Experimental report

12/02/2025

Proposal: **5-41-1243** **Council:** 10/2023

Title: Magnetic structure elucidation of CaCu3Ti4O12 using neutron scattering

Research area: Physics

This proposal is a new proposal

Main proposer: **Yung Hsiang TUNG**

Experimental team: Po Chun CHANG
Yung Hsiang TUNG
Sabreen HAMMOUDA
Yixi SU

Local contacts: Iurii KIBALIN
Bachir OULADDIAF
Eric RESSOUCHE

Samples: $CaCu_3Ti_4O_{12}$
Fe_3GaTe_2

Instrument	Requested days	Allocated days	From	To
ORIENTEXPRESS	1	1	12/04/2024	13/04/2024
D23	7	0		
D10	7	6	09/04/2024	15/04/2024

Abstract:

The perovskite-like compound, $CaCu_3Ti_4O_{12}$, with its unique dielectric properties, has sparked scientific curiosity due to its divergence from conventional high dielectric constants in ferroelectric systems. Recent studies have primarily aimed to understand the mechanisms underpinning these properties, with specific focus on its magnetism and the role of anisotropy in determining its magnetic structure. Contradictory assertions concerning its magnetic structure, along with discrepancies in single crystal measurements and theoretical predictions, underscore the necessity for further detailed investigations. This proposal endeavors to comprehensively elucidate the compound's magnetic structure using the D10 four-circle diffractometer, and to reveal the temperature dependence of the specific magnetic reflections. The findings aim to complement another inelastic scattering experiment, merging insights on magnetic structure with spin wave dynamics to provide a holistic understanding of the material's magnetic interactions.

1. Brief Description of the Experiment:

Neutron diffraction experiments were performed at the D10 diffractometer using thermal neutrons with an incident wavelength of 1.26 Å. A Cu monochromator was chosen to ensure high momentum resolution while minimizing contamination from higher-order reflections and reducing absorption.

The measurements were conducted at three different temperature conditions:

- 50 K (above T_N): To investigate the nuclear structure in the paramagnetic state.
- 2 K (well below T_N): For detailed determination of the magnetic structure.
- Temperature range 2 K–30 K: To track temperature-dependent changes in magnetic reflections.

2. Main Results:

The experiment was designed to measure both nuclear and magnetic reflections independently. A total of 273 (26 after the reduction process) nuclear and 97 valid magnetic Bragg reflections (after integration checking) were collected, allowing for precise refinement of the nuclear and magnetic structure.

3.1 Nuclear and Magnetic Structure

At 50 K, the system is in the paramagnetic state, and only nuclear reflections were observed. The nuclear structure was refined using 26 nuclear reflections, yielding the lattice parameter: a = 7.361 and α = 120 and Goodness factors: R^2 = 8.75, R^2_w = 4.52. The refined atomic positions and occupancy factors are consistent with the cubic ***I m -3*** symmetry. At 2 K, the antiferromagnetic structure was confirmed, with a propagation vector (1,0,0). The magnetic moment of Cu^{2+} ions was determined to be 0.796(3) μ_B per Cu, with the following components: α = 0.562, β = 0.417, and γ = 0.714. The goodness-of-fit factors were R^2 = 2.15, R^2_w = 2.22, confirming the accuracy of the refined magnetic model. There was no significant structural distortion observed between 2 K and 50 K, confirming that the magnetic transition does not induce a structural phase transition.

3.2 Temperature-Dependent Neutron Diffraction (2 K–30 K)

The intensity of the magnetic reflections (110), (111) and (100) was monitored as a function of temperature. The magnetic peak intensity decreased with increasing temperature and vanished at T_N = 26 K for reflections (111) and (100), while the reflection (110) remains unchanged, confirming the antiferromagnetic transition. The squared magnetic structure factor $|F|^2$ followed a power law near T_N yields the critical exponent β was 0.307(13) for (111) and 0.319(17) for (100), which match the three-dimensional Ising model ($\beta \approx 0.326$), indicating strongly anisotropic spin interactions.

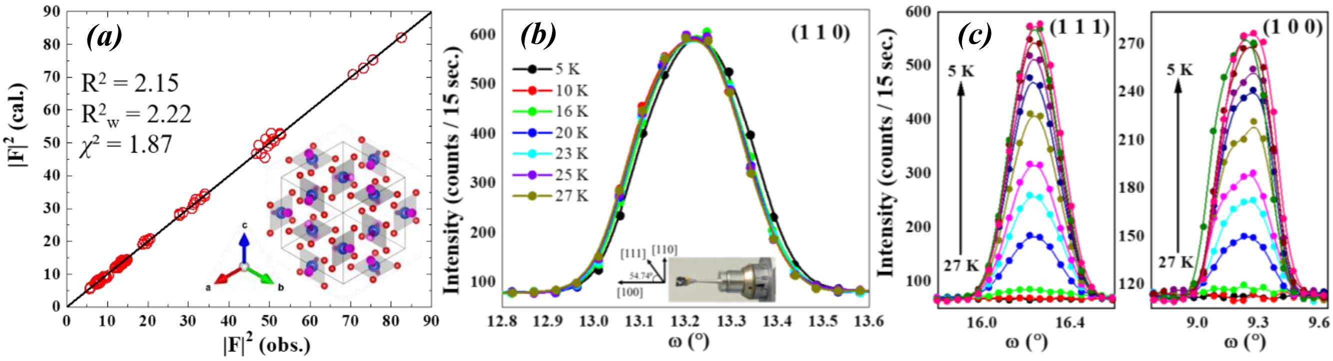

FIG. 1: (a) Refinement results for magnetic reflections at 2 K. The inset depicts the resolved magnetic structure oriented along the [111] crystallographic direction. (b)(c) Temperature-dependent square of the structure factor and its corresponding rocking curves for the reflections (110), (111), and (100).

3. Further Comments and Outlook:

- The neutron diffraction data confirm that $CaCu_3Ti_4O_{12}$ remains structurally stable across the magnetic transition.
- The temperature-dependent behavior follows a second-order phase transition with a three-dimensional Ising-like spin ordering.
- Future work should explore:
    1) Neutron diffraction under an applied magnetic field to investigate potential metamagnetic behavior.
    2) Further inelastic neutron scattering (INS) studies to track spin dynamics across the transition.