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# Neutron diffraction study in pulsed high magnetic field

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## ABSTRACT
We have developed a technique for neutron diffraction experiments in pulsed high magnetic fields to determine magnetic structures in high field. Our system is capable to produce pulsed fields up to 20 T every 2 sec. The experiment was done using MRP diffractmeter at KENS. The magnet was the so-called Bitter type water cooling solenoid with 40 mm inner diameter, 240 mm outer diameter and 220 mm length. It was horizontally installed on the goniometer. The flow type FRP cryostat was set in the magnet and the temperature was down to almost 4.2 K but there was a heating problem for metal sample when a pulsed magnetic field was applied. The first application has been done for a determination of high field magnetic structure of metamagnetic $PrCo_2Si_2$.

## I. INTRODUCTION
As is well known, many magnetic materials show magnetic phase transitions in high magnetic field and it is important to investigate magnetic structures in such high field phases by neutron diffraction. However, the magnetic field available in neutron diffraction experiments has been limited below 10 T so far even a superconducting magnet is used. The discussion in this paper is on the method to do neutron diffraction experiments in magnetic fields above 10 T.

There are several ways to produce high magnetic field, and they are classified usually to two categories. One is DC field and another is pulsed field. DC field can be produced by using conventional electromagnet, superconducting magnet, water cooling Bitter type magnet and hybrid magnet. The electromagnet which produces only 2 or 3 T is out of question for our purpose. The available field by using superconducting magnet is now up to 20 T but when used for neutron diffraction experiment, it may be limited to 10 T due to complicated configuration of equipments. The water cooling Bitter type magnet can provide 20 T and the hybrid magnet which is the combination of Bitter type magnet and large superconducting magnet is capable to generate 30 T[1]. The electric power necessary for Bitter type magnet[2] is calculated by

$$H = G\sqrt{\frac{W_{DC}}{\rho}}, \qquad (1)$$

where H is the produced field, G geometrical factor of the magnet, $W_{DC}$ electric power, $\rho$ resistivity of the coil material. $W_{DC}$ is roughly estimated to be a few MW for 20 T in a space of about 100 $cm^3$. This power is converted to heat and then cooling system for the magnet is also huge. Then we consider it is almost impossible to use them for neutron diffraction experiments.

On the other hand, techniques for producing pulsed high magnetic field and its application to physics have been well developed in this decade. We do not discuss the destructive methods which can produce fields above 100 T[3] because it is considered to be impossible to combine with neutron diffraction experiments. In the non-destructive way, it is possible to obtain 80 T[4] by a method developed in Osaka University. But it is not so

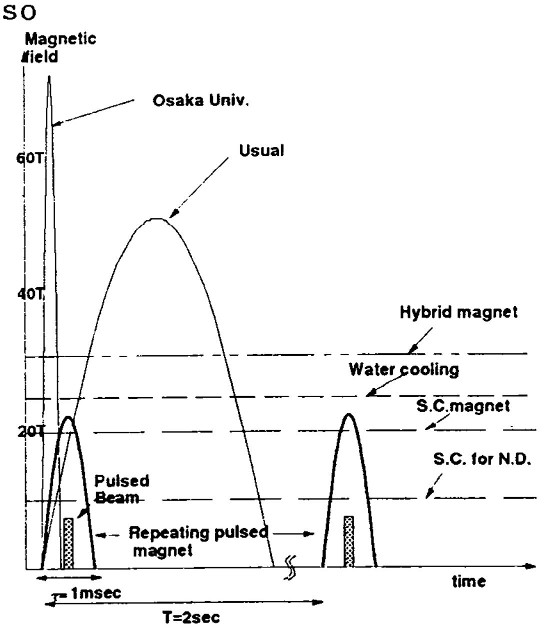
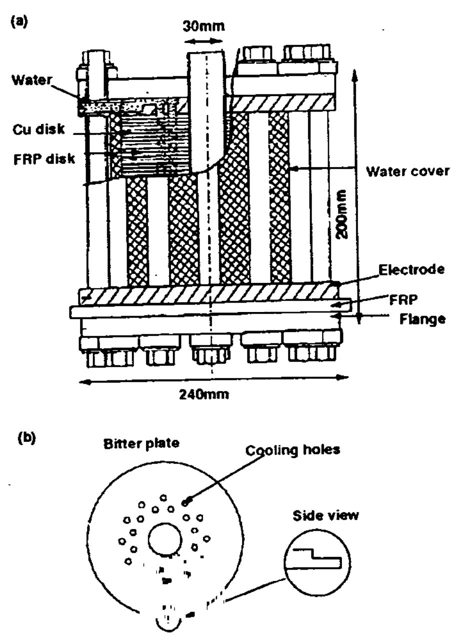



Fig.1 Methos to produce high magnetic and time duration




Fg.2 (a) Cut view of the magnet and (b) top view of a disk compact to be installed in neutron facility. The most conventional and popular way is to use a magnet wound by copper wire and cooled by liquid nitrogen. This method is capable to produce fields up to 40 T and is widely used. The maximum fields obtained by these methods are summarized in Fig.1 with time duration of pulsed field. Grössinger *et al*[5] have tried to use single shot pulsed field with a reactor operated in a flushed manner, but the beam intensity was not strong enough to detect the diffraction.

We have considered that repeating pulsed fields synchronized with the neutron beam pulses will be useful for high field neutron diffraction experiments and have developed the technique for it[6].

## II. REPEATING PULSED FIELD

The repeating pulsed field is defined as a pulsed field which is produced cyclically with an interval T as is shown in Fig.1. What we have to consider to design the repeating pulsed field are the followings:

1) Type of magnet.
2) Field shape as a function of time
3) Pulse width
4) Interval of pulsed field
5) Available maximum field
6) Configuration of installation

1) Type of magnet

Repeating pulsed field is the intermediate between a DC field and a single shot pulsed field. The heat is integrated by every shot and then the magnet must be cooled. The easiest way for cooling is to use a Bitter type magnet. We made disks with thickness of 1 mm and a shape shown in Fig.2(b) and stacked around 100 disks with insulator disks. The cut view of the magnet is shown in Fig.2(a).

2) Field shape

We use capacitor discharge method as is discussed in reference 6, then the field shape is inevitably half-sin curve. For neutron diffraction experiments, however, rectangular shape is favorable. It is possible using a complicated circuit if a budget is enough but difficult by using a simple circuit like ours. We are planning to make a flat-top field as is shown in Fig.3 in an economical way.

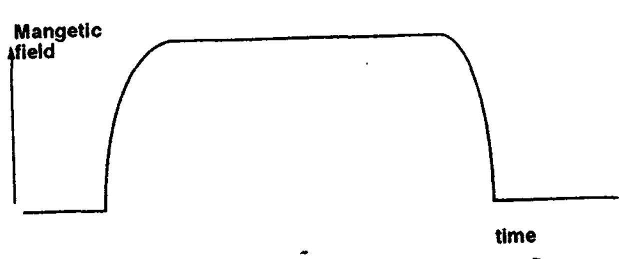



Fig.3 Shape of flat-top field

3) Pulse width

This is determined by the inductance L of the magnet and the capacitance C of the capacitor bank as $\tau = \sqrt{LC}$. When we use Bitter type magnet, L is determined by the volume of the field space and is of the order of 100 $\mu$H. From the field intensity and the volume of the field space, the energy of the capacitor bank is given and from the limitation of available charging voltage, C is estimated to be of the order of mF. Then $\tau$ is of the order of msec. In our case, the energy of capacitor bank is 60 kJ with C=1.2 mF and $\tau$ is 1 msec.

4) Interval of pulsed field

This is determined by charging ability of the power supply and ability of the cooling system. The cooling rate is reduced by the relation

$$W_{RP} = (\tau/2T)\cdot W_{DC}, \qquad (2)$$

for half-sin shape, where $W_{RP}$ is the electric power consumed by repeating pulsed field system. Both charging ability and cooling ability partly depend on budget but T~0.1 sec may be a limit bcause the system must be compact and easy for treatment. In our case T=2 sec as is discussed in reference 6.

5) Available maximum field

This is limited by the strength of the magnet material. By normal copper, we obtained fields up to 20 T. If we use reinforced copper, 30 T may be available. For one shot pulsed field, maraging steel is used in Osaka University[4]. It has a strength capable to produce fields up to 40 T. When it is used for repeating pulsed field, $W_{DC}$ is estimated from Eq.(1) to be about 40 times larger than that of copper case due to almost 10 times larger resistivity. At present time, we can produce 20 T field by using copper disk magnet.

6) Configuration of installation

As will be discussed later, we now use a magnet whose field direction is almost parallel to the neutron beam direction. Sample is set at the center of the magnet and liquid He flow type cryostat is used. In this configuration, however, we consume too much liquid He and also the available scattering angle is small due to small diameter of the magnet. We are now planning to build a split type magnet with vertical field. This will enable us to use vertical cryostat and to have large scattering angle. But the maximum field in the gap will be reduced to about 75 % of that of solenoid.

# III. HIGH FIELD NEUTRON DIFFRACTION OF $PrCo_2Si_2$

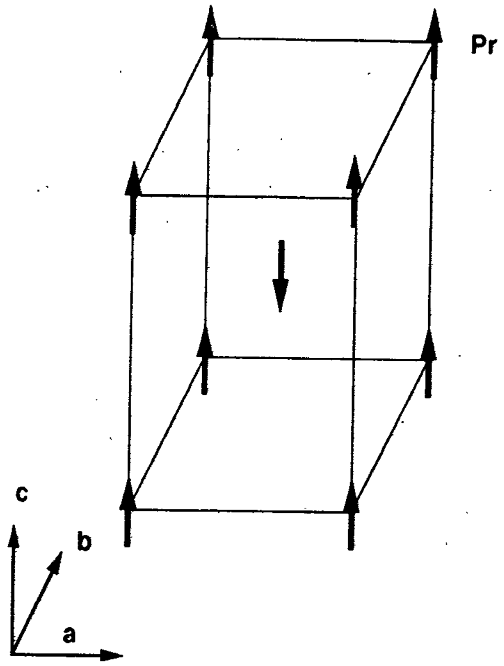



Fig.4 Magnetic structure of $PrCo_2Si_2$

Only Pr has magnetic moment in this crystal and it aligns ferromagnetically in the c-plane while antiferromagnetic coupling with the adjacent layer below $T_1$=9 K as is shown in Fig.4. Between $T_1$ and $T_2$=17 K, it turns to a long pitch incommensurate structure with a propagation vector $k$=0.926 and has another structure with $k$=0.777 between $T_2$ and $T_N$=30 K, as are shown in Figs.5 and 6[7]. On the other hand, when an external field is applied along the c-axis at 4.2 K, it shows metamagnetic transitions at 1.2, 3.8 and 12.2 T[8] due to Ising like strong anisotropy. From the field induced

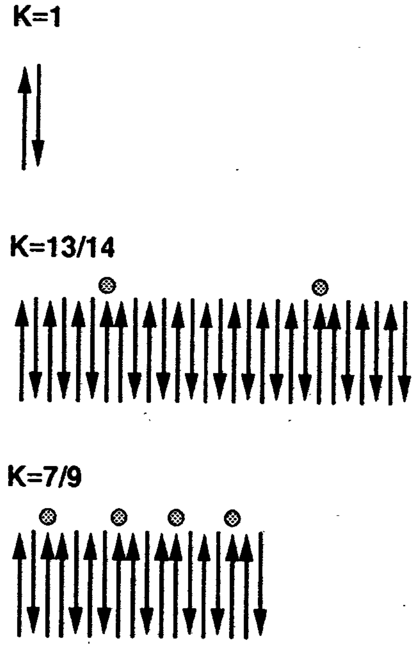



Fig.5 Magnetic alignments of antiferromagnetic and incommensurate states

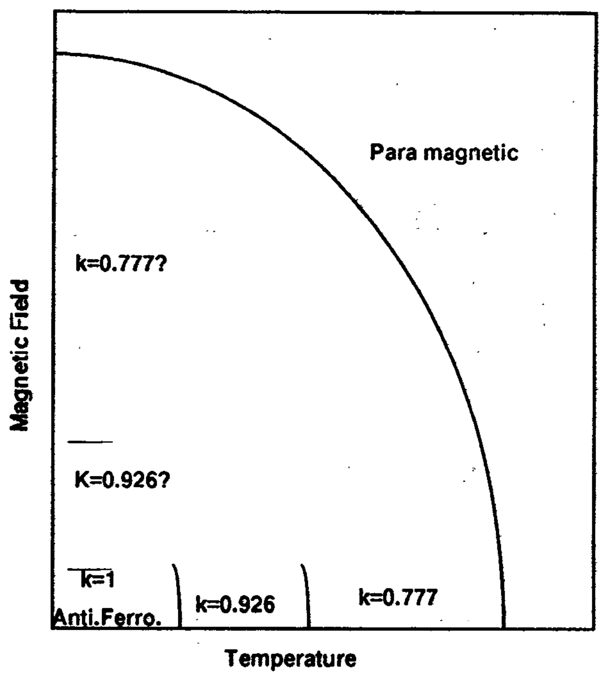



Fig.6 Inferred phase diagram

moment, the magnetic structures in these states are inferred to be same as those appears in the intermediate temperatures. The results are schematically summarized in Fig.6. Our present purpose is to confirm these structures by neutron diffraction.

The sample size was 6mm in diameter and 7 mm thickness and the magnetic field was parallel to the c-axis. The beam size was about 15 × 10 $mm^2$. The diffraction angle was kept at 2θ=10° and the horizontal rotation of the sample was limited within a few degrees because of small aperture of the magnet. This configuration was enough to scan the area of reciprocal space concerning the expected propagation vectors. The wave length of used neutron was about 1 Å. Since the metallic materials cannot be used for both cryostat and sample holder due to eddy current heating, we used a FRP cryostat and a BN ceramics sample holder. The magnet and cryostat was set up on a goniometer which adjusted the angle between the incident neutron beam and the c-axis of the sample. The experimental set up is shown in Fig.7.

First we observed diffraction changing temperature at zero field to adjust the configuration of equipment and to check the sensitivity and accuracy of the system. Next we applied external fields up to 16 T at 4.2 K and obtained the expected Bragg reflection corresponding to the complicated magnetic structures. The experiment was done by time of flight method. The wave length $\lambda$ of neutron beam is proportional to time after spallation. Fig.8

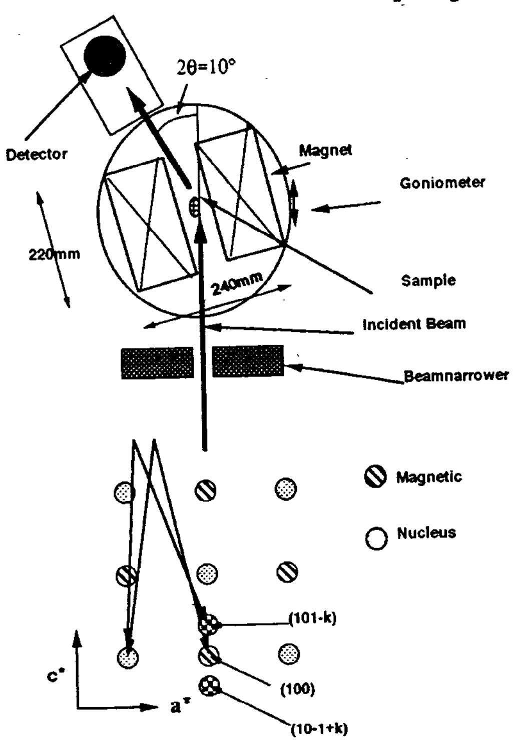



Fig.7 Experimental set up for high field neutron diffraction experiment

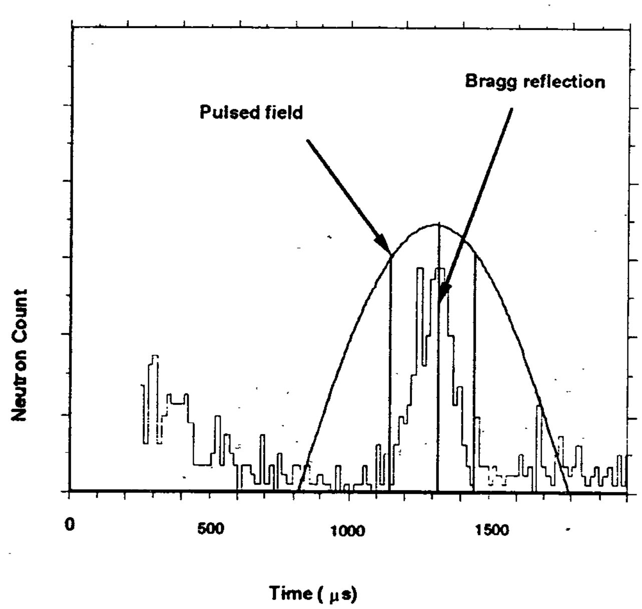



Fig.8 Synchronized pulsed field to neutron beam of relevant wave length

shows the time relation between the neutron beam with relevant $\lambda$ and the generated pulsed field. To observe a Bragg reflection at high field, the field pulse was synchronized so as to coincide with the neutron beam of desired $\lambda$ at the field peak. The observed Bragg reflection had a width due to resolution of detector and mosaic structure of the specimen and the resultant width was about 200 msec in time spectrum. Because of half-sin shape of the field, field change during this time was about 10%. But this was not a serious problem in this case because the structure was supposed to be same in a phase when an applied field was adjusted at the intermediate of the transition fields. The interval of the pulsed field was 2.5 sec because of the accelerator operation program and then it took 40 times longer time than the ordinary experiment. However the intensity of the Bragg reflection strong enough to take data in an hour or two. We scanned mainly around (1,0,0) magnetic peak and observed (1,0,1-$k$) satellite peak arising from the incommensurate structure with propagation vector $k$. The scanning was done by changing an angle of the incident neutron beam to the c-axis of the sample. The value of $k$ was determined by the change of incidence angle of neutron beam. Heating of sample by eddy current occurred during field generation, then the temperature was higher than 4.2 K when the Bragg reflection observed in high field. The real temperature of the specimen in a pulsed field was estimated by the following way. We applied a pulsed field just before the time corresponding to the Bragg reflection of antiferromagnetic state, and measured the reflection intensity at zero field, which reflected the temperature just after the field application. As the relation between temperature and intensity of Bragg reflection had been obtained by Shigeoka *et al.*[7], we calibrated temperature using it.

An example of the obtained data is shown in Fig.9, in which the Bragg reflection by (1,0,0.777) corresponding to the incommensurate state is seen at 11.5 T but it disappears at 14 T. All the data obtained by our experiments are summarized in Fig.10 as a phase diagram. The phases determined by the present experiment is consistent with the expected ones from the magnetization measurement.

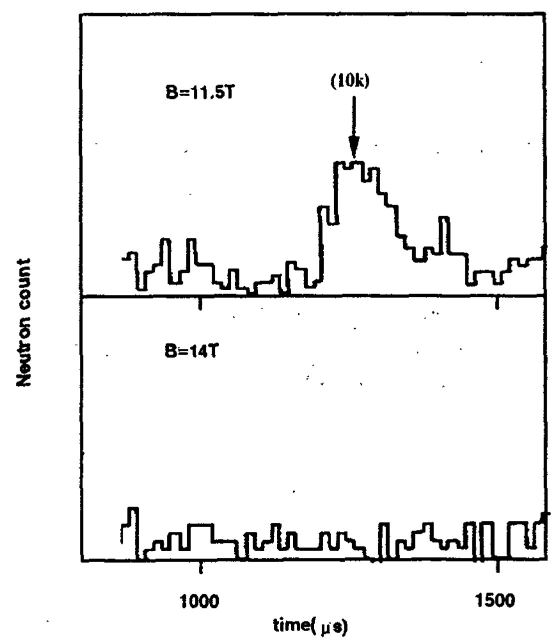



Fig.9 Bragg reflection at 11.5 T which disappears at 14 T.

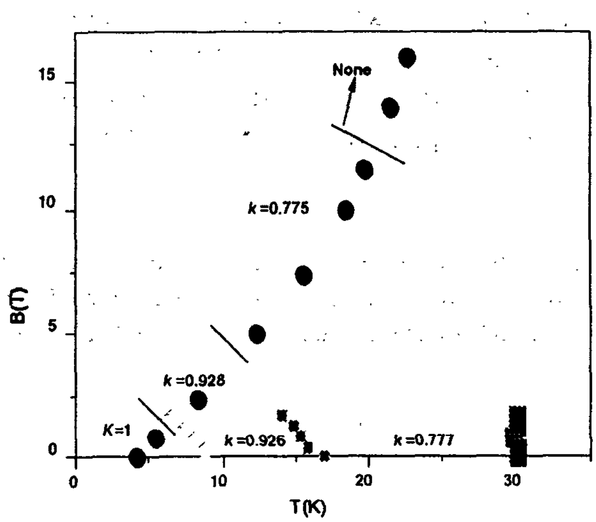



Fig.10 Confirmed phase diagram
● is observed point

## IV. CONCLUSION

We have succeeded in neutron diffraction experiments in a high magnetic field up to 16 T and shown the repeating pulsed magnetic field is quite useful for pulse neutron experiments.


## ACKNOWLEDGMENT

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Q(R.Pynn): Can one look at time-dependent phenomena after a magnetic pulse has been applied?

A(M.Motokawa): Yes, It is easy to look at time-dependent phenomena after a field pulse. One can see the time dependence of diffraction after an pulsed field applied just before the relevant $\lambda$(or k).