Observation of Nuclear Antiferromagnetic Order in Copper by Neutron Diffraction at Nanokelvin Temperatures

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Long-range nuclear antiferromagnetic order in the nuclear-spin system of a ^{65}Cu single crystal has been observed by neutron diffraction. By use of a linear position-sensitive detector, the time evolution of the antiferromagnetic (100) Bragg peak was followed during the warmup of the nuclear-spin system. The peak intensity was found to depend strongly on the external magnetic field between $B=0$ and the critical field $B_c=0.25$ mT.

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In this Letter we report the first observation of spontaneous nuclear order in an elemental metal by neutron diffraction. Our study was made on a ^{65}Cu single crystal. Since the ordering takes place at about 60 nK, a new temperature regime has been opened for neutron diffraction experiments.

Elemental copper represents one of the simplest systems for the study of nuclear magnetism. It is a well understood metal, and the nuclear spins interact via the known dipolar and indirect conduction-electron–mediated Ruderman-Kittel interactions. In general, nuclear magnetism differs from its electronic counterpart in several important ways: The nuclear magnetic moments are highly localized and thus represent point probes for the interactions, the energy scale of importance is extremely small, and, hence, the critical temperature of the spontaneous nuclear ordering is lowered by several orders of magnitude. Finally, at low temperatures the nuclear-spin system in copper is only weakly coupled to the lattice; thus the nuclear spins can be cooled to the ordered state by adiabatic demagnetization while the lattice and the conduction electrons are kept at a relatively high temperature.

Experimental evidence for spontaneous antiferromagnetic ordering in copper was found in studies of the low-frequency ac susceptibility on a polycrystalline sample at temperatures below $T_N \approx 60$ nK.[1] In later experiments on a single crystal, all three Cartesian components were studied, and an entropy-field phase diagram was deduced.[2] It showed three distinct regions and an upper critical field of 0.25 mT. The theoretical aspect of these results has been dealt with in a series of papers,[3] but none of them has provided a full understanding of the phase diagram. An interesting aspect is connected with the fcc structure, which may lead to frustrated spin configurations.[4]

For a full understanding of any magnetic system it is fundamental to establish the magnetic ground state. For this purpose neutron diffraction is unique. For *nuclear* magnets it is the spin-dependent part of the *nuclear* cross section which makes possible the determination of ordered spin arrangements.[5] The previous neutron-scattering studies of nuclear ordering comprise the work on LiH by Roinel *et al.*[6] and on ^{3}He by Benoit *et al.*[7] In these two systems the experimental conditions and the physics of ordering are very different from those in copper. After demagnetization in the rotating frame LiH orders in a strong magnetic field, and in ^{3}He the ordering is due to particle exchange, which is a strong quantum effect.

The aim of the present neutron-scattering experiments is to determine the ordered ground state in elemental copper. The indirect Ruderman-Kittel interaction and the dipolar interactions are competing, since they favor antiferromagnetic and ferromagnetic structures, respectively. However, all theoretical calculations for the zero-field case predict an antiferromagnetic structure, where the magnetic translation period is equal to the cubic lattice constant. This leads to the appearance of (100) superlattice diffraction peaks in the nuclear ordered phase, and the spin structure is characterized by planes perpendicular to the propagation vector, which has a net spin component that alternates from one plane to the next.

The experiments were performed at the HMI-TAS8 diffractometer, located at the neutron guide from the cold source in the DR-3 reactor at Risø. A wavelength of 4.7 Å was selected by a vertically curved pyrolytic graphite monochromator. The beam was filtered for higher-order contamination by means of a 20-cm cooled BeO filter. For most of the experiments a linear position-sensitive detector was used to measure the Bragg peak profile, since mechanical movements of the spectrometer would heat the sample.

A new two-stage nuclear demagnetization cryostat, especially designed for studies of nuclear magnets by neutron diffraction, was constructed for these experiments, and based on the principles of a similar apparatus in Helsinki.[8]

^{65}Cu was chosen as the sample material because the large spin-dependent part of the scattering length[9] for this isotope yields a gain factor of 7 in the scattered intensity as compared to natural copper. Our specimen is a slablike single crystal with approximate dimensions of $35\times7\times0.6$ mm^3 with the $[01\bar{1}]$ direction along the longest edge.

In order to reach all cubic directions, the sample was mounted with the [100] and [011] axes in the scattering plane. The external field, produced by a split-pair magnet, was aligned along the $[01\bar{1}]$ axis within 4°. In these experiments the crystal was oriented for the measurement of the (100) Bragg peak by use of the (200) reflection and the $\lambda/2$ component, which is otherwise removed from the beam by the BeO filter.

During our experiments the specimen was fully polarized in a $B=4.5$-T field by the adiabatic demagnetization of the first nuclear stage to about 100 μK. The final cooling of the sample spins was obtained by demagnetization from 4.5 T to a field below 0.3 mT, while the lattice was kept in thermal contact with the first nuclear stage. After the final demagnetization, the nuclear-spin temperature begins to relax towards the electron temperature T_e. We measured, in the absence of the neutron beam, spin-lattice relaxation times $\tau_1 \approx 40$ min at $T_e = 80$ μK. In the neutron beam, with a flux of $\approx 10^5$ n/cm^2 s, τ_1 was reduced to about 20 min. This is due to an increase in T_e of the sample, resulting from beam heating.[10] We estimate, from the observed increase in the relaxation rate and from the measured thermal conductivity between the specimen and the first nuclear stage, that the beam heating is about 1 nW, a value which agrees well with calculations.[11]

The measurements were performed as follows: After the sample had been cooled into the ordered phase, the scattered-neutron intensity was monitored as a function of time. Simultaneously, we measured the static susceptibility $\chi'(0)$, in the direction of the external magnetic field, by means of a pickup coil located around the lower part of the sample which was not exposed to the neutron beam. As a result of field inhomogeneity and stray fields the general accuracy in the values of B is $\approx \pm 0.01$ mT. To reduce heating caused by the neutrons, the beam was opened only just before the final experimental field was reached.

Reliable estimates for the spin temperature in the ordered state are not available yet since it would require a full understanding of the magnetic structures and the thermodynamics of the system. Hence the data will be presented as a function of time, the only fixed point being the previously reported zero-field ordering temperature of 60 nK. Our neutron diffraction data for $B=0$ are shown in Fig. 1. The upper part illustrates the time dependence of the intensity integrated over scattering angle 2Θ. During the first minute the signal increases slightly. After this the neutron counting rate diminishes indicating a decay in the antiferromagnetic sublattice polarization as the nuclei warm up through the spin-

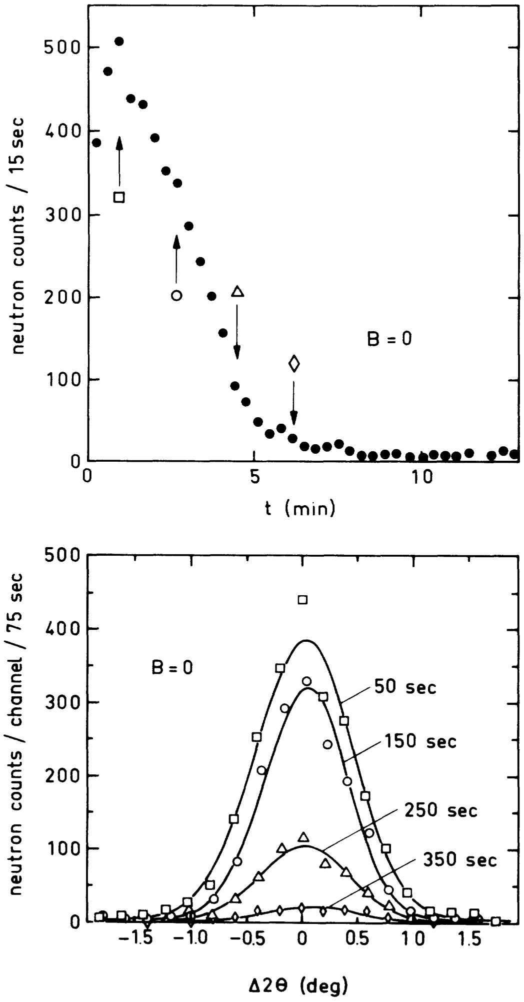

FIG. 1. Upper: The time dependence of the integrated neutron intensity from the (100) antiferromagnetic Bragg peak after zero field was reached. Lower: The time evolution of the (100) peak, as observed in the linear position-sensitive detector. The four 75-s measuring intervals are centered at the times indicated for each curve by the corresponding symbol in the upper part of the figure. The solid curves are the best Gaussian fits to the experimental points.

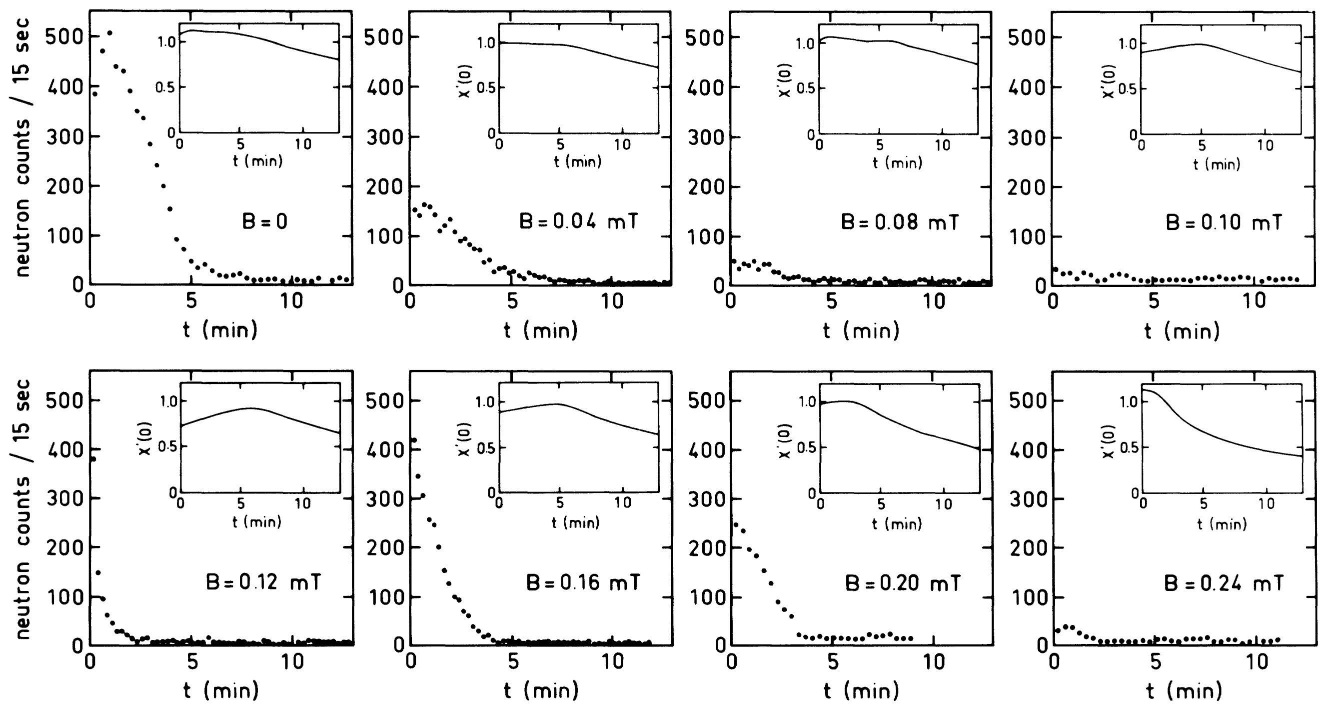

FIG. 2. Integrated neutron intensity and susceptibility $\chi'(0)$ as functions of time after final demagnetization to the field indicated on each frame; $\chi'(0)$ is in arbitrary units. The linear detector was employed at $B=0$, 0.10, 0.20, and 0.24 mT. The efficiency of this detector at $\lambda=4.7$ Å was measured to be about 10% less than that of the single detector used in the other fields.

lattice relaxation process. 5 min after the end of the final demagnetization the decrease of the signal becomes slower, and after 7–8 min the background level was reached.

To demonstrate that the scattered intensity is, indeed, a Bragg peak, we used a linear position-sensitive detector. The time development of the peak is shown in the lower part of Fig. 1 at four subsequent 75-s measuring periods.

To obtain more information about the phase diagram for nuclear ordering in copper, experiments were also made at several nonzero external fields. Special care was taken to maintain similar initial conditions in each experiment before demagnetization to the ordered phases. We estimate that, just before entering the ordered states, the initial polarization was $p_i=0.96\pm0.01$, corresponding to $S_i=0.1R\ln4$; $R\ln4$ is the maximum entropy of a spin-$\frac{3}{2}$ system.

Our experimental data showing the variation of the neutron intensity and the static susceptibility $\chi'(0)$, as functions of time after reaching the final field, are presented in Fig. 2. At $B=0.04$ mT the qualitative behavior of the neutron signal is the same as at $B=0$, but the intensity is less. At $B=0.08$ mT the neutron intensity is further reduced. In all these fields $\chi'(0)$ is similar, showing us for the first 4-5 min almost a plateau, indicating an antiferromagnetic state as in previous observations.[2] At $B=0.10$ mT the neutron intensity is almost zero during the entire experiment, while in contrast to the case at lower fields, the susceptibility shows a clear increase for the first 4 min. At $B=0.12$ mT the neutron data are drastically different from those at 0.10 mT. The intensity is very high, as at $B=0$, immediately after the final field has been reached and, in contrast to lower fields, shows no increase but a very rapid decrease at the beginning of the experiment; after about 2.5 min no neutron signal is observable. The susceptibility increases almost 20% during the first 4 min. The neutron intensity thus disappears clearly before the system reaches the susceptibility maximum. At 0.16 mT the characteristics are again somewhat different. The neutron intensity is very high initially, as at 0.12 mT, but decreases more slowly. The disappearance of the neutron signal is now coexistent with the maximum of $\chi'(0)$. The behavior of the susceptibility is qualitatively the same as at $B=0.10$ and 0.12 mT, showing a clear increase in the beginning. At $B=0.20$ and 0.24 mT the neutron signal is similar to that at $B=0$, but the intensity is smaller especially at 0.24 mT. The susceptibility increase at $B=0.16$ mT is reduced to a plateau at $B=0.20$ mT, and at 0.24 mT only a short nonexponential decay was observed initially. Finally, at $B=0.30$ mT (not shown in Fig. 2) we found no signs of ordering; thus no neutron intensity was seen and an exponentially decreasing susceptibility signal was observed.

Our neutron data above $B=0.16$ mT suggest that, at elevated fields, the nuclear spins tilt towards B, as in a magnetic spin-flop phase. Thereby the contribution to the antiferromagnetic peak becomes weaker. By extrapolating to the field at which the neutron intensity disappears, one obtains a critical field $B_c \approx 0.25$ mT, as observed before.[2]

The susceptibility measurements in Helsinki differ from the present work in two ways: The earlier sample was of natural copper and the magnetic field was applied along [001], whereas we have used a ^{65}Cu sample and a $[01\bar{1}]$ field direction. In spite of these differences, the magnetic field dependence of $\chi'(0)$ is very similar. According to theory[3] the ordering is not affected significantly by the 7% difference in the magnetic moments of the two isotopes.

Our neutron data confirm that at least two antiferromagnetic phases characterized by the (100) reflection exist in copper, one at the lowest fields and another at fields close to B_c. The rapid transient variation and the low level of the (100) neutron intensity at intermediate fields may be due to a third phase as was concluded previously.[2] The initial increase in the neutron signal during the first minute observed at low fields may be due to a long nucleation time for this phase[2] and will be investigated further. More experiments close to the proposed phase boundaries and at other reflections are needed to map out the phase diagram fully.

We have demonstrated that neutron diffraction can be used for studies of nuclear ordering even at nanokelvin temperatures, and unambiguously proved antiferromagnetic ordering in copper. Our studies on the magnetic field dependence of the (100) antiferromagnetic Bragg reflection show that at least two antiferromagnetic phases exist in copper below $B_c = 0.25$ mT.

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