DEVELOPMENT AND USE OF PULSED HIGH MAGNETIC FIELDS FOR NEUTRON DIFFRACTION EXPERIMENTS

M.Motokawa, M.Arai, M.Mino, K.Ubukata, T.Bokui and M.Fujita
Department of Physics, Faculty of Science,
Kobe University, Rokkodai, Nada, Kobe 657, Japan

abstract

Following the first report presented at the previous ICANS in Tsukuba on the development and use of pulsed high magnetic fields for neutron diffraction experiments, recent technical development and measurements of neutron diffraction in high fields are reported.

In addition to the 20 Tesla magnet whose field direction is horizontal and almost parallel to the neutron beam from the target, vertical field up to 16 Tesla is available. This is more convenient than the horizontal method to obtain a large scattering angle and a stable temperature by use of glass Dewar. A new magnet for 30 Tesla field is designed and now under construction by using a strong metal and by a new machining method. Another method by a copper wire coil which is immersed in a liquid nitrogen vessel is also tried.

After determination of the complicated magnetic structure of $PrCo_2Si_2$ in high field, we have performed to observe a field induced structure change due to the quantum effect in $CsCuCl_3$.

1. Introduction

One of the most interesting phenomena in solid state physics is considered to be the phase transition of materials induced by changing temperature, pressure or magnetic field. It is usually accompanied by a symmetry change which causes a structure transition and it is well known that the direct way to look in the symmetry of the structure is X-ray or neutron scattering. Especially the latter is useful for mapping the dispersion relations of the elementary excitations on the $\omega-k$ diagram. This is one of the reasons why the combination of neutron diffraction technique with the extreme condition of low temperature, high pressure or high field is regarded as the important and urgent subject.

Regarding the general technique for high magnetic field, it has been developed intensively in this decade and now 80 Tesla field is available without destruction in a pulsed way, while the static field is now up to 30 Tesla. These high fields are under the practical use for various experiments such as the measurements of magnetization, optics, magnetoresistance, magnetic resonance and so on. As mentioned in the previous paper [1], however, such high field techniques had been considered to be difficult to use for neutron diffraction experiment. For example, a static high field up to 30 Tesla is almost impossible to use for neutron scattering experiments because of its hugeness in both size and budget. The pulsed field had been also unuseful because the data accumulation is necessary for the neutron diffraction experiments while the pulsed field was usually single shot. So the standard method is to use a superconducting magnet with the neutron beam from a reactor. Because of the complicated configuration of equipment, i.e. installation of cryostat for the superconducting magnet, sample cryostat and goniometer, the available field has been limited lower than 10 Tesla. We considered that if the pulsed field is repeated with coincidence of the pulsed neutron beam, this method would be a breakthrough to develop the high field neutron diffraction experiments. This is the motivation of developing the repeating pulsed field. As the first step of our project, we succeeded to obtain high fields of 20 Tesla every 2 seconds [2].

2. Survey of the previous work

The profile of our equipment is as follows [1][2][3][4]:

(i) Capacitor bank
Capacitance: 1.2 mF
Maximum stored energy: 50 kJ at 10kV
Repeating frequency: less than 0.5 Hz

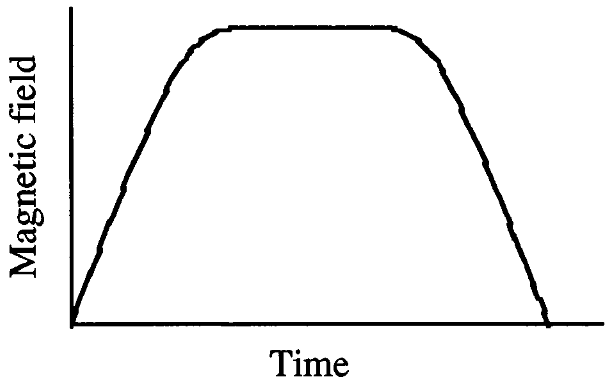
(ii) Magnet
Inductance: 80 μH
Inner diameter: 40 mm
Outer diameter: 144 mm
Maximum field: 20 Tesla at 20 kA
Duration time : 1 msecnd
Field shape: half sine curve
Flow rate of cooling water: 0.33 l/min
Temperature difference: 6°C at 20 Tesla
Life time: 20000 times at 20 Tesla
Field direction: horizontal

(iii) Cryogenics
Cryostat: Liquid helium flow type
Lowest temperature: 4.2 K
Sample size: 7×7 mm^2 and thickness of a few mm

(iv) Others
Neutron beam: Spallation neutrons at KENS
Scattering angle: $2\vartheta = 10°$
Spectrometer: TOF method

The first successful trial of measurement was the determination of the complicated magnetic structure at high field of $PrCo_2Si_2$ [3][4] which shows a metamagnetic transition at 1.2, 3.8, 6.7 and 12.2 Tesla at 4.2 K when an external field is parallel to the c-axis.

At present time, we have to apply a pulsed field at the time when the reflection corresponding to the high field structure appears in TOF method. So if we have no idea for the high field structure, it is quite difficult to estimate the time to apply a field. This problem is technically solved by using a flat-top pulsed field and a position sensitive detector as shown in Fig.1.



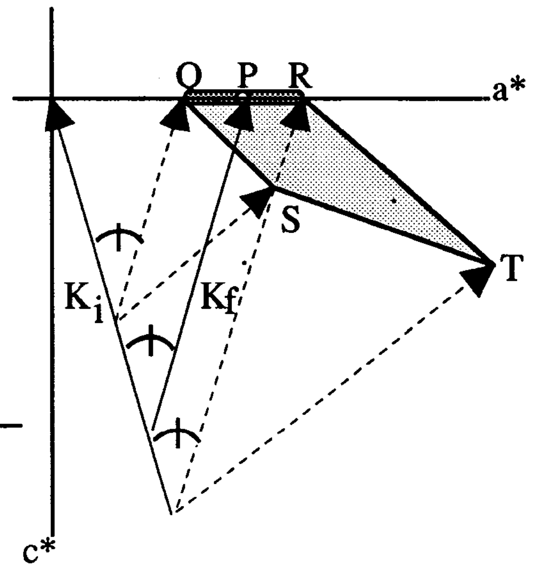


Fig.1 Shape of flat top field (left) and observable area (right). Using the present equipment, we can observe the reflection only at the point P by applying an external field. But it can be expanded to the region between Q and R by using a flat top field and more expanded to the shaded area QRST by using a position sensitive detector.

3. Vertical configuration

The magnet coil mentioned above is of the so-called Bitter type which are originally developed for static high magnetic fields. We used it for pulsed high field experiments, but when it is used for the neutron diffraction experiments, the problem is the small scattering angle [1][3]. This is one of the disadvantages of this configuration. To make a large scattering angle, we have developed a split type magnet as is shown in Fig.2 at the expense of the maximum field strength. This is set as the magnetic field is vertical. The scattering angle is arbitrary in principle but because there must be a hard spacer between the split parts of the magnet, the direction of the neutron beam is fixed as is shown in Fig.2. The available scattering angle is 30° in this case. The biggest advantage of this configuration is that a glass Dewar system is available as is shown in Fig.2. So the temperature of the specimen can be kept at 4.2 K or 1.4 K very easily and the consumption of liquid helium is small. As mentioned above, in the case of horizontal configuration, the cooling system for the specimen is a liquid Helium flow type cryostat which consumes a huge amount of liquid helium to maintain the temperature at 4.2 K. Then the sample temperature is usually kept between 5 and 10 K to save liquid helium. The disadvantage of this magnet system is the reduction of the maximum field strength. The split type magnet is also composed of Bitter disks as well. Due to the large gap at the center of the magnet, however, the available field is up to 16 Tesla. The design of this magnet is not completely finished. We still have some problems like water leak.

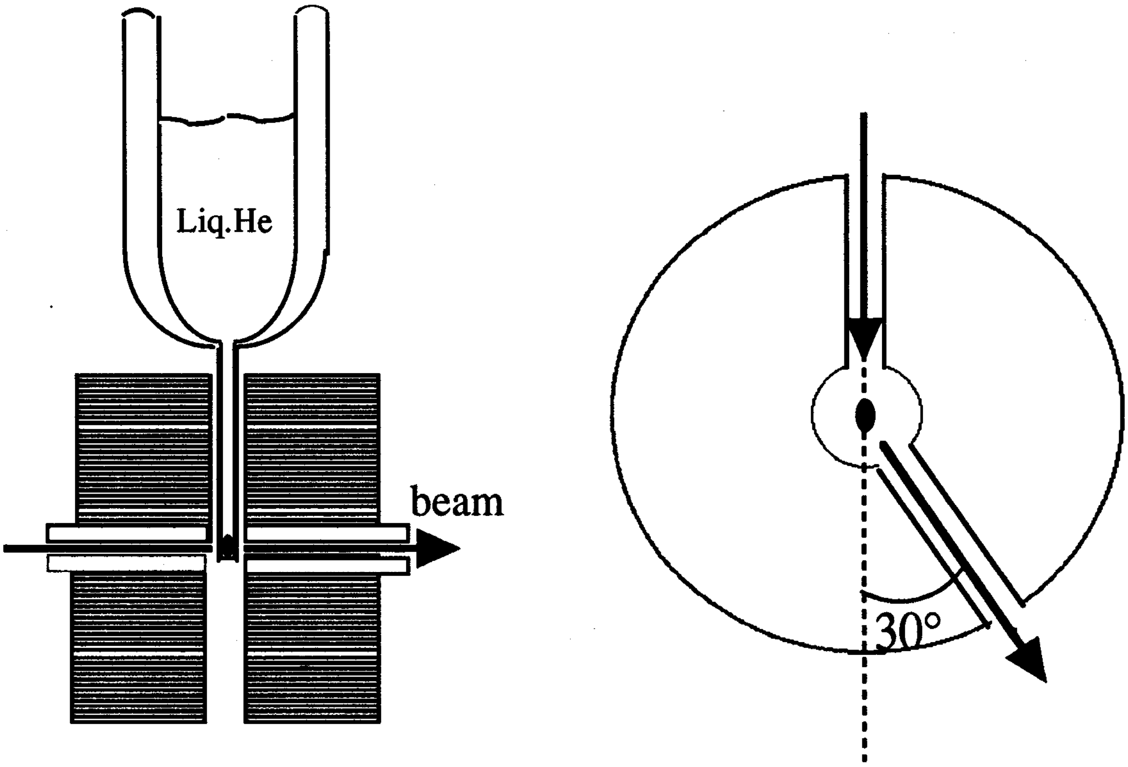


Fig.2 Cut view(left) and top view (right) of the configuration of split type magnet.

4. Design and construction of strong magnet

The magnets described in the above sections are composed of so-called Bitter disks which are copper disks with holes for water flow. The 1 mm thick disks are punched out from a large copper plate. They are stacked with insulator sheets sandwiched between the disks. 100 disks are used for the horizontal configuration magnet coil. The maximum field is limited by the strength of material and structure and by cooling rate as mentioned in the previous papers [1][2]. In our case, the material is copper which is not strong enough to produce a magnetic field higher than 20 Tesla. After 20000 times shots of the pulsed field of 20 Tesla, the inside part of the Bitter disks slightly bent[3]. The structure of the magnet is designed as the disks mechanically contact each other. This may make electrical and mechanical problems after the magnet is used for a long time. Water cooling rate is not a big problem at present. The ion exchanged water is circulated at the rate comparable to the flow rate of filling water to a bathtub. A small problem we have now is the high back pressure of the water circulation system. This problem will be solved soon by using a pump connected to the outlet of the magnet.

In order to obtain higher field than 20 Tesla, it is obvious that we have to use a strong material for the magnet. As the strong material for high field magnet, copper materials reinforced by Beryllium, almina or silver particles are known. Maraging steel is also used. These materials are strong but low in electric conductivity. If maraging steel is used, the produced field is up to 40 Tesla but the electric conductivity is 10 times less than that of normal copper and the energy loss increases 40 times more. So it seems to be difficult to use such a steel for our purpose. The reinforced copper with Beryllium is stronger than the normal copper but brittle and electric conductivity is less than 90 %. So the reinforced coppers with almina or silver seem to be good candidates. From the yielding strength of these materials, 30 Tesla will be available by using the same capacitor bank.

We designed a new magnet using the reinforced coppers with almina and are now machining it at the machine shop in KEK. In this case, the disks mentioned above are not used. We cut a magnet coil from a big bulk material using a electric spark cutter. Suppose a bulk cylinder with a hole of inner diameter and holes for water path similar to the Bitter type magnet disk, a wire electrode is put diagonally perpendicular to the cylinder and the cylinder is rotated slowly with spark machining. After 50 rotations, 100 turns coil will be obtained as a double helix. In this case there is no mechanical connection and this magnet coil must be strong. The disadvantage is that the machining costs expensive.

5. Wire wound magnet

As another way, we are trying a different kind of magnet. When a magnet coil is wound by copper wire and a high field is produced, the force acting on the coil is outward and it is converted to the tangential force. To prevent expansion and destruction of the coil, a strong reinforcement must be used. The usual way to provide external reinforcement is to use a thick stainless steel cylinder and impregnate with epoxy resin in the space between the coil and the stainless steel shell. We suggested a novel idea to use for the reinforcement of the magnet [5]. We used almina powder and water instead of epoxy resin. Once it was frozen at liquid nitrogen temperature, this mixture was like a ceramic and very strong. By this method we have been constructing pulsed field magnets in our laboratory. The magnets usually used are multilayer, so it takes a few minutes to cool down to the initial temperature after field production. We considered if the magnet coil is single layered, the cooling time would be short. After some trials, however, it has turned out that the magnet has been broken below 20 Tesla because the electric current is so strong due to single layer. We are now improving some parts of the magnet to obtain higher than 20 Tesla.

6. Magnetic phase transition of $CsCuCl_3$

$CsCuCl_3$ is one of the hexagonal ABX_3 type compounds. The magnetic properties have been studied many researchers. It has linear chains of Cu^{2+} inos along the c-axis and they make triangular lattices in the c-plane. The intrachain coupling is ferromagnetic, whereas the interchain coupling is antiferromagnetic. Below T_a=423 K, Cl^- octahedron surrounding a Cu^{2+} has a distortion due to the cooperative Jahn-Teller effect, and it causes the antisymmetric Dzyaloshinsky-Moriya interaction between spins along the c-axis. This compound shows 120° magnetic structure in the c-plane and have a

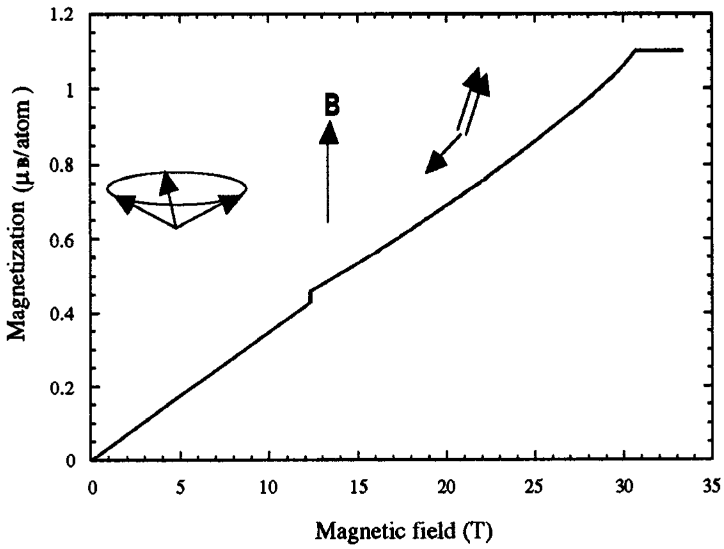


Fig.3 Magnetization curve at 1.1 K for H//c and proposed magnetic structures

long pitch helical spin structure along the c-axis below Tn=10.5K. It has been experimentally found that the magnetization has a small jump at B_c=12.5 Tesla at 1.1K in a field parallel to the c-axis as shown in Fig.3 [6]. It has been difficult to explain this phase transition for a long time within the classical theory. Recently, a new theoretical interpretation for this small jump has been proposed by Shiba and Nikuni [7]. They have pointed out that the quantum fluctuation is important in $CsCuCl_3$ because of S=1/2. They proposed a model that the umbrella-like structure changes to the coplanar structure at B_c as shown in the inserts of Fig.4. In this model, two spins are parallel while one is almost opposite above B_c. This kind of magnetic phase transition have never been observed. We have studied neutron diffraction experiments in a pulsed high magnetic field in order to investigate the magnetic structure of this material.

The magnetic field was applied parallel to the c-axis at the sample temperature of 7K at which B_c =10 Tesla. The intensities of the magnetic reflection (1/3,1/3,0) and (1/3,1/3,0.085) were measured as a function of the applied magnetic field, because a magnetic unit cell is three times as large as a chemical one in the c-plane. First we focused our attention on the (1/3,1/3,0.085) reflection due to the component of the magnetic moments perpendicular to the c-axis. In addition to the 120° umbrella-like structure at low field, this spin system has a helical modulation along the c-axis which makes a satellite along the c*-axis in the reciprocal lattice space. The intensity decrease as a field is increased below B_c as shown in Fig.4 due to the decrease of the perpendicular component of the moments. The calculated intensity is in good agreement with the experimental results. It shows an abrupt decrease at B_c and a gradual change above it. Assuming the model shown in Fig.3, we

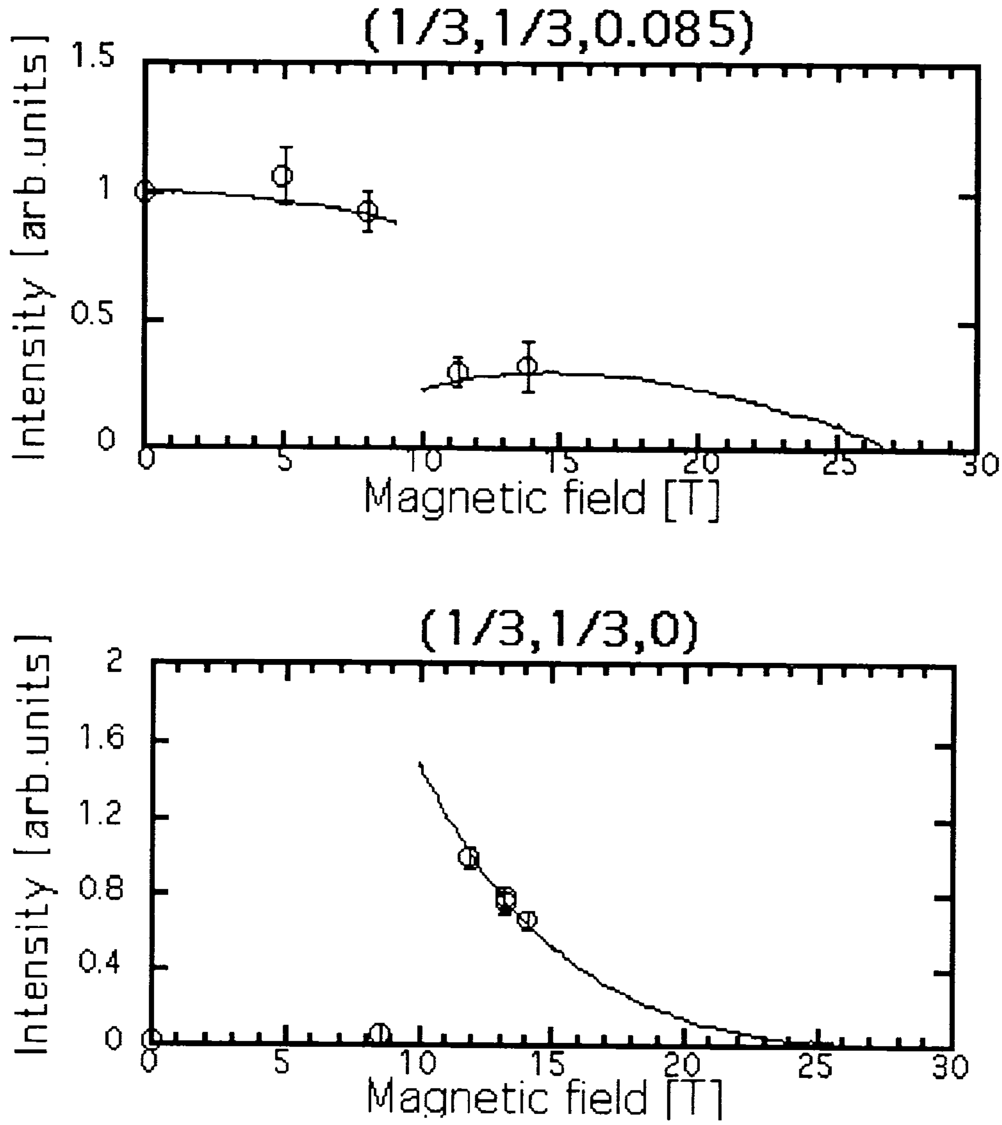


Fig.4 Field dependences of the reflection intensities. solid lines are calculated ones.

calculated the field dependence of the intensity above B_c and found a good coincidence between experiment and calculation as shown by the solid line in Fig.4. The intensity is to become weak as an applied field is close to the saturation field B_s=27 Tesla reflecting decrease of the perpendicular component.

On the other hand, (1/3,1/3,0) reflection is due to the parallel component of the magnetic moments to the c-axis. At the umbrella-like structure below B_c, the c-component of the magnetic moments is identical each other and the reflection must appear at (1,1,0). This reflection overlays on the nuclear reflection and it is difficult to be discriminated from the strong nuclear one. Therefore (1/3,1/3,0) reflection can not be observed below B_c. Above B_c, however, the proposed structure shown in Fig.3 contributes to (1/3,1/3,0) reflection observed at 13.2 Tesla. This magnetic reflection disappears below B_c as shown in Fig.4. We calculated the field dependence of the intensity for the proposed structure and found a very good agreement between calculation and the experimental results as shown in Fig.4.

7. Summary

We have developed pulsed field system for the neutron scattering experiments at high magnetic field. It is practical and will be useful if it is used with a stronger beam intensity. We look forward to the new project in Japan and also our technique will be useful even at ISIS.

At present time, the research for high Tc superconductors are intensively progressing and the practical use of them will come in future. If superconducting magnet which is capable of producing higher fields than 20 Tesla at room temperature or even liquid nitrogen temperature may appear. The advantage of our technique will disappear.

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