NUCLEAR MAGNETIC ORDERING AT NANOKELVIN TEMPERATURES

If you cool a suitable metal such as copper to sufficiently low temperatures, its nuclei will align spontaneously; susceptibility and neutron diffraction experiments have detected the effect and have set a new low-temperature record in the process.

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Just as electrons engage in spontaneous magnetic ordering, accounting for such phenomena as ferromagnetic domains, so too can nuclei order spontaneously. However, because nuclear magnetic moments are very much smaller than electron magnetic moments, spontaneous nuclear ordering occurs only at extremely low temperatures. As we will see, experiments conducted in the course of research on nuclear ordering have produced spin temperatures as low as 25 nanokelvins in copper and 2 nanokelvins in silver.

Electronic magnetism shows a wide spectrum of ordering phenomena, extending from a few millikelvins in cerium magnesium nitrate to above room temperature in iron. In nuclei, we expect spontaneous ordering when the thermal energy kT of the nuclear magnetic dipoles becomes smaller than their interaction energy. With nuclear moments three orders of magnitude smaller than their electronic counterparts, and with dipolar interactions proportional to the squares of the magnetic moments, nuclear spin systems should show order only at submicrokelvin temperatures. Solid He^3 is an exception, owing to the strong quantum mechanical exchange force that arises from the overlap of electron wavefunctions caused by the large zero-point motion. So are van Vleck paramagnets such as $PrNi_5$, which are paramagnetic materials in which an applied magnetic field is enhanced considerably at the nucleus by inner-shell electrons. In these systems, the transition temperature is relatively high, around 1 mK.

The experimental work discussed in this article represents 17 years of labor at Helsinki University of Technology and, more recently, four years of work at Risø National Laboratory near Copenhagen, Denmark. The Helsinki experiments involved susceptibility measurements on a single crystal of copper. They yielded the field-vs-entropy diagram, which revealed three antiferromagnetic phases. The Risø work used neutron scattering to observe the (100) Bragg reflection from a specimen with ordered nuclei and revealed two antiferromagnetic phases. It is likely, but not certain, that the ordered state has been reached by the Helsinki group in silver as well; in any case, a new low-temperature record, 2 nanokelvins, has been set.

Theory is the most important reason for experiments on spontaneous nuclear ordering in simple metals, because nuclei in solids are the magnetic systems most amenable to calculations: Nuclear spins are well localized, the magnetic degrees of freedom do not couple to lattice distortions and the spin–spin interactions are known. The orienting forces in simple metals are expected to be dominated by the direct dipole–dipole coupling and by the Ruderman–Kittel indirect exchange force.[1] The Zeeman interaction with the external magnetic field is also present, but quadrupolar forces are absent because of cubic symmetry. Thus the Hamiltonian can be written

$$H = H_D + H_{RK} + H_Z$$

The anisotropic dipolar part and the Zeeman term are known exactly. The isotropic Ruderman–Kittel interaction is mediated by the conduction electrons, which carry energy from one spin to another by scattering from the nuclei. In copper the dipolar and electron exchange forces are comparable, whereas the much weaker interactions in silver are exchange dominated. In both metals the ordering is predicted to be antiferromagnetic due to the particular distances between the nuclei in the crystal lattices.

Experimental work

The pioneering experiments on copper by Nicholas Kurti and his coworkers at Oxford established in 1956 the

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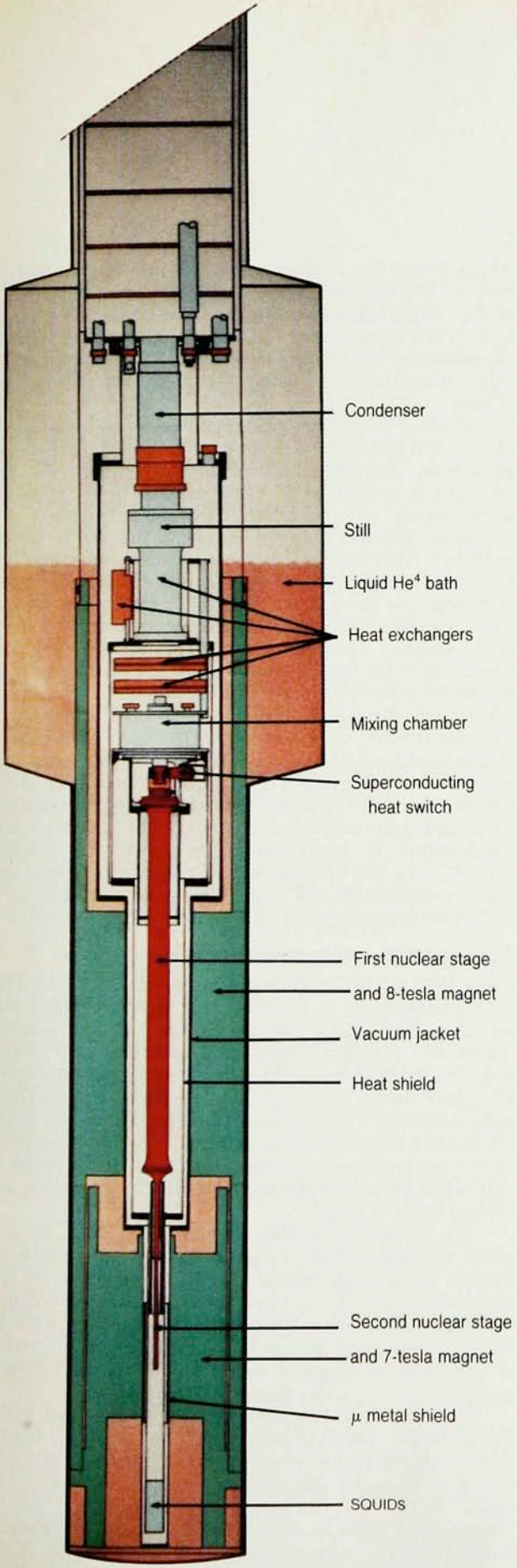


Refrigerator. The cascade nuclear demagnetization refrigerator shown here schematically is used in nuclear magnetic ordering experiments in Helsinki. Ten moles of copper in an 8-tesla magnetic field make up the large first nuclear stage. The sample itself is the small second nuclear stage. The copper or silver specimen is welded to the upper stage, without a heat switch. This means that the conduction electron temperature is the same in both stages and that one relies on the slowness of the spin–lattice relaxation process for sufficient thermal isolation of the nuclear spin system in the sample. (From reference 5.) **Figure 1**

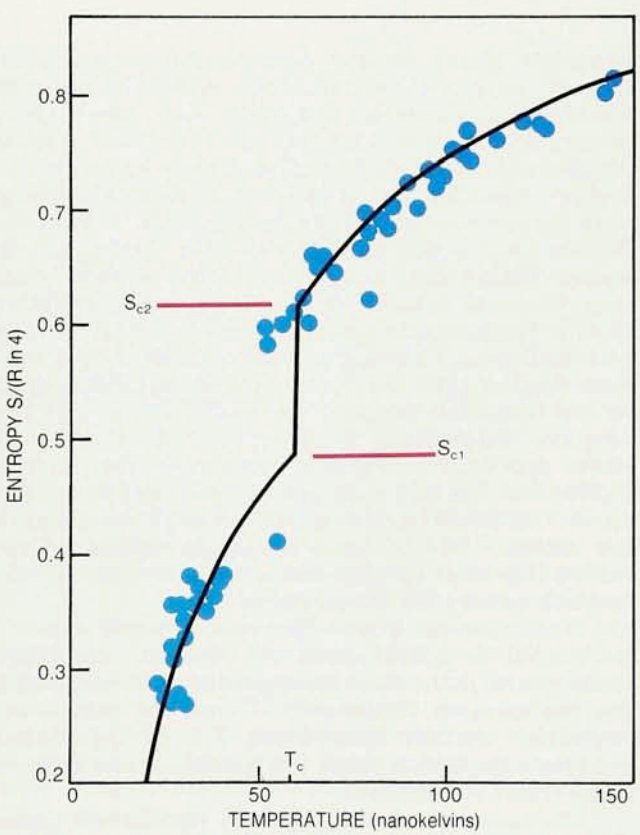
feasibility of the nuclear demagnetization method of cooling.[2] In spite of the limitations imposed by cryogenic techniques available at that time, the Oxford group succeeded in reaching 1 μK. Fifteen years later, Anatole Abragam, Maurice Goldman and their coworkers at Saclay made the first studies[3] of nuclear cooperative phenomena, on insulators like CaF_2 and LiH. In Helsinki, we began our work[4,5] on copper in 1973. Among several nuclear cooling schemes, the "brute force" method, which is applicable to metallic samples, is the most straightforward.[6] In this technique the nuclear spins are first polarized, using a high initial magnetic field H_i and a low precooling temperature T_i, and then adiabatically demagnetized to a small field H_f. The nuclei thereby cool to a very low temperature T_f, given by $T_i(H_f^2 + h^2)^{1/2}/H_i$, where spin–spin interactions dominate. The internal dipolar field h is 0.36 millitesla in copper and 0.04 mT in silver. One then expects that if $H_f < h$ and if the entropy is low enough, the Ruderman–Kittel and dipolar interactions will produce spontaneous order in the nuclear spin assembly at very low temperatures.

It is important to note that near absolute zero it is meaningful to speak about two distinct equilibrium temperatures in the same specimen and at the same time: the nuclear spin temperature T and the lattice and conduction electron temperature T_e. During nuclear ordering experiments these two quantities can differ by many orders of magnitude.

The nuclei reach local thermal equilibrium among themselves in a time characterized by the spin–spin relaxation time τ_2, which is on the order of a few milliseconds, whereas the approach to equilibrium between nuclear spins and conduction electrons is governed by the spin–lattice relaxation time τ_1. At low temperatures τ_2 is much less than τ_1, which makes a separate nuclear spin temperature meaningful and real. The spin–lattice relaxation time is inversely proportional to the conduction electron temperature. Typically, in

copper at a temperature T_e of 50 μK, the relaxation time τ_1 is 3 hours.

To reach the necessary ultralow temperatures and to maintain them for a sufficiently long time, one needs a special cascade refrigerator; figure 1 is a schematic diagram of the double nuclear demagnetization cryostat in Helsinki. A typical experiment is performed as follows. The large upper nuclear stage is first magnetized to 8 T and the second nuclear stage to 7 T; both are cooled by the dilution refrigerator to 15–20 mK. The superconducting heat switch between the mixing chamber of the dilution refrigerator and the first nuclear stage is then opened, and the 8 T field is reduced, in two steps, to 20 mT. With a copper specimen this produces a conduction electron and lattice temperature $T_e = 50$ μK in both nuclear stages. Next, the specimen—that is, the second nuclear stage—is demagnetized from 7 T to zero field in about 20 minutes,



Nuclear spin entropy of copper as a function of temperature with no external magnetic field. The vertical line at a temperature T_c of 58 nK indicates a phase change. The latent heat, given by $T_c(S_{c2} - S_{c1})$, is 0.09 μJ/mol. The entropy S is given in units of its maximum value, $R \ln(2I + 1)$, where the nuclear spin I of copper is 3/2. (From reference 5.) **Figure 2**

which brings the nuclear spins well into the nanokelvin region. The sample then immediately begins to warm up slowly, owing to heat leaking from the conduction electron system at a temperature T_e of 50 μK. The nuclear spins in the specimen are monitored by low-frequency ac-susceptibility measurements, using SQUIDS as the sensing elements.

Finding the absolute thermodynamic temperature of the nuclear spin system is not a trivial matter in these experiments. The second law of thermodynamics, $T = dQ/dS$, must be used directly. The procedure in the paramagnetic regime is as follows:[5] One starts from a 1 mT magnetic field and from a nuclear spin entropy S_1, which is known from susceptibility measurements because the sample is still in the region where the equations of the paramagnetic state apply. The specimen is then demagnetized adiabatically, that is, at constant entropy, to zero field, whereby the spin system reaches the very low temperature that one wishes to know but which cannot be deduced directly from susceptibility measurements. Next, a known heat pulse dQ is applied, causing the entropy to increase from S_1 to S_2. The spins are then adiabatically magnetized from 0 back to 1 mT. The entropy S_2 can now be found, in the high field region, again from susceptibility measurements, by relating the susceptibility χ to polarization, and polarization to entropy. The unknown low temperature in zero field is then given by $T = dQ/(S_2 - S_1)$. In the ordered region the measurements are more complicated; the susceptibility is employed as a thermometric parameter by writing $T = (dQ/d\chi)/(dS/d\chi)$.

Susceptibility measurements on copper

From 1973 to 1986 in Helsinki we carried out susceptibility measurements on copper; many physicists participated, including Gösta Ehnholm, Matti Huiku, Jacques Jacquinot, Markku Loponen, Lounasmaa and Jouko Soini.[4,5] Figure 2 shows the entropy-vs-temperature curve of copper below 150 nK, which we deduced from these measurements. The vertical line emphasizes the phase change at a temperature T_c of 58 nK. The first-order jump in entropy is quite clear from this picture. In the course of this work we cooled the copper nuclei to the record low temperature of 25 nK.

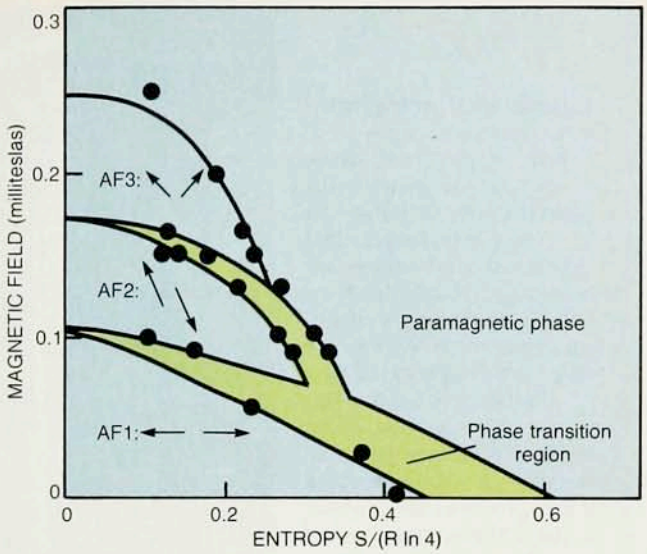
We obtained the data in figure 2 using a natural polycrystalline sample of copper. In later experiments we focused on a single-crystal specimen and measured the magnetic susceptibility in all three Cartesian directions.[5] This crystal was a slab 0.5 mm in the x-direction, 5 mm in the y-direction and 20 mm in the longitudinal z-direction of the external magnetic field. The z-axis was approximately in the crystallographic [001] direction.

We carried out experiments in several final fields. For example, when the field was 0, the susceptibilities χ_x and χ_z stayed almost constant for the first five minutes, while χ_y changed significantly, reaching a clear maximum. At 0.15 mT, the transverse components χ_x and χ_y behaved the same as at low fields, but the longitudinal susceptibility χ_z increased clearly at the beginning of the

warm-up and reached a maximum. At 0.20 mT, χ_z decayed at all times, while χ_y had a low maximum and χ_x was flat.

For drawing conclusions from these observations, we must recall that below the Néel point the antiferromagnetic susceptibility transverse to sublattice magnetization is constant while the parallel susceptibility goes to zero as the temperature approaches zero. Consequently, when the field is 0, the nuclear magnetization in copper is mainly along the y axis because the susceptibility changes were largest in this direction; at 0.15 mT, the sublattice magnetization has components in both the z and y directions; and at 0.20 mT, the "paramagnetic" behavior of χ_z shows that the spins are leaning toward the external magnetic field, while the small increase in χ_y indicates an antiferromagnetic component of magnetization parallel to the y axis.

These spin arrangements, labeled AF1, AF2 and AF3, are illustrated schematically in figure 3, which shows the field-vs-entropy diagram of copper. The colored areas are regions where a first order phase change takes place.



Phases of copper as they appear in a plot of the external magnetic field against entropy. The three antiferromagnetic phases, AF1, AF2 and AF3, are shown, as are their spin arrangements relative to an applied field that points up, along the z axis. The y axis in the spin configuration is to the right; the x axis is out of the plane of the paper. (From reference 5.) **Figure 3**

Neutron diffraction measurements on copper

The difficult neutron diffraction experiments were carried out in the DR-3 reactor hall at Risø by a collaboration between the Hahn–Meitner Institute of West Berlin, Helsinki University of Technology and Risø. Participating in this project were Kurt Clausen, Matti Huiku, Teppo Jyrkkiö, Kazuhisa Kakurai, Jørgen Kjems, Lounasmaa, Konrad Siemensmeyer and Michael Steiner.[7]

Because susceptibility measurements probe the spin system macroscopically at a wavevector of zero only, they give no detailed information about the ordered structures. The proper technique is neutron diffraction, because the scattering cross section, which is the result of the strong interaction between the nucleus and a neutron, is spin dependent. The long-range order of nuclear spins in an antiferromagnetic state gives rise to additional Bragg reflections, yielding directly the translational symmetry of the ordered state.

Copper has an fcc lattice that allows reflections only when the Bragg indices (hkl) are all even or all odd. Antiferromagnetic ordering of the nuclear spin system gives rise to additional peaks with (hkl) mixed. With no external magnetic field, all theoretical calculations predict an antiferromagnetic structure where the magnetic translation period is equal to the cubic lattice constant; this yields a (100) Bragg reflection in the ordered phase. For a larger magnetic unit cell, nonintegral indices would appear.

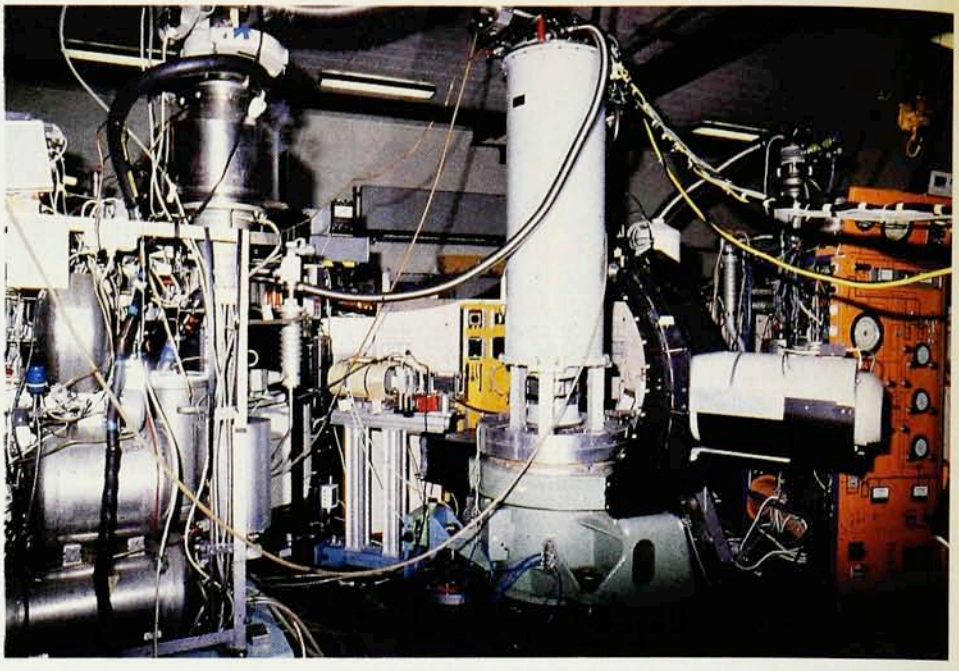
The cryostat and detector that we used at Risø (figure 4) may be moved independently in the scattering plane before an experiment is started. Instead of using natural copper as in the Helsinki work, we used Cu^{65} as the sample material at Risø to gain a factor of seven in the neutron scattering intensity. The specimen was mounted so that its longest edge, parallel to the [011] crystallographic direction, was aligned with the external magnetic field.

Applying a flux of 10^5 neutrons/cm^2 sec reduced the spin–lattice relaxation time to about 20 minutes. This was due to the 1 nanowatt beam heating, mainly caused by prompt gamma rays and by the beta decay of the nuclei formed in the neutron capture process. The experiments monitored the intensity of the (100) Bragg reflection after the sample was cooled below the ordering temperature; the longitudinal susceptibility χ_z was simultaneously measured.

Figures 5a and b show the neutron intensity and longitudinal susceptibility as functions of time after the field has been reduced to zero. For the first minute the neutron signal shows a small increase. Then, during the period of the susceptibility plateau, the neutron count diminishes rapidly, indicating a fast decrease in the antiferromagnetic sublattice polarization as the nuclei warm up through the spin–lattice relaxation process. Later measurements using a linear, position-sensitive detector proved that the observed scattered intensity was a Bragg peak. Figure 5c shows clearly how the peak decreases with time.

To obtain more information about the phase diagram of copper with ordered nuclei, we also measured neutron intensities at several low fields. At 0.04 mT the qualitative behavior of the neutron count was similar to that at zero field, but the intensity was smaller. At 0.08 mT the neutron signal was further reduced. At 0.12 mT there was clear change: The neutron intensity was very high immediately after the final field had been reached and showed no increase, but a very rapid decrease at the beginning of the experiment; after about 2.5 minutes no signal was observable; this shows that a phase change was in progress. At 0.16 mT the characteristics were again somewhat different: The intensity was very high initially, but decreased much more slowly. At 0.20 and 0.24 mT the neutron signal was qualitatively the same as at 0.16 mT, but the intensity was smaller, especially at 0.24 mT.

The neutron data above 0.16 mT suggest that at



Experimental arrangement for neutron scattering work at Risø. At the center of the picture is a cryostat on a spectrometer turntable.[7] A new two-stage nuclear demagnetization refrigerator was constructed in Helsinki for the scattering experiments. Cold neutrons enter from the left and are first reflected by a graphite monochromator crystal. The neutrons, which have a 4.7 Å wavelength, then pass through a BeO filter and hit the sample in the cryostat. The scattered neutrons are detected at the right by counters provided by the Hahn–Meitner Institute. **Figure 4**

elevated fields the nuclear spins tilt toward the external magnetic field, thereby weakening the antiferromagnetic peak. Extrapolating to the field at which the neutron intensity disappears gives a critical field H_c of 0.25 mT; this value is the same as that deduced from susceptibility experiments (see figure 3). The drastic change in the neutron intensity between 0.08 and 0.12 mT probably indicates a transition in this region. The field-vs-entropy diagram of copper, according to the Risø data, would then consist of only two antiferromagnetic phases; in intermediate fields, around 0.10 mT, the antiferromagnetic order seems to vanish because the neutron signal was absent after equilibrium.

Combining the susceptibility data obtained in Helsinki[5] and the neutron diffraction results from Risø,[7] one can thus conclude that copper has three antiferromagnetic phases when the external field is in the crystallographic [001] direction, but only two phases when the field is in the [011] direction.

Susceptibility measurements on silver

Work in Helsinki on the magnetic susceptibility of silver has been carried out by Arto Annila, Pertti Hakonen, Aarne Oja, Yasumasa Takano and Shi Yin, among others. The experimental setup is very similar to that used for studying copper; the cryostat (see figure 1), for example, is the same. The sample, which again acts as the second nuclear stage, consists of 28 rectangular silver foils, 5×30 mm^2 and 25 microns thick. The interactions between nuclei in silver are much smaller than those in copper, pushing estimates for the ordering temperature into the picokelvin range. How could we reach this new record low temperature?

According to the data on copper (figure 2), the lower critical entropy S_{c1}, below which the ordered state is reached in an adiabatic demagnetization experiment, is 48% of the maximum spin entropy. Taking an additional 20% as the safety margin, one has to remove about 70% of the entropy before the second stage demagnetization. This means that the silver sample must be cooled to 200 μK in a 7 T field.

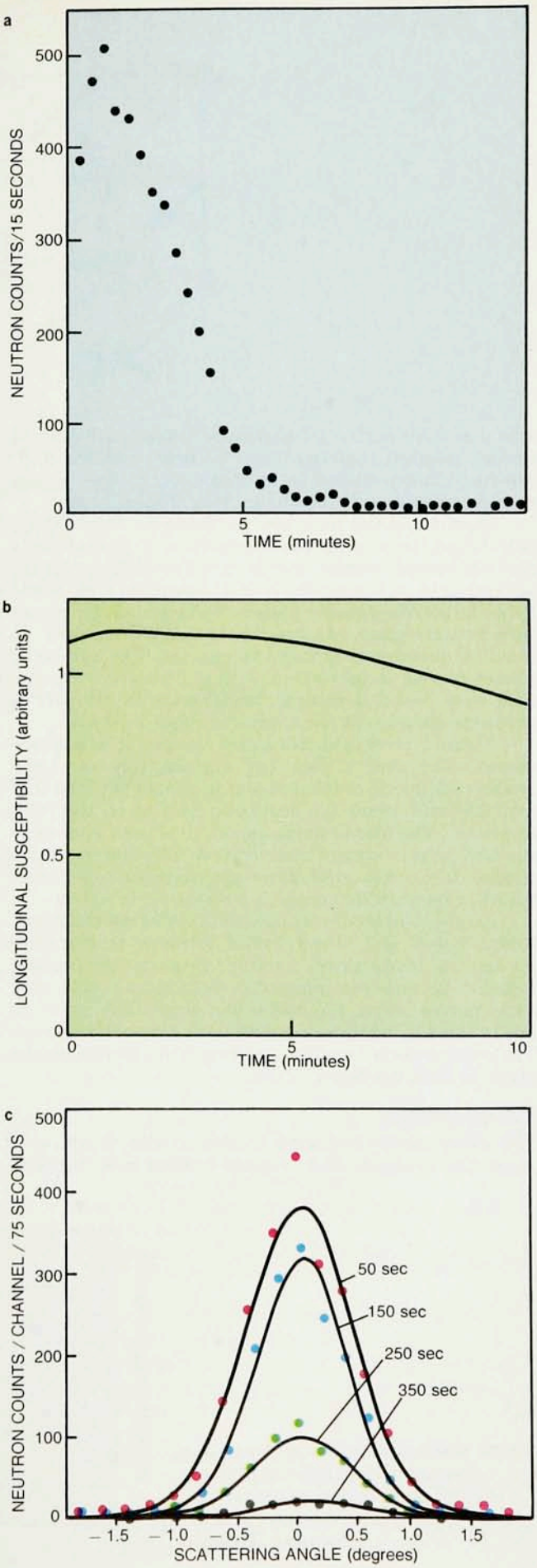
There is a new problem, however. With copper the spin–lattice relaxation time is too short for convenience, but in silver it is too long, 14 hours. This means that precooling the sample is a tedious process. Therefore, owing to the limited refrigerating capacity of the first nuclear stage and the unavoidable external heat leak, it was not possible to wait long enough to cool the sample to a temperature T_e of 200 μK.

Have we nevertheless been successful in getting silver into the ordered state? Figure 6 shows magnetic susceptibility data obtained by Annila, Oja and Takano; in this run χ seemed to remain nearly constant for about one hour. We have observed similar warm-up curves in several experiments but not in all of the runs. Slight indications of an initial increase in susceptibility have also been seen, as in figure 6. When comparing these results with observations on copper, one can claim to have at least come close to the ordering transition in silver. Unfortunately, the scatter of the data is quite large, owing to the poor signal-to-noise ratio. In any case, Hakonen and Yin recently have been able to observe 2 nanokelvins in their silver specimen; this is the lowest spin temperature ever measured.

It is possible, by a fast reversal of the external magnetic field, to get the spins into a state in which the higher energy levels are more populated than the lower ones, corresponding to a negative absolute temperature in the Boltzmann factor $\exp(-H/kT)$. We did this in the nuclear spin system of silver, while the conduction electrons remained at about 200 μK. However, a large amount of polarization was lost in the "nonadiabatic" fast passage. At negative absolute temperatures the ordered structure in silver is expected to be ferromagnetic.

Theory of nuclear ordering

A fair amount of theoretical work has been done on the structures of copper with ordered nuclei, and some has been done on silver. For example, the interactions between the nuclear spins in copper have been calculated from first principles.[8] When the phase change in copper was first observed at zero field, the nature of the transition was puzzling: A second-order process was expected but a first-order phase change was found instead. However, a renormalization group theory calculation predicts[9] a first-order transition in UO_2, as observed experimentally. This result seems to be applicable to nuclear ordering in copper, which belongs to the same universality class as the



Neutron diffraction data on copper with ordered nuclei.[7] **a:** Time dependence of the integrated neutron intensity from the (100) antiferromagnetic Bragg peak of a single crystal of Cu^{65}. **b:** Longitudinal susceptibility χ_z. **c:** Time evolution of the (100) peak with no applied magnetic field, as observed by a linear position-sensitive detector. The four 75-sec measuring intervals are centered at the times indicated for each curve, which are the best Gaussian fits to the experimental points. **Figure 5**

electronic antiferromagnet UO_2.

In general, at first the agreement between experiments and theory was rather poor; mean-field calculations on copper, for example, predicted only one antiferromagnetic phase and a transition temperature of 230 nK, four times the experimentally observed value. Much of the early theoretical work was devoted, indeed, to explaining why the observed critical temperature of 58 nK (figure 2) is so much lower than the mean-field estimate. It was then shown that fluctuations significantly lower the transition temperature.[10]

During the last three years the theoretical situation has improved dramatically. In Helsinki, for example, Oja and Hanna Viertiö have used the theory of noninteracting spin waves to investigate the ground state of nuclear spins in fcc metals at zero temperature.[11] They found that the zero-point energies lift the degeneracy of the mean-field solution; and they calculated phase diagrams that possess a rich structure that depends on the direction and magnitude of the external magnetic field and on the relative strength of the Ruderman–Kittel interaction. The ground state for a strongly exchange dominated system such as gold differs considerably from that of a system such as copper in which dipolar and exchange interactions are comparable; silver falls in between.

At Risø, Per-Anker Lindgård has employed a four-spin cluster model and second-order perturbation theory to calculate how the isotropic and dipolar nearest-neighbor interactions lift the degeneracy of the mean-field ground state.[12] This gave a phase diagram consisting of three ordered spin structures when the external magnetic field is in the [001] direction and two phases in the [011] direction, in agreement with experimental findings on copper in Helsinki and Risø.

The ground state at zero temperature is determined solely by quantum effects because thermal fluctuations vanish. However, spin wave analysis indicates that at temperatures higher than 30 nK thermal rather than quantum fluctuations make the dominant contribution to the free energy. A classical Monte Carlo calculation can thus be expected to describe the behavior of the copper spin system reasonably well at the experimentally accessible temperatures above 30 nK. Steven Frisken and David Miller at the University of New South Wales in Australia,

who first made such a calculation on copper with ordered nuclei, assumed that the magnetic field is in the [001] direction and included interactions up to the eighth-nearest neighbors.[13] They found four ordered phases.

Viertiö and Oja have also performed a Monte Carlo calculation, truncating the interactions to include only first and second-nearest neighbors.[14] For copper, with the applied field in the crystal's [001] direction, they found three antiferromagnetic phases, with first-order transitions between them, as a function of the external field. In the [011] direction they found two phases. The number of phases is thus in agreement with experiments. In zero field they found a critical temperature of 65 nK, in excellent accord with the measured value of 58 nK.

Figure 7 shows the calculated nuclear spin arrangements. The theory does not explain, however, why antiferromagnetic order is absent in copper between 0.08 and 0.12 mT when the magnetic field is in the [011] direction. The Monte Carlo method has been applied to the nuclear spin system of silver as well.[14] The transition temperature to the ordered state was found to be as low as 0.5 nK, that is, in the picokelvin range.

At the University of Bayreuth in West Germany, Frank Pobell and Georg Eska's ultralow temperature group has investigated nuclear ordering in thallium metal.[15] Because the spin–lattice relaxation time is very short in this metal, the nuclei and conduction electrons are in thermal equilibrium with each other at all times. The group reports[16] nuclear ordering into a ferromagnetic phase in thallium below 77 μK.

Future trends

The experiments discussed in this article prove, once again, the existence of a separate nuclear spin temperature.[17] The relaxation process between nuclei and conduction electrons in copper and silver is slow enough so that the spin system can be treated separately with equilibrium thermodynamics.

The measurements at Risø have shown that one can use neutron diffraction to study nuclear ordering at nanokelvin temperatures. The heat leak to the sample is serious but tolerable. A new temperature regime has thus been opened for neutron scattering experiments.

The work on copper is continuing. The (100) Bragg reflection must first be investigated with the external magnetic field in the [001] direction, where three different antiferromagnetic phases are expected. One should also try to detect neutrons at other Bragg reflections, for example (011) and ($\frac{1}{2}$00). Experiments using polarized neutrons would be very useful.

One can use nuclear magnets to investigate the crossover from order to disorder: Substitutions of the magnetic Pt^{195} by nonmagnetic platinum isotopes yield a unique system of materials for the clean study of random magnetism and spin glasses over the whole "impurity" range from 0 to 100%. The spin–lattice relaxation time in platinum is so short that the nuclei and the conduction electrons are at the same temperature throughout the experiment. The ordering temperature in platinum should be about 1–2 orders of magnitude lower than in thallium. Therefore, a three-stage nuclear refrigerator might be needed. Similar studies could be made on palladium, which also has one magnetic and several nonmagnetic isotopes.

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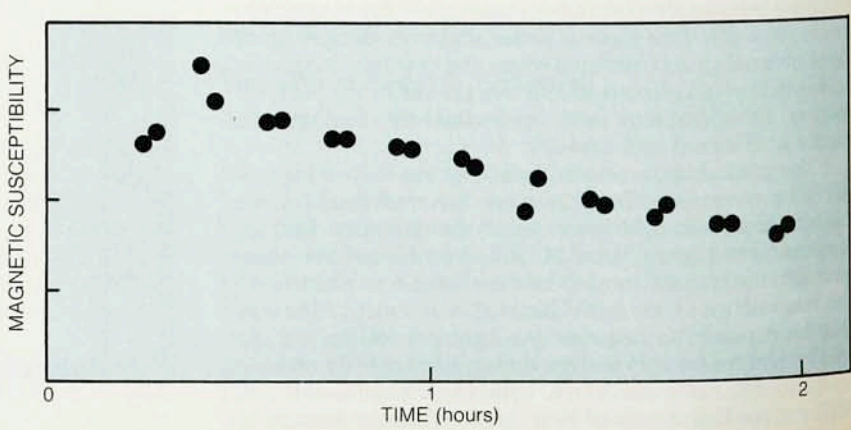
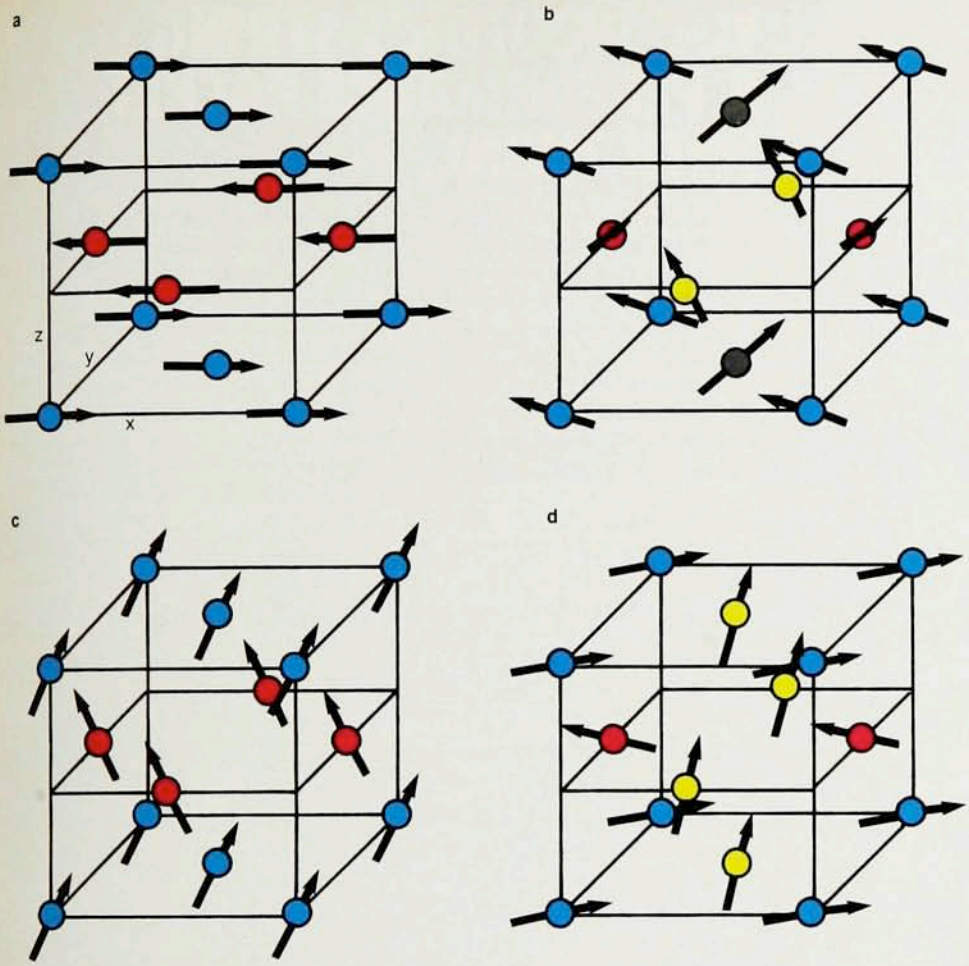


Static susceptibility of silver nuclei as a function of time after the field was turned off. (Courtesy of Arto Annila, Aarne Oja and Yasumasa Takano.) **Figure 6**



Spin configurations of copper nuclei at 16 nK, obtained from Monte Carlo calculations. **a:** Spin arrangement in zero field. **b:** Arrangement in a 0.15-mT field parallel to the [001] crystallographic direction. **c:** Arrangement in a field close to the critical field of 0.25 mT and parallel to [001]. **d:** Arrangement when the field is between 0.05 and 0.25 mT and in the [011] direction. Spins in the same direction are represented by the same color. (From reference 14.) **Figure 7**

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