

Stroboscopic Time-of-Flight Neutron Diffraction in Long Pulsed Magnetic Fields

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We present proof-of-principle experiments of stroboscopic time-of-flight (TOF) neutron diffraction in long pulsed magnetic fields. By utilizing electric double-layer capacitors, we developed a long pulsed magnet for neutron diffraction measurements, which generates pulsed magnetic fields with the full widths at the half maximum of more than 10^3 ms. The field variation is slow enough to be approximated as a steady field within the time scale of a polychromatic neutron pulse passing through a sample placed in a distance of the order of 10^1 m from the neutron source. This enables us to efficiently explore the reciprocal space using a wide range of neutron wavelength in high magnetic fields. We applied this technique to investigate field-induced magnetic phases in the triangular lattice antiferromagnets $\text{CuFe}_{1-x}\text{Ga}_x\text{O}_2$ ($x = 0, 0.035$).

I. INTRODUCTION

Exploring new quantum states of matter in extreme conditions, such as low temperatures, high magnetic fields, and high pressures, is one of the central topics in condensed matter physics. Among them, novel field-induced phases have been extensively investigated; for example, spin-lattice-coupled magnetization plateaus in frustrated spin systems [1–4], field-induced flop of the spin-driven electric polarization in multiferroics [5, 6], spin-nematic states in quantum spin systems [7, 8] etc. Neutron scattering is one of the most powerful techniques to study these exotic phenomena because it can probe Fourier-transformed time-space correlation functions of atomic positions and magnetic moments; specifically crystal and magnetic structures are determined by the elastic scattering, and phonons and magnons are measured by the inelastic scatterings. However, the highest magnetic field for neutron scattering instruments is limited to approximately 15 T even for state-of-the-art superconducting magnets. Although there was a superconducting and non-superconducting hybrid magnet for neutron scattering with the highest magnetic field of 26 T in the Helmholtz Zentrum Berlin (HZB)[9], it is no longer available since the research reactor in HZB was shut down in 2019.

An alternative is a pulsed magnetic field[10–13]. For instance, Nofiri and co-workers successfully observed a magnetic Bragg peak in a field-induced phase of multiferroic MnWO_4 by time-of-flight (TOF) pulsed neutron diffraction measurements with pulsed magnetic fields up to 30 T[11]. However, the widths of the pulsed magnetic fields are often limited to several milliseconds, which

are shorter than a time spread of a polychromatic neutron pulse after flying a typical source-to-sample distance in existing TOF neutron diffractometers, as we explain later. In this case, the high magnetic field is achieved only for a limited wavelength range of the neutrons, and this situation makes explorations of new magnetic peaks in the reciprocal space difficult.

To overcome this problem, in the present study, we have developed a long pulsed magnet for neutron scattering experiments. By utilizing electric double-layer capacitors (EDLC)[14], we successfully generated pulsed magnetic fields whose full widths at the half maximum exceed 100 ms. We established the low-temperature neutron scattering environment with this long pulsed magnet, and performed stroboscopic TOF neutron diffraction measurements on the triangular lattice antiferromagnets $\text{CuFe}_{1-x}\text{Ga}_x\text{O}_2$ ($x = 0, 0.035$). By virtue of the extended time scale of the magnetic field, we can use a wide wavelength range of the incident neutron beams, which enables us to map out the neutron diffraction intensities in the reciprocal space in high magnetic fields.

CuFeO_2 has been extensively investigated as a geometrically frustrated system [15, 16]. Crystal structure of this compound belongs to the space group of $R\bar{3}m$, and consists of triangular lattice layers of magnetic Fe^{3+} ions, which are rhombohedrally stacked along the c axis. In zero magnetic field, this compound exhibits a collinear four-sublattice (4SL) antiferromagnetic order, in which the spins are arranged to form an up-up-down-down sequence running along the $[110]$ direction, below 11 K. The spins in the adjacent layers are antiferromagnetically aligned, and thus the magnetic modulation wave vector (q -vector) is described as $q = (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$. The 4SL antiferromagnetic order breaks the threefold rotational symmetry about the c axis of the crystal, and thus results in three magnetic domains corresponding to the q -vectors of $q = (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$, $(-\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ and $(\frac{1}{2}, -\frac{1}{2}, \frac{1}{2})$, which are inter-

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