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Magnetic diffraction in solid ^3He

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Résumé. — L'observation d'un signal de diffraction magnétique d'un cristal cubique centré d' ^3He montre directement l'existence d'un ordre antiferromagnétique de vecteur de propagation $(1/2, 0, 0)$.

Abstract. — Magnetic diffraction in a bcc single crystal of ^3He directly proves the occurrence of an antiferromagnetic ordering with a propagation vector $(1/2, 0, 0)$.

The originality of the magnetic ordering of solid ^3He is that the large zero point motion leads to atom-atom exchange processes. This mechanism involves the whole motion of the nucleus with its electronic shell. Thus solid ^3He represents one of the more attractive example of a magnetic coupling correlated with the direct displacement of the atoms. Evidence of the atom exchange is given by an ordering temperature $T_N \sim 1 \text{ mK}$ [1] three orders of magnitude greater than that predicted for dipolar magnetic nuclear interactions. The tunnelling origin of the exchange leads also to a drastic diminution of T_N with the decrease of the volume (V) : $T_N \sim V^{1/3}$ [2]. Interest in the magnetic studies is reinforced by the fact that an ordinary nearest neighbour Heisenberg exchange model fails to explain the magnetic behaviour at low temperature ($T \lesssim T_N$) although it seems to describe the high temperature experiments. Due to the hard core potential, triple and four spin exchanges are important [3, 4]. Recently, it has been proposed [5] that a new magnetic ordering (a spin nematic state) may occur with the particularity that the atoms may carry no sublattice magnetization below T_N at the opposite of the case of antiferromagnetic structures. Only neutron scattering experiments can answer whether or not a magnetization exist on each site below T_N .

Neutron diffraction is an unambiguous way to determine the existence of a sublattice magnetization and a magnetic structure. In the present case of nuclear moment ordering, magnetic diffraction would occur not through the usual dipole-dipole interaction which is too weak, but through the nuclear interaction which is spin dependent. The difference between the values b_+ and b_- of ^3He recently measured [6] is enough for a superstructure magnetic Bragg reflection to be detected. However, performing this experiment on solid ^3He below 1 mK is a challenge.

The magnetic ordering occurs at very low temperature. The huge neutron- ^3He absorption cross section implies a detection of a weak diffraction signal and a drastic sample heating by the neutron beam. Consequently the magnetic phase can be observed only for a short time [7]. We report the first observation of magnetic diffraction on a bcc single crystal of solid ^3He .

1. The experimental background.

1.1 THE NEUTRON SPECTROMETER. — The experiment has been performed on the polarized neutron spectrometer DN2 installed at the reactor Melusine of the « Centre d'Etudes Nucléaires de Grenoble ». The beam is made monochromatic and polarized by an Heussler crystal. Its wavelength, $1.74 \text{ \AA}$ has been chosen large enough to produce larger intensities on the magnetic Bragg reflections of ^3He , and to reduce the background around these low angle reflections. A set of filters (Al_2O_3 single crystal, Sm_2O_3 powder) permits use of either the $\lambda = 1.74 \text{ \AA}$ beam (polarized) or the $\lambda/2 = 0.87 \text{ \AA}$ beam (practically unpolarized) to search for magnetic or nuclear reflections.

Two counters are operated simultaneously (Fig. 1) : one (C_T) for transmission measurements and the other (C_D), which can be tilted above the basal plane, for Bragg reflection intensities.

A concrete block with a large hole in its centre is mounted above the spectrometer. It lies on three pillars with rubber suspension. It eliminates vibrations and supports the rotating mechanism for the sample cryostat.

1.2 THE ULTRA LOW TEMPERATURE CRYOSTAT. — Ultra low temperatures are obtained in two stages. The first stage consists of a dilution refrigerator which allows a pre-cooling down to 10 mK . The second stage is a nuclear demagnetization of copper which allows the sample container to cool down to 0.5 mK . It is made of a bundle of 10 moles of copper wires magnetized in a field of 8 T . The 2 stages are connected by a superconducting thermal switch made of pure lead wires, which conducts heat when the field is on and does not conduct when the field is off.

Fig. 1. — View of the experimental set up. N_0 is the incoming neutron beam, M an Heussler crystal monochromator, RF a resonant flipper, F filters for producing unpolarized ($\lambda = 0.87 \text{ \AA}$) or polarized ($\lambda = 1.74 \text{ \AA}$) beam, MG the magnetic guide, C_D and C_T the counters used respectively for the measurement of a (h, k, l) reflection and of the flipping ratio. A field H of 80 mT polarizes the ^3He target.

2. Controlling the experimental parameters.

Observing a magnetic Bragg reflection on a ^3He single crystal, cooled down below 1 mK, requires that many difficulties be brought under control.

2.1 CONTROL OF THE ^3He CRYSTAL GROWTH AND ORIENTATION. — The ^3He single crystal has to be grown *in situ* in a flat copper cell, under pressure, at very low temperature. Furthermore, at $\lambda = 1.74 \text{ \AA}$, the desirable thickness is less than 0.1 mm. To grow such a crystal, we have used a copper cell and filled it with liquid ^3He through a very thin capillary. The geometry of the cell does not allow solidification at constant pressure because it implies a continuous flow of liquid in the cell. Crystallization occurs then at constant volume with a continuous decrease of pressure from 44 bar at $T = 1.15 \text{ K}$ to 34 bar at 0.8 K [8].

The orientation of such a crystal is not known. It is determined by a systematic search for the nuclear reflections of type (110), by rotating the cryostat around the vertical axis, and this for the different positions of the counter C_D above the horizontal plane, along the Debye-Scherrer cone.

2.2 CONTROL OF THE ^3He MOLAR VOLUME. — As the Néel temperature decreases dramatically with the molar volume ($T_N \sim V_M^{1/3}$) [2] it is of the utmost importance to grow a crystal as close as possible to the melting curve at low temperature. It is therefore necessary to control accurately the ^3He molar volume in the experimental cell. This is done by taking advantage of the very large absorption cross section of ^3He for neutrons : transmission measurement gives a direct calibration of the molar volume in the target when the thickness of the cell is known [7]. The determination of the molar volume by adjustment of the capillary pressure is then easy. Such a measurement also permits following any change of the molar volume during the crystallization process due to a possible displacement of matter from the cell to the capillary.

2.3 CONTROL OF THE ^3He CRYSTALLIZATION IN A SILVER SKELETON. — Cooling solid ^3He and thermal equilibrium in the copper cell is made very difficult by the Kapitza resistance which increases dramatically at low temperature [7]. The only way to overcome these difficulties was to increase the exchange area between the metallic cell and the solid ^3He . We, therefore, filled the copper cell with pressed sintered silver powder and grew the helium crystal in the holes of the powder [9]. Thermal conductivity is thus provided by the metallic silver skeleton while the ^3He single crystal, in the holes between the silver grains, forms a continuum which extends over the whole experimental cell. Thermal equilibrium below 1 mK is obtained after a few hours as the exchange surface has been enhanced and the cooling distances inside helium have been reduced to the dimension of the holes.

The striking phenomena is that a single crystal grew systematically in a silver powder of a 0.3μ mean grain size for a cooling rate lower than 50 mK/hour [10]. The packing factor was 50 % and the thickness of the ^3He crystal was found equal to 0.08 mm by neutron transmission.

2.4 CONTROL OF THE ^3He TEMPERATURE. — It is of the greatest importance to follow the temperature of the solid helium sample, at each stage of the experiment preparation. At these very low temperatures, the use of an external thermometer (cobalt for nuclear orientation, platinum for NMR) is very delicate for geometrical reasons, and the thermal equilibrium between the helium sample and the external thermometer is not guaranteed.

Instead, we preferred to use the neutron beam itself to measure the temperature. As the neutron absorption cross section of ^3He is spin dependent [11], we have measured the flipping ratio R_T (the ratio of the transmission of incoming neutrons polarized parallel to antiparallel with respect to the nuclear polarization of ^3He nuclei) by the counter C_T in the transmitted polarized beam (Fig. 1). It gives a direct determination of the average magnetization if ^3He are polarized per-

manently by a small magnetic field ($H = 80$ mT). The flipping ratio is directly correlated to the spin temperature. As a discontinuity in the susceptibility appears at the first order transition, the complete crossing of the sample through the transition is detected with a high sensitivity.

3. Search for the magnetic structure.

Usually, when the experiments can be performed at thermal equilibrium, the long range structure is sought by a complete exploration of the reciprocal space. Such a study is prohibited here by the fast warming of the sample. The choice was made to select one particular point of reciprocal space and to make a temperature scan to test the evidence of the magnetic reflection.

The search is made easier if complementary information is given as to possible magnetic structures. The NMR experiments [12] have proved the breakdown of cubic symmetry below T_N and the occurrence of a new 4 fold symmetry axis $(1, 0, 0)$ corresponding to the observation of three domains. The simplest structure agreeing with this symmetry is the udd phase with $(1, 0, 0)$ ferromagnetic planes arranged in the sequence two spin up planes followed by two spin down planes [12]. Its corresponding wavevector is $(1/2, 0, 0)$.

After orientation of the crystal by searching for $(1, 1, 0)$ reflections, the detector (C_D) is moved to the selected position and the sample cooled below T_N . At the beam opening (time t_0), the diffraction signal (N) of the counter C_D and the flipping ratio measured by transmission with the counter C_T are simultaneously recorded. For C_D located on a $(1/2, 0, 0)$ reflection, these two measurements are shown on figure 2.

Fig. 2. — Simultaneous time variation (a) of the neutron counts N in 80 seconds ($\bar{\Gamma}$) measured by the counter C_D on a $(1/2, 0, 0)$ reflection and (b) of the measured flipping ratio R_T ($\bar{\Gamma}$) detected by an experiment with polarized neutrons. The theoretical flipping ratio in (c) corresponds to the susceptibility measurements of reference (2). The average signal and its rms deviations ($\bar{\Gamma}$) in (a) is obtained by a linear regression. The dashed line in (b) represents the flipping ratio estimated from (c) assuming a linear time conversion of the nuclei at T_N and the thermal equilibrium after the maximum at $t_1 - t_0 \sim 500$ s.

The measurement of the flipping ratio proves that, at the beam opening, the ^3He nuclei are in a magnetic phase. It gives also a direct measurement of the time t_1 during which the ^3He atoms become almost completely paramagnetic. From t_0 to t_1 , with a constant neutron flux, there is a linear conversion of the ordered phase to the paramagnetic phase at the ordering temperature due to the large discontinuous jump of the entropy at the first order transition. If a magnetic signal exists, it must coincide with a linear decrease from t_0 to t_1 while a constant background must be recovered after t_1 .

In figure 2, the difference between the measured flipping ratio and that estimated assuming a linear transformation of the ^3He atoms to T_N is due to the approximation that the nuclei has reached a thermal equilibrium at t_1 with $T = T_N$. In fact, part of the sample has already reached temperatures higher than T_N at t_1 due to the high flux irradiation.

Figure 2 demonstrates the occurrence of a magnetic diffraction corresponding to a propagation vector $(1/2, 0, 0)$. A linear regression gives for the magnetic intensity $N = 8.7 \times 10^{-2}$ neutron per second with a mean quadratic deviation of 2×10^{-2} neutron per second. This corresponds to the estimation based on the $(1, 1, 0)$ nuclear intensity. Two experiments performed with $T < T_N$ at t_0 confirm this observation.

4. Conclusion.

The neutron experiment resolves without ambiguity that solid ^3He on the melting curve has an antiferromagnetic ordering with a propagation vector $(1/2, 0, 0)$. A spin nematic state is clearly ruled out [5]. The observed magnetic structure is the simplest structure compatible with previous NMR data [12]. It cannot be explained by a simple Heisenberg model but by exchange between three and four particle rings [4]. The microscopic description of ^3He is still an open problem. Our result gives a strong stimulation for searching now the ordered phase of solid ^3He in high magnetic fields.

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