Neutron diffraction experiments with 40T pulsed magnets

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Abstract. Aiming at realising neutron scattering experiments under $B=40$ T magnetic fields, we are developing diffusive techniques for neutron diffraction with a long pulse magnet. For the present experiments, we succeeded in observing the spin-flop transition of the antiferromagnet MnF$_2$ around $B=10$ T using a 20 T pulsed magnet on a neutron spectrometer installed at a reactor. 35 T pulsed magnetic fields were also successfully generated.

1. Introduction
Magnetic field is a unique and important parameter which can control magnetic moments in materials directly and precisely. Particularly, high magnetic field experiments are indispensable to study strongly correlated electrons systems, in which high magnetic fields cause novel and exotic magnetic behaviour. On the other hand, neutron scattering provides a direct probe of space-time correlation of spins in materials. Thus, neutron scattering under high magnetic fields is eagerly anticipated in novel material science.

Currently, the highest magnetic field for neutron scattering experiments was $B=25$ T, which was performed by Motokawa et al. using a repeating pulsed magnet in Neutron Science Laboratory of High Energy Accelerator Research Organization in Japan [1, 2]. However, neutron scattering experiments over $B=10$ T are still difficult and limited for the present; therefore, easier and more diffusive techniques are ambition. Therefore, aiming at performing neutron scattering experiments under $B=40$ T, we are developing easier and more diffusive techniques for neutron scattering with long pulsed magnets.

2. Experimental
We performed pulsed magnetic field experiments on the triple axis spectrometer, AKANE, of Institute for Materials Research, Tohoku Univ. installed at the reactor JRR-3M in Japan Atomic Energy Agency in Tokai, Japan. The sample of the experiments was MnF$_2$: MnF$_2$ is a simple antiferromagnet ($T_N=68$ K), which shows a spin-flop transition at $B=9.27$ T for $B//c$-axis [3].

Figure 1 shows the circuit diagram of the double-coils magnet system with a choke coil, which make the dumping of the magnetic field slower. The parameters of the system are shown in the figure caption. For the single-coil mode, the choke coil was removed. Figure 2 shows the
calibration curve between the coil current and magnetic field generated by the single-coil mode determined by off-beam measurements with a pick-up coil. We have confirmed that a magnetic field of 35.2 T could be generated stably by the system. Figure 3 shows the pulse shapes of the magnetic field generated by the single- and double-coil modes; for the double-coil mode, the width of half maximum was $\sim 4.5$ msec and the width of the bottom was $\sim 15$ msec, which are $\sim 15$ times longer than those for the single-coil mode. Since the space in which magnetic field is generated is quite small ($\phi 5 \text{mm} \times 10 \text{mm}$), the total energy can be reduced: 3300 J and 80 J for the double coil and the single coil modes, respectively [4].

**Figure 1.** Circuit diagram of the double-coils pulsed magnet system. C, THR, D indicate condenser, thyristor, and diode, respectively. The choke coil was cooled by liquid-N$_2$ during the experiments. For the single-coil magnet mode, the choke coil was removed. The parameters of the circuit diagram are as follows: $L_1 \sim 0.1 \mu\text{H}$, $L_2 \sim 1 \mu\text{H}$, $C = 3.36 \text{ mF}$ for the double coil mode, $C = 0.96 \text{ mF}$ for the single coil mode.

**Figure 2.** Calibration curve between the coil current and generated magnetic field of the single-coil magnet. The highest magnetic field in this measurements was $B = 35.2$ T

**Figure 3.** Comparison of the pulse shape of magnetic fields generated with the single-coil (dashed line) and double coil (solid line) pulsed magnets. $E_{\text{total}}$ is the total energy consumed in the choke and magnet coils in this system. The interval of each pulse was $\sim 7$ min for the single-coil magnet and $\sim 17$ min for the double-coils magnet.

For the neutron scattering experiments, we used the double-coil mode to make a long pulse; the maximum magnetic field was set to $B = 20$ T for the safety. The magnet coil, in which the single crystalline sample of MnF$_2$ ($\phi 4.7 \text{mm} \times 9 \text{mm}$) was fixed, was set in a liquid He refrigerator so that the coil and the sample were cooled down to $T = 4.2$ K through exchange He gas; the exchange gas maintained homogeneity of temperature of the coil and the sample as well. The magnetic field in the sample space was estimated from the coil current through the calibration curve obtained by off-beam measurements.
The scattering plane was the $a^*-c^*$ plane so that we could measure the (100) antiferromagnetic reflection under magnetic fields applied along the $c$-axis. For the experiments, the wave length of the neutrons was $\lambda=2.0\,\text{Å}$; the collimation condition was Guide tube-Open-Sample-Blank-Blank, which gives highest scattering intensity in AKANE. The single 1D neutron detector of AKANE was fixed at the peak top position of the (100) antiferromagnetic reflection. During the pulsed magnetic fields experiments, neutrons counts and coil currents were measured with an oscilloscope (channel width: $4\,\mu\text{sec}$) to resolve time structure so that magnetic field dependence of neutron counts could be obtained.

3. Results

Figure 4 indicates the peak profile of the (100) magnetic reflection of MnF$_2$ which was set in the magnet coil at $T=4.2\,\text{K}$ under $B=0\,\text{T}$. Note that the background level was $\sim 1/150$ or less of the peak top intensity even though the sample was much smaller than the coil; therefore, the contamination of the background can be neglected for the experiments under pulsed magnetic fields.

![Figure 4](image1)

**Figure 4.** (100) magnetic reflection of MnF$_2$, which was set in the magnet, under $B=0\,\text{T}$ at $T=4.2\,\text{K}$ obtained on AKANE. The background level was $\sim 1/150$ or less of the peak top intensity. During the high field experiments, the detector was fixed at the peak top position of the (100) reflection.

![Figure 5](image2)

**Figure 5.** Time Dependence of neutron counts at the peak top position of (100) magnetic reflection at $T\sim 4.2\,\text{K}$ obtained after 100 shots (upper panel). The background level was estimated as $\sim 1$ counts for this condition. Time dependence of magnetic field was also indicated as the solid line in the lower panel. The dashed lines indicate the boundaries of the region where $B \geq 10\,\text{T}$ were applied.
Figure 5 shows time dependence of neutron counts at the peak top obtained after 100 shots. It needed \( \sim 15 \text{ min.} \) to cool down the coil and the sample enough, and \( \sim 2 \text{ min.} \) to charge the condenser bank, so that the interval between each pulse of \( \sim 17 \text{ min.} \) was needed. Consequently, the actual measurements duration was about 28 hr. Since the background was negligible small under the present conditions as shown in figure 4, the scattering in figure 5 was due to the \((100)\) magnetic reflection. As shown in figure 5, the intensity of the \((100)\) magnetic reflection decreased when magnetic fields over 10 T were applied. Note that the intensity recovered when the magnetic field decreased and became lower than \( B=10 \text{ T} \), indicating that the decrease of the intensity was not due to the heating by the coil current. Thus, figure 5 indicates that we succeeded in observing the spin-flop transition of MnF\(_2\) at \( B=10 \text{ T} \). This is the first neutron diffraction data obtained under \( B=20 \text{ T} \) using spectrometers in reactor facilities in Japan.

Based on the present results, we are improving the magnet system to obtain higher magnetic fields: reducing the electric resistivity of the choke and magnet coils to minimize the energy loss, and increasing the electric capacitance of the condenser bank \((C \sim 7 \text{ mF})\). We expect that these improvements make possible to realize neutron diffraction experiments under \( B=40 \text{ T} \) pulsed magnetic fields.

On the other hand, there exists some problems to be solved; though it was expected that the intensity above \( B=10 \text{ T} \) is a half of that under \( B=0 \text{ T} \) or less, assuming isotropic distribution of magnetic domains in the spin-flop state, the change in the present experiments was only \( \sim 25 \% \). Though the reason is not clear at the moment, there exists some possibilities: (1) inhomogeneity of generated magnetic fields because the sample length was nearly the same as that of the coil, (2) lopsided domain distribution probably caused by misalignment between the c-axis and magnetic field, (3) the background under pulsed magnetic fields becomes unexpectedly higher. To enhance the homogeneity of the magnetic field, the sample length should be shorter than the coil length. To check the contamination of the background under magnetic field, observation of the peak profiles is important, which could not be observed in the present experiments.

4. **New devices to enhance beam flux and efficiency.**

Though we succeeded in observing the spin-flop transition of MnF\(_2\), enhancement of neutron beam flux is seriously required for practical investigation of novel materials because (1) the sample size will become smaller to enhance homogeneity of the generated magnetic field, (2) the measuring duration is extremely short, (3) beamtimes of neutron scattering experiments are limited. Therefore, to enhance the effective counting rate additionally, we are developing new devices: neutron focusing device and position sensitive neutron detector (PSD) system. The neutron focusing device consists of neutron suppermirrors, which can reflect neutron beam with high reflectivity, so that the beam density at the sample position can be enhanced. The PSD system can measure in wide \( 2\theta \) range simultaneously without step scans; therefore, using the PSD system, we will be able to obtain peak profiles under pulsed magnetic fields. Using these new devices, neutron diffraction experiments under \( B=40 \text{ T} \) are in progress.

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