Experimental report

17/03/2021

Proposal: **5-11-440**

Council: 4/2020

Title: The nuclear structure of CVT grownFe3-xGeTe2

Research area: Physics

This proposal is a new proposal

Main proposer: **Chris STOCK**

Experimental team: Navid QURESHI
Edmond CHAN

Local contacts: Oscar Ramon FABELO ROSA
Nebil AYAPE KATCHO

Samples: Fe3GeTe2

Instrument	Requested days	Allocated days	From	To
D9	4	5	17/02/2021	22/02/2021

Abstract:

We have been investigating the magnetic properties of two dimensional honeycomb magnets. The bulk properties in these materials is highly sensitive to the stacking of the two dimensional sheets and hence the crystal structure. We wish to characterize the crystallographic and magnetic structure of our crystals of Fe3GeTe2 grown using chemical vapor transport. Inelastic measurements have found different magnetic dispersion relations in these crystals from those reported and grown using the flux technique. We will establish the stacking of the 2d sheets in our crystals as well as the details of iron concentration. The experiment requires the use of D9.

Proposal 5-11-440, 5 days on D9
The nuclear structure of CVT grown $Fe_{3-x}GeTe_2$

Introduction

Magnetic properties two-dimensional magnets are highly related to the stacking of the two-dimensional layers. Ferromagnetic $Fe_{3-x}GeTe_2$ crystallizes in $P6_3/mmc$ space-group, and its structural and magnetic properties are highly linked to the iron concentration [1]. The single-crystal studied in this D9 experiment was synthesized using chemical vapor transport, and inelastic measurements from MACS, NIST, pointed out different dynamics from samples grown by flux [2]. The aim of this D9 experiment is to characterize the nuclear and magnetic structure of this crystal and compare it to the literature.

We have collected nuclear reflections (including ferromagnetic contributions below T_C) at four different temperatures: 300K, 98K, 60K, 30K. The temperature dependence of 3 Bragg peaks was tracked while heating up from 30K to 300K, in order to study the magnetic order parameter.

Structure refinement

The nuclear structure was refined in $P6_3/mmc$ space-group in Fullprof. Scale, extinction parameters, atomic positions and individual isotropic displacement were refined. The occupancy of Fe2 (Wyckoff position 2c) was refined, and the magnetic structure is ferromagnetic, with a component along z-axis for both Fe1 and Fe2 (magnetic space-group $P6_3/mm'c'$), as shown in Fig.1.(a). The refinement with antiferromagnetic interlayer (space-group $P6_3'/mm'c$) shown in Fig.1.(b) was attempted but not successful. This structure was theoretically predicted for undoped Fe_3GeTe_2 [3], whereas the ferromagnetic interlayer should be the ground state for $Fe_{3-x}GeTe_2$ ($0.11<x<0.36$) [4].

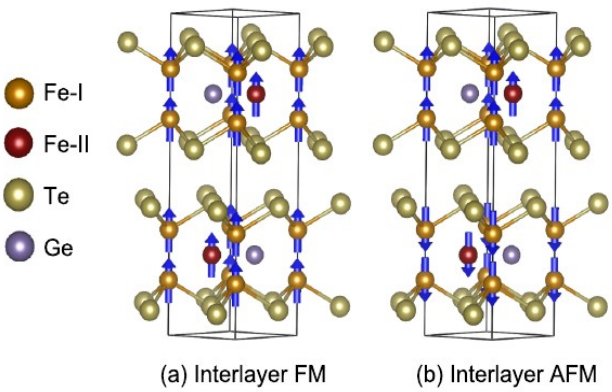

Fig. 1: Magnetic and nuclear structure in the case of an (a) interlayer ferromagnet, (b) interlayer antiferromagnet

For the 30K dataset, the scale and extinction parameters are fixed, and taken from the 60K refinement, because the least-squares algorithm found the optimum for a higher scale factor (1343 at 30K vs. 1247 at 60K), with an unphysical negative primary extinction parameter, and a magnetic moment of $1.3(2)\mu_B$, inferior to the $1.5(3)\mu_B$ found at 60K.The best refinements are found fixing the Fe1 and Fe2 magnetic moments to the same values.

The main refined parameters are reported in Table 1. For the 300K dataset, refinement with the ferromagnetic model was attempted, leading to a non-zero moment of $0.7(5)\mu_B$, with similar statistics as the purely nuclear model. It is important to point out that the different R_{Bragg} values for the temperatures are directly linked to the number of independent reflections which where measured. The reflections being measured in increasing order of Q, and the differences between observed and calculated intensities being more important for high-Q, the agreement factors are worst while the number of reflections are increasing. Absorption effects were not taken into account in these refinements, further corrections are necessary to improve these fits.

T(K)	300		98	60	30
a=b(Å)	3.9920(2)		3.9794(5)	3.9785(5)	3.9779(1)
c(Å)	16.3431(25)		16.3037(34)	16.2859(40)	16.2813(24)
V(Å^3)	225.5515		223.5949	223.2467	223.1191
Number of reflections	480		382	645	200
Independent reflections	159		128	189	76
R_{int}	1.95		1.6	1.52	1.44
	Only nuclear	Nuclear + magnetic			(scale and extinction fixed)
scale	1258(35)	1242(36)	1273(46)	1248(44)	1250
Biso Fe1	0.672(27)	0.649(27)	0.310(32)	0.240(23)	0.232(28)
Biso Fe2	0.830(72)	0.805(71)	0.639(91)	0.516(66)	0.445(105)
Biso Ge1	1.909(84)	1.848(83)	1.699(95)	1.578(85)	1.732(87)
Biso Te1	0.815(38)	0.786(37)	0.349(44)	0.254(30)	0.239(42)
Mz_Fe1		0.69(52)	1.44(33)	1.54(36)	1.65(17)
occ_Fe2	0.0717(19)	0.0719(20)	0.0725(24)	0.0716(25)	0.0720(15)
1-x	0.860(23)	0.863(24)	0.870(29)	0.859(30)	0.864(18)
R_{Bragg}	6.1505	6.1427	6.23	8.2537	4.43

Table 1: Magnetic and nuclear refinement (from Fullprof) results for the 4 measured temperatures.

Temperature dependence of Bragg peaks integrated intensities

The intensities of three Bragg peaks were tracked as a function of the temperature (while heating up from 30K to 300K). The amplitude squared of the magnetic structure factor is proportional to the integrated intensity for each reflection. Also, neutrons are only sensitive to magnetic moments perpendicular to the scattering vector $\vec{Q}$. If Fe moments are effectively along c-axis, reflections with $\vec{Q} \parallel \hat{c}$ should not give any magnetic contribution but only a nuclear one. Also, integrated intensities of reflections with $\vec{Q} \perp \hat{c}$ should be directly linked to the magnetic order parameter.

Reflections (2 2 0) and (3 0 0) indicate a second order magnetic transition (Fig. 2). The integrated intensities are decently fitted to a classical power law. This gives a critical exponent $\beta = 0.36(1)$ or $\beta = 0.38(1)$ fairly compatible to 3D Heisenberg model ($\beta = 0.365$)

as reported in [5]. The Curie temperature is found around 265(2) K, which is much higher than the value $T_C = 203\,\mathrm{K}$ measured from powder diffraction data for $Fe_{2.85}GeTe_2$ in [1].

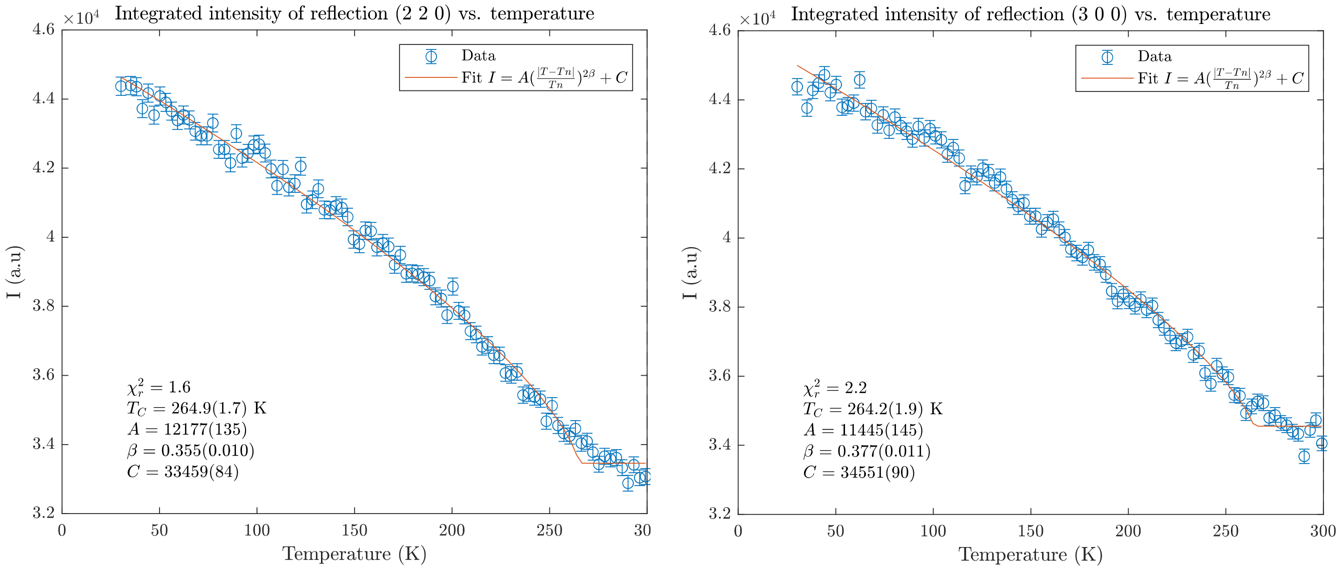

Fig. 2: Integrated intensities for reflections (220) and (300), and fits with power law.

Surprisingly, reflection (0 0 10) shows a non-negligible variation as a function of the temperature (Fig. 3). As shown in the structure refinement, where magnetic and nuclear contributions are both taken into account, the scale factor is not varying as a function of the temperature. Furthermore, the behavior of the variation seems to change after the transition temperature. This could be an indication of the presence of magnetic moments in the (ab)-plane, which is not allowed by the maximal magnetic space-groups from paramagnetic $P6_3/mmc$ space-group. Other models using subgroups of these magnetic space-groups can be further studied. For this reflection, the intensity can be fairly fitted to a linear law ($\beta = 0.5$), with a transition temperature $T_C = 256(13)\,\mathrm{K}$ compatible with the previous fits. Further macroscopic measurements can be useful to complete the studies on the magnetic structure of this single-crystal.

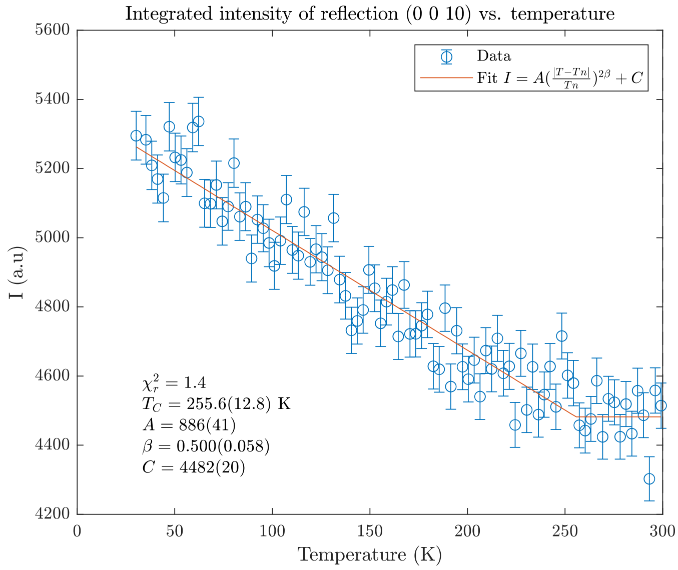

Fig. 3: Integrated intensities for reflection (0 0 10)

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