskamp, Diddens, Steenland, and Tolhoek.[13] Finally, I want to mention the experiments of Jastram, Sapp, Schroeder, and Daunt [14] at Ohio State University, who studied the angular correlation in the γ-ray cascade from oriented Co^{60} nuclei. Through combination of the various nuclear orientation results with data obtained by conventional nuclear physics methods and by paramagnetic resonance, it has been possible to establish in detail the radioactive decay schemes of some 20 nuclei and even to determine, though with no high accuracy, the nuclear magnetic moments of most of these radioactive nuclei. These and similar nuclear orientation experiments have

been carried out at the Universities of Illinois, Leiden, Ohio State, Oxford, at the National Bureau of Standards, and at the Oak Ridge National Laboratories.

One might say that these are useful but rather humdrum results. But no one will deny that not so long ago nuclear orientation has had its full share of glamour and limelight, that is if a headline in *The New York Times* and places of honor on the glossy pages of *Life* and *Time* may be taken as criteria. I refer of course to the celebrated Bureau of Standards experiment on parity conservation carried out by Drs. Ambler and Hudson in collaboration with their nuclear physicist colleagues Dr. Hayward and Mr. Hoppes of the Bureau and Professor Wu of Columbia.[15] But we shall hear about this work at a later session and I only want to make one point. It seems to me that the indispensable and essential part that low-temperature physics played in this experiment is, regretfully, overlooked. I need hardly emphasize this to the present audience, and I hope that when talking to noncryogenists about this experiment they will make it clear that there was more to it than simply reaching into a super ice box, obligingly kept by the Bureau, withdrawing from it a chunk of cerium magnesium nitrate suitably doped with Co^{60} and placing it into a magnetic field. No one who bothers to understand properly the Bureau of Standards experiment can fail to have genuine respect and admiration for the insight, ingenuity, and technical skill that Drs. Ambler and Hudson brought to bear on this problem. Also, it is as well to remember that it was *this* experiment that gave the first conclusive proof of the correctness of the Lee-Yang hypothesis and that, except for the work of Telegdi and Friedman,[16] *all* other noncryogenic parity experiments, the whole avalanche of them, started on their course *after* the first positive results from the Bureau of Standards had become known.

HAVING discussed briefly nuclear orientation and its bearing on nuclear physics, let me turn to the main preoccupation of this conference and see how nuclear orientation can help in solid-state physics and cryogenics. If one knows in detail the decay scheme and hence the γ-ray anisotropy to be expected, one can use oriented radioactive nuclei as indicators for certain properties. Let me give two examples:

1. In the magnetic hfs polarization (method II) experiments mentioned earlier we observed the anisotropy of the γ-radiation from Co^{60} incorporated in cerium magnesium nitrate which after being cooled by demagnetization was placed in a magnetic field of a few hundred oersted. Now it was known from paramagnetic resonance experiments carried out down to liquid-helium temperatures that even in the absence of a magnetic field there should be a sizable anisotropy owing to the effect of the crystalline field, according to method IV. The experimental result was not at all what had been expected. Not only were the γ-ray anisotropies much too small, but instead of increasing steadily with falling temperature they passed through a maximum and eventually decreased at the lowest temperatures. It was clear therefore that in this temperature range the magnetic behavior of the Co ions was different from that at about 1°K. The most likely explanation was that this change in behavior was due to interaction of the Co ions with the Ce ions, a plausible hypothesis on account of the cooperative ordering of the Ce ions known to occur below .01°K.

To test this hypothesis we decided to do away with Ce^{3+} as cooling agent and instead to incorporate our radioactive indicator, the Co^{60}, into the isomorphous diamagnetic lanthanum salt to which we added in rather small concentration some stable Co^{59} to act as cooling agent. It is true that we could not get to as low a temperature as with the cerium salt but, as Fig. 3 shows, this enabled us to prove the correctness of our explanation: considerably higher anisotropies were found, at a given temperature, with the lanthanum salt, and the experimental points fit the predicted curve reasonably well.

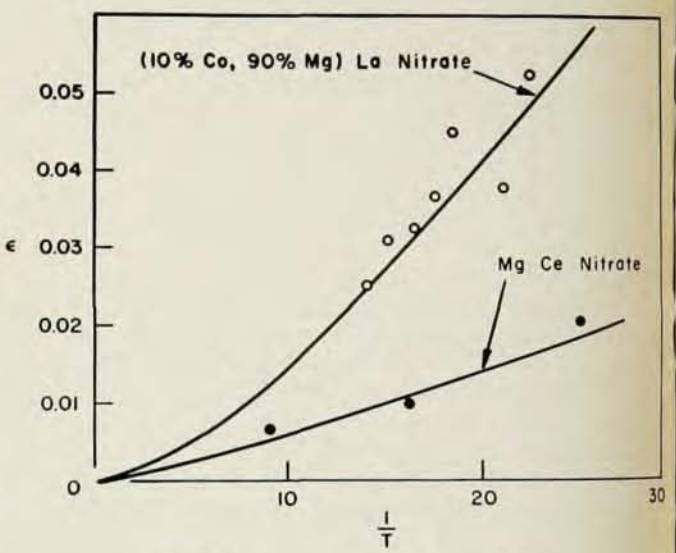

Fig. 3. Anisotropy of the γ-rays from Co^{60}.

2. Another example of nuclear orientation used as a tool in solid-state physics is the determination of the hfs coupling in a ferromagnetic metal. Within a ferromagnetic domain the electron spins are all parallel to each other; hence, if the temperature is low enough for the hfs coupling energy between nuclear and electronic spins to be of the order of kT, the nuclear spins too will be oriented parallel. If one takes a single crystal of metallic cobalt, in which, as is well known, the domains are all aligned along the hexagonal axis, and if one incorporates some Co^{60}, the γ-emission should become anisotropic at low enough temperatures and the degree of anisotropy will be a measure of the hfs coupling. Fig. 4 shows the results of experiments carried out in Oxford in collaboration with Grace, Johnson, Scurlock, and Taylor.[17] The γ-ray anisotropy is proportional to $1/T^2$, as is to be expected at these relatively high tem-

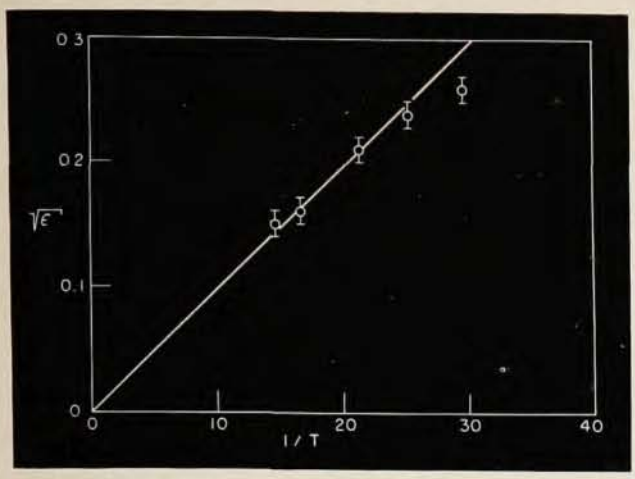

Fig. 4. Anisotropy of Co^{60} γ-radiation from a single crystal of cobalt metal.

peratures, and from the slope of the line one can calculate the hfs coupling constant. This turns out to be about the same as in the salts of Co^{++} which indicates that just as the paramagnetism of the salts, so the ferromagnetism of the metal is due chiefly to the 3*d* electrons and that the contribution from the 4*s* electrons is rather small, a conclusion which agrees with what Kittel and Mitchell [18] deduced from ferromagnetic resonance data. I think that this sort of experiment, especially when combined with specific heat measurements, will be useful in getting information about the electronic structure of ferromagnetic, and also of ferrimagnetic and antiferromagnetic, substances.

FINALLY, I want to turn to a purely cryogenic application of oriented nuclei and talk about nuclear cooling. The idea of producing very low temperatures by isentropic demagnetization of polarized nuclei is nearly as old as the ordinary magnetic cooling method based on electron paramagnetism. It was mentioned explicitly in 1933 by Gorter and by Simon and myself, but it is quite likely that many others have thought of it too; it is really so obvious. The temperature, T_f, one can attain by isentropic demagnetization of a spin system can be written in first approximation

$$T_f = H_{\text{eff}} \frac{T_i}{H_i}$$

where T_i and H_i are the initial temperature and field, and H_{eff} an effective magnetic field to represent the interaction of the spin with its surroundings. If H_{eff} is of magnetic origin (dipole field) then, for a given separation of the dipoles, it will be about a thousand times smaller for a nuclear paramagnetic than for an electron paramagnetic. Hence nuclear demagnetization should lead to temperatures orders of magnitude lower than ordinary magnetic cooling. The main difficulty is that, as mentioned earlier, very large values of H_i/T_i are needed because of the smallness of the nuclear magnetic moment. The possibilities were discussed in considerable detail by Simon in 1939 [19] and it is undoubtedly because of the difficulties that little progress has been made until recently. The work I want to describe was carried out in the Clarendon Laboratory, Oxford, by Dr. F. N. H. Robinson, the late Sir Francis Simon, Mr. D. A. Spohr, and myself; [20] the final experiments were performed in the early summer of 1956.

Fig. 5 shows the four steps of a magnetic cooling experiment. The electronic stage, which provides the heat sink at 10^{-2}°K, is a paramagnetic salt that can be cooled by adiabatic demagnetization. The nuclear stage is connected to it by means of a heat-conducting link. The first step (a) is the isothermal magnetization of the electronic stage at 1°K. This is followed (b) by adiabatic demagnetization which brings both the electronic and the nuclear stage to about 10^{-2}°K. In the third step (c) the nuclear stage is magnetized isothermally, the heat of nuclear magnetization being absorbed by the electronic stage. Finally, after thermal contact between the two stages has been broken (d), the nuclear stage is demagnetized.

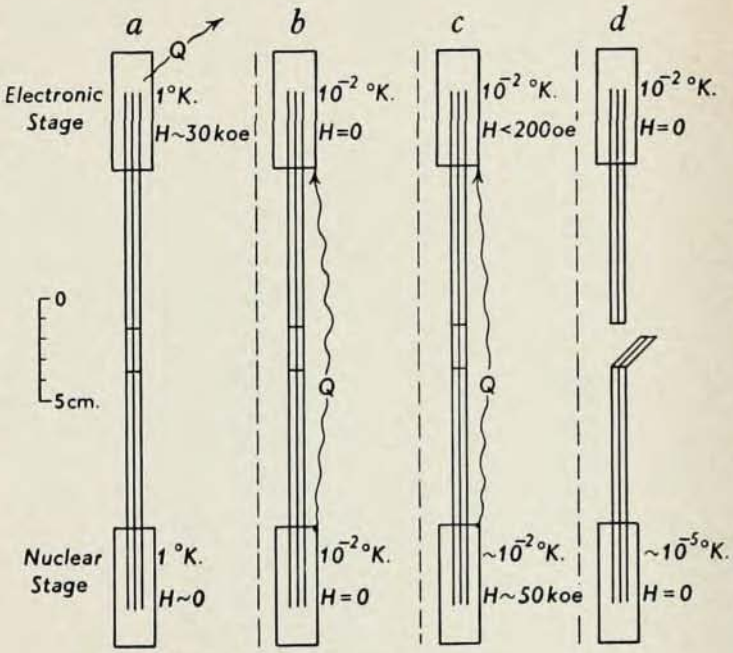

Fig. 5. The four steps in nuclear cooling.

In designing an apparatus for a nuclear cooling experiment the two main considerations were the provision of adequate energy transfer between the nuclear spin system and the electronic stage, and the elimination of heat influx to the nuclear stage and heat generation in it. As to the first point a satisfactory technique was developed to ensure good heat contact between the thermal link and the electronic stage; the more fundamental difficulty of adequate energy transfer from the nuclear spin system was taken care of by using a metal for the nuclear stage. For copper the nuclear spin relaxation

time is about 100 secs at 10^{-2}°K and varies inversely with temperature.

The reduction of the heat influx to the nuclear stage to about 1 erg/min was achieved by surrounding the specimen with a thermal shield kept at 0.1°K. Finally, heat generated in the metallic specimen by eddy currents was reduced by subdividing the specimen and by shielding.

The cryostat designed to satisfy these conditions is shown in Fig. 6. It shows thermal shields at 20°K, 4°K, 1°K, and 0.1°K; the latter consists of magnetically cooled manganous ammonium sulphate with copper wires and liquid helium ensuring fair temperature equilibrium throughout this shield. The electronic stage consisted of chromic potassium alum with 1500 enamelled copper wires of 0.1 mm diameter embedded into it. This bundle acted both as the thermal link and, its lower end, as the nuclear stage. No attempt was made to incorporate a thermal switch; the link used was something of a compromise: the heat transfer it provided was neither too good nor too bad. The temperature of the nuclear stage was to be determined by its nuclear magnetic susceptibility. A system of coils surrounding the nuclear stage and connected to the usual ballistic system was provided for this purpose.

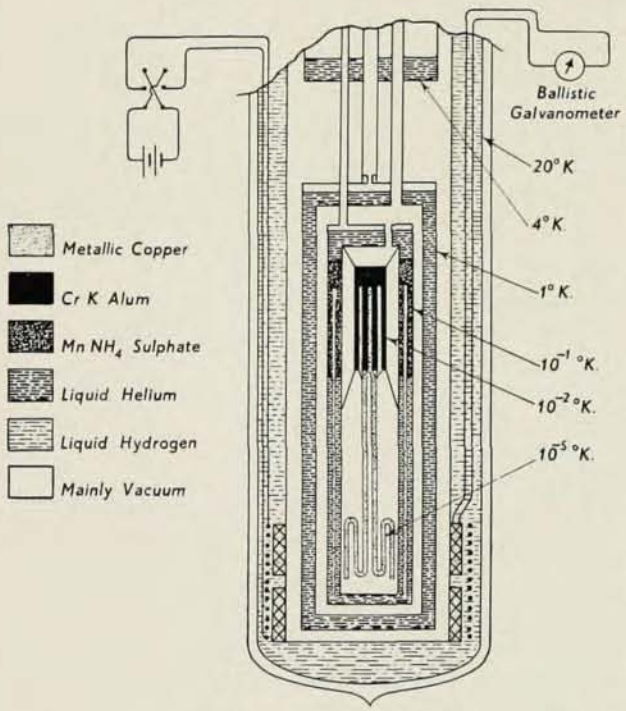

Fig. 6. Cryostat for nuclear cooling.

Fig. 7 shows the results of a series of nuclear demagnetizations, all starting from 0.012°K and from fields ranging between 3.5 and 28 koersteds. It gives the deflections of the ballistic galvanometer from which the absolute temperatures can be obtained by Curie's law

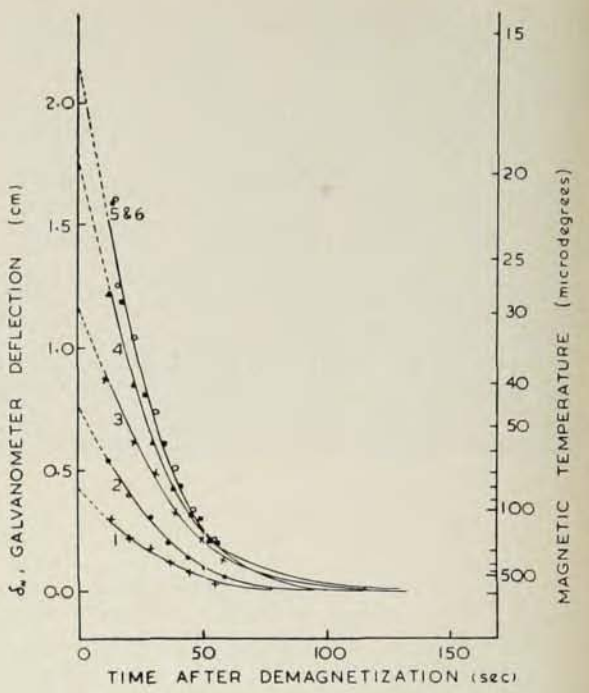

Fig. 7. Ballistic galvanometer deflection and absolute temperature (by Curie's law) after nuclear demagnetization. $T_i = 0.012$°K. H_i (in kilo oersteds) for the different curves = (1): 3.5; (2): 6.9; (3): 10.4; (4): 13.9; (5): 20.9; (6): 27.8.

(right-hand side ordinate scale) as a function of time after demagnetization. We see that, as expected, higher initial fields lead to progressively lower temperatures, the lowest actually measured temperature being about 20 microdegrees. The temperatures reached immediately after demagnetization can be obtained by extrapolating the warming-up curves to zero time. Substituting these values of T_f into the relation

$$T_f = H_{\text{eff}} \frac{T_i}{H_i}$$

we find that this relation is well obeyed with $H_{\text{eff}} = 27$ oersted.

This value of H_{eff} is considerably larger than $H_{\text{eff}} \approx 4$ oersted predicted for the various interaction mechanisms between the nuclear magnetic moments. On the other hand, as pointed out by Kittel,[21] one can expect considerably larger coupling energies due to the interaction between the nuclear electric quadrupole moment and the crystalline electric field which in our rather strained specimen is certainly not of pure cubic symmetry. It is therefore likely that this electric quadrupole coupling is responsible for the abnormally high value of H_{eff}.

The obvious question one asks at this point is whether these results justify the effort that has gone into this experiment. I should perhaps explain that we regard these experiments as something exploratory, simply a

proof of the feasibility of the method. To describe their position in the field of nuclear cooling an operatic metaphor might be permissible. I would say that these experiments certainly do not correspond to a grand finale; but they may, perhaps, be likened to the closing bars of the overture.

What then are the most obvious future trends? First of all, it should be possible to increase four- or even five-fold the value of H_i/T_i and thereby to reduce the nuclear entropy by about 40% from its ideal value, as compared with the present 2%. Demagnetization would then lead to below the nuclear Curie point and one would be able to see whether the nuclear ordering is of a ferromagnetic or antiferromagnetic nature and to study the possible influence of nuclear spin ordering on the electric and magnetic properties of a metal.

Another worthwhile problem to tackle is the cooling of other specimens into the microdegree temperature range. At first sight this seems rather problematic since, as mentioned before, nuclear spin relaxation times increase with falling temperature and, even in a metal, they will be about 10^5 sec at 10^{-5}°K. But, as was first pointed out by Casimir, one has to be very careful about using this relaxation time indiscriminately to characterize any spin equilibration process. The basic physical quantity that determines spin relaxation is the transition probability for a spin flip; its reciprocal is approximately equal to the thermal relaxation time of a spin system placed in an *infinitely large* heat reservoir. But in our case the heat reservoir, namely the conduction electrons, is *extremely small* and the equivalence does not hold. Kittel [21] has analyzed the situation in detail but a simple qualitative reasoning shows that in a nuclear cooling experiment the conduction electrons will follow the nuclear spins very promptly. It is true that it takes at an average 10^2 secs at 10^{-2}°K or 10^5 sec at 10^{-5}°K for a nuclear spin to flip, and to exchange its energy with the conduction electrons. But because of the smallness of the electronic specific heat very few nuclear spins need flip to cool down the conduction electrons, and the process instead of taking 10^5 secs only takes about 10 secs. It seems therefore that there is no fundamental obstacle to cooling other systems to these temperatures and there is at least a chance that eventually microdegrees will assume a similar place in cryogenics as do millidegrees today.

THIS brings me to the end of what I wanted to say about nuclear orientation and nuclear cooling. I have been wondering whether I should apologize for having chosen as my subject on this particular occasion something not in Fritz London's special fields of interest. I don't think I need do that. Fritz London's tastes for problems in physics were truly catholic; and pretty well any problem, in whatever branch of physics or chemistry, commanded his acute attention and absorbing interest. In fact, in so many fields did London's work exert a lasting influence that one might ask why low-temperature physics should have been chosen to pay homage to his memory.

I think that can be fully justified by Fritz London's unique position in cryophysics. We all know—a mere look at the program of this conference shows it clearly—that low-temperature physics has increased by leaps and bounds in the last years, and has ceased to be the closely knit, unified, almost self-contained discipline of one or two decades ago. It may well be that before long we shall have to define or redefine the boundaries of cryophysics. But, in whatever way this demarcation will be done, it is sure that the phenomena of liquid helium and of superconductivity will always form an essential, sine-qua-non part of cryophysics. Now it is fair to say that no other theoretical physicist has made a deeper impact on these two subjects than did Fritz London. It is sad that he did not live to see the recent exciting developments in the theories of superconductivity and of liquid helium which may well bring us to the solution of these fascinating problems, but though he is no longer with us, "his soul goes marching on". I shall end this talk with a quotation from Profesor Felix Bloch's moving tribute to Fritz London, which appeared in the second, posthumously published volume of *Superfluids:* "If, as London explained in the preface to this volume, the time comes for a third volume on the quantum theory of superfluids, and, even if the author of this future volume should be able to reveal in it the last mysteries of superfluidity, there can be no doubt that it will be permeated by London's spirit."

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