ICANS-XI International Collaboration on Advanced Neutron Sources
KEK, Tsukuba, October 22-26, 1990

Neutron Scattering at Ultra-low Temperatures

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Abstract

Two examples of neutron scattering experiments at ultra-low temperatures are described where the spin-dependent part of the nuclear scattering is used.

Introduction

In the very beginning of the application of the thermal neutron scattering, all possible contributions to the scattering cross-section have been calculated theoretically[1]. Despite this fact and although Shull and Ferrier [2] demonstrated experimentally the observability of the spin-dependent part of the cross-section quite early, it has been utilized in only few neutron scattering experiments [3]. The reason for this is straightforward: Because of the smallness of the nuclear spin moment , the nuclear temperature at which the nuclear spin polarization $<I> \neq 0$ occurs, is at least three orders of magnitude lower than that needed for the electronic magnetic moment to be polarized. This mK temperature range became first commonly available after the recent development of the commercial dilution units.
By now, starting from this mK temperature range, the nuclear adiabatic demagnetization can be applied to reach even lower μK and nK nuclear temperatures. At these ultra-low temperatures spontaneous nuclear spin ordering can be expected. Remembering the unique role of the magnetic neutron scattering for investigating the electronic magnetic properties of solids, one easily can think of the importance of the spin dependent cross-section term in connection with the nuclear spin ordering and its properties. In this contribution I would like to demonstrate the importance of the neutron scattering at these low temperatures by which new and unique informations of the nuclear spin system can be obtained.

Neutron scattering cross-section

The fundamental quantities for neutron scattering at nucleus with spin I are the spin-independent scattering length

$$b_0 = \frac{b^+(I+1) + b^- I}{(2I+1)}$$

and the spin-dependent scattering length

$$b = 2\frac{b^+ - b^-}{(2I+1)}$$

where b^+ and b^- are respectively the scattering length for neutron spin parallel and antiparallel to nuclear spin. In the case of the unpolarized nuclear spins the spin-dependent scattering length gives the well known spin incoherent cross-section

$$\sigma_{inc} = \frac{1}{4} I(I+1) |b|^2$$

With $\vec{P}_n = \langle \vec{I} \rangle / I$ defined as the nuclear polarization vector, the coherent cross-section for an electronically nonmagnetic sample can be written in the following three contributions:

$$\left(\frac{d\sigma}{d\Omega}\right) = \left| \sum_{v} \exp(i\vec{Q}\vec{R}_v)\, b_{0_v} \right|^2 \tag{1}$$

$$+ \frac{1}{4} \left| \sum_{v} \exp(i\vec{Q}\vec{R}_v)\, b_v I\, \vec{P}_n \right|^2 \tag{2}$$

$$+ \mathrm{Re} \left\{ \sum_{v\mu} \exp\left[i\vec{Q}\left(\vec{R}_v - \vec{R}_\mu\right)\right] b_{0_v}^{*} b_\mu I\, \vec{P}_n \vec{P}^N \right\} \tag{3}$$

where $\vec{P}^N$ represents the neutron polarization. For the simplicity only the special (but most common) case of the nuclear polarization along the external field is considered here for the term including the neutron polarization P^N (Term (3)).
The first term is the spin-independent part, the second is the term purely due to the nuclear polarization and the third term is the interference term of the first two contributions. As can be seen the third term can be only observed if $P^N \neq 0$, i.e. only with the use of polarized neutrons.

In the following the observation of the nuclear spin ordering in Cu through the second term and the polarized neutron thermometry using the third term will be described as examples for the use of the spin-dependent scattering length.

Nuclear spin ordering in Cu

(The details of this experiment are given in ref. 4)
The first experimental evidence for spontaneous antiferromagnetic (AF) nuclear spin ordering in Cu was found in studies of the low-frequency ac susceptibility on a polycrystalline sample at temperatures below 60nK[5] and subsequently the AF order in the field was studied in detail on a single crystal[6]. The latter experiment yielded three different regimes in the field and entropy phase diagram. To obtain more insight into the complicated phase diagram, a direct study of the periodicity of the AF order is mandatory, which can be only performed with neutron scattering using the spin-dependent cross-section term (2).

The Hamiltonian of the Cu nuclear spin system consists out of the indirect Ruderman-Kittel and the dipolar interaction. All theoretical calculations for the zero-field case predict an AF structure where the magnetic translation period is equal to the cubic lattice constant. This would lead to the appearence of (100) superlattice peaks in the nuclear spin ordered phase. A two axis neutron diffractometer was specially modified for this ultra-low temperature application, including the vibration damping and the position sensitive detector, and installed at the neutron guide from the cold source in the DR-3 reactor at Risø. A new two-stage nuclear demagnetization cryostat with the split type magnet for the second stage was constructed to allow the neutron beam access to the sample. ^{65}Cu was choosen as the sample material because of the large spin-dependent scattering length for this isotope. The slablike single crystal with the approximate dimensions of 35*7*0.6 mm^3 with the [011] direction along the largest edge. [100] and [011] axes were oriented into the scattering plane and the external field, a vertical field, was applied along the [01$\bar{1}$] direction. The whole nuclear demagnetization stage is precooled by the dilution unit. Using the first demagnetization stage the sample in the field of 4.5T is cooled to a lattice temperature of about 100mK. The demagnetization of the second stage (the sample) from 4.5T to a field below 0.3mT cooles the nuclear spin system to the final temperatures $T<T_N$=60nK. After the final demagnetization the nuclear spin temperature begins to relax towards the electron temperature. The spin-lattice relaxation time in the experiment with a neutron flux of 10^5 n/cm^2/sec was ~20min. which allowed the observation of the ordered phase for 7~8 min.. Fig.1 shows the time evolution of the (100) peak after the demagnetization to the zero field, as observed in the position sensitive detector. The decay of the AF nuclear spin Bragg peak as the nuclei warm up with time is clearly seen. To obtain more information about the phase diagrm, experiments were also performed at nonzero external fields. In fig.2 the neutron intensity contour diagram of the (100) Bragg peak as a function of time and external magnetic field is shown. It is noteworthy that there is a region around 0.07mT where the (100) intensity is effectively zero, although the susceptibility measurement carried out simultaneously clearly indicates AF ordering. This is a clear indication of another type of the spin ordering in this phase diagram regime. The more recent neutron scattering results indeed show that (0,2/3,2/3) type nuclear spin Bragg peaks are present in this regime[7]. The commensurate ordering vector $\vec{k}=(2\pi/a)(0,2/3,2/3)$ causing this type of Bragg peaks has not been observed in any electronic fcc AF systems sofar. In the fcc nuclear spin system Cu, the comparable size of the Ruderman-Kittel and dipolar interactions and the quantum effects might stabilize this novel ordering vector. This results add a new aspect to the old problem of the frustration in the fcc AF system.

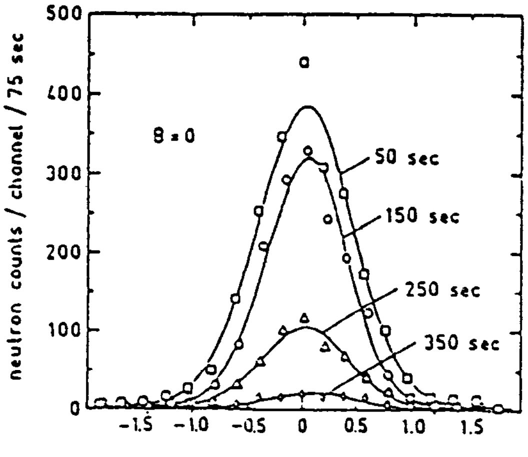

Fig.1

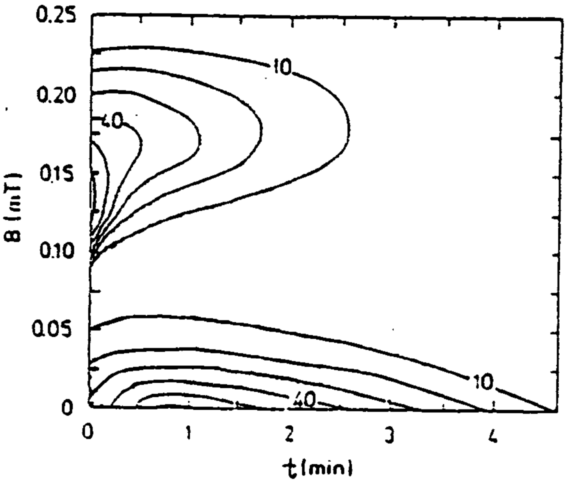

Fig.2

Polarized neutron thermometry

(The details of this method are described in ref. [8].)

In the following measurements we have used polarized neutrons to determine the nuclear temperature of Cu in the range between 100μK and 20mK. Using the cross-section formula above the Bragg intensity for the fcc lattice with four atoms per unit cell can be written as

$$I_{\pm}=16\left(b_0^2 \pm b_0 b I P_n \left|P^N\right| + \left(\frac{I^2}{4}\right) b^2 P_n^2\right)$$

for an extinction-free case.

The flipping ratio (FR), i.e. the ratio between the intensity of scattered neutrons in the two spin state then is

$$FR(P_n) = \frac{I_+}{I_-} \quad .$$

This equation gives the relationship between the nuclear polarization and the FR, a quantity which is less sensitive to experimental errors than the measurement of absolute intensities. In the paramagnetic regime the nuclear polarization P_n is related to the nuclear temperature T_n via the Brillouin function B_I:

$$P_n = B_I\left(\gamma_n \hbar I B / kT_n \right)$$

where γ_n is the gyromagnetic ratio of the nucleus and B is the applied external field.

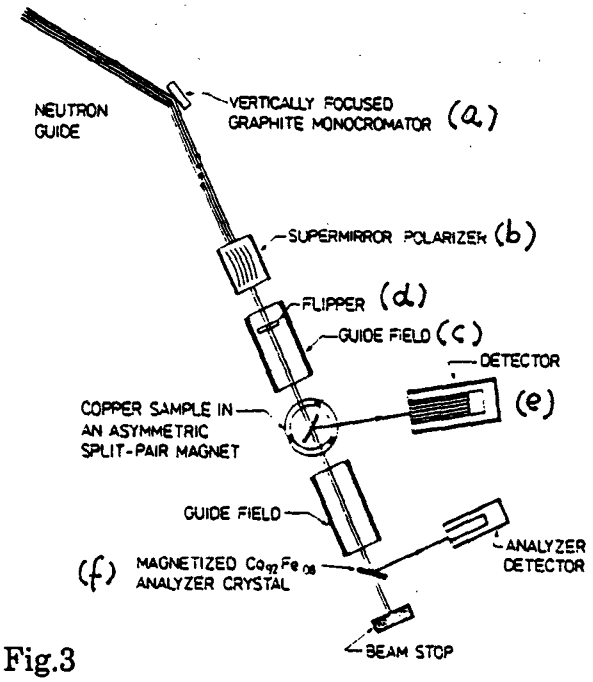

Fig.3

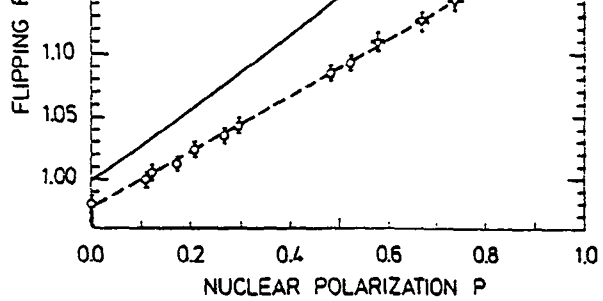

Fig.4

The following neutron diffractometer set up was used (fig.3). Neutrons from the cold neutron guide are monochromatized using a vertically pyrolytic graphite monochromator(a). The beam is then polarized by a supermirror bender (b). Permanent magnets are used to magnetize the vertical guide fields before and after the cryostat(c). A dc-flipper(d), located in the guide field in front of the cryostat , determines the + or - polarization of the beam. The Bragg reflected neutrons at the sample is detected by the main counter (e). The sample enviroment is identical to that of the nuclear spin ordering experiment mentioned above. The beam polarization is monitored by an analyser system in the through going beam behind the sample using a magnetized $Co_{92}Fe_{08}$ analyzer crystal(f).

Although this thermometry is in princip calibration free, the extinction effect of the single crystal Bragg intensity does cause the deviation of the relation $I \propto /F/^2$ and hence a calibration over the entire temperature range is required. Because of the limited space available I only show the result of such a calibration (fig.4) without going into detail but mentioning that the Co thermometry was used in the B/T range of B<4T and T>3mK. For even larger B/T-values the temperatures were determined by extrapolating back the warm-up curve to the measurable temperature after having first cooled the system below 1mK. The more interested readers are referred to the original paper [8]. The calibration depicted in fig.4 is for $\lambda = 2.4$ Å. The solid curve is the calculated extinction free curve and without the μmetal correction, which causes the measured curve not intersepting FR=1 at P_n=0. Once this calibration is done the nuclear polarization, hence the nuclear temperature at given external field, can be determined by measuring the FR.

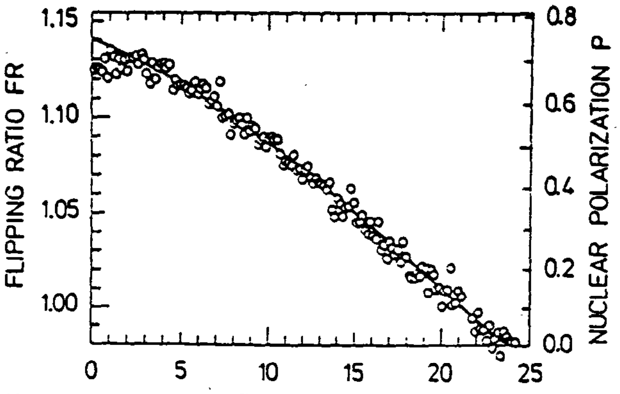

Fig.5

As an application a typical heat leak measurement of the Cu system is shown in fig.5. This measurement was carried out in an external field of 1T and it covers temperatures from 0.5 mK upwards. The curve through the experimental points corresponds to a constant heat leak of $\dot{Q}$=14nW, which describes the experimental data well over the 24 hours. For studies of nuclear ordering such an heat leak measurement is of extreme importance to reduce the heat input to the nuclear stages to a very low level.

To conclude I have picked up two examples to demonstrate the use of the spin dependent part of the nuclear scattering length. Unique and important informations on the nuclear spin system can be obtained. To utilize this possibility neutron scattering at ultra-low temperatures is indispensable.

Acknowledgement
The results described here were obtained in the joint low temperature neutron scattering project with Low Temperature Laboratory at Helsinki University of Technology, Finnland, Neutron Scattering Group at Risø National Laboratory, Denmark and Neutron Scattering Group at Hahn-Meitner-Institut, Berlin. In this context I thank K.N. Clausen, M.T. Huiku, T.A. Jyrkkiö, J.K. Kjems, O.V. Lounasmaa, K. Siemensmeyer and M. Steiner without whose help this contribution could not have been written.

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Q(R.Pynn): Do you think there is any possibility of doing inelastic scattering experiments from nuclear ordered systems?

A(K.Kakurai): To be honest, I haven't done any quantitative collection. But it will be difficult because of the following two reasons:

1) Expected low excitation energies in these systems and hence an high energy resolution needed,
2) Low intensities combined with the limited flux available for the measurement.