Experimental report

15/09/2022

Proposal: 5-11-441 **Council:** 4/2020

Title: The nuclear structure of CVT grownVI3

Research area: Materials

This proposal is a new proposal

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Samples: VI3

Instrument	Requested days	Allocated days	From	To
D9	5	6	08/07/2021	14/07/2021

Abstract:

Despite the strong two dimensionality of Van der Waals magnets, we have observed a strong effect on the bulk properties with stacking of the two dimensional layers. To understand the bulk properties of crystals, we therefore need to characterize the crystal structure. This proposal aims at understanding the magnetic and crystal structure of our VI3 crystals grown using chemical vapor transport (CVT). Diffraction measurements have found that the structure may not be the hexagonal structure reported in the literature in our single crystals. We request time on D9 to characterize the nuclear structure (at high temperatures) and also the magnetic structure (for temperatures below 50 K).

Proposal 5-11-441, 6 days on D9 (08/07/2021-14/07/2021)
The nuclear structure of CVT grownVI_3

Introduction

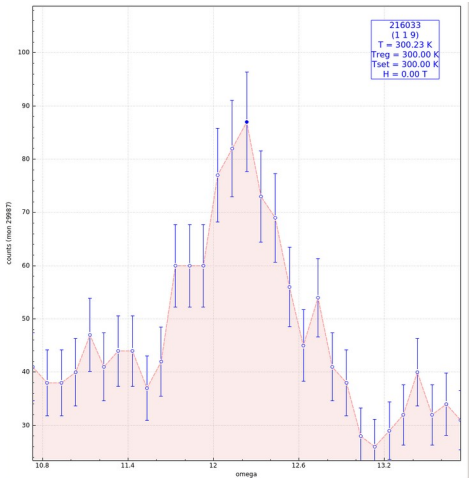
VI_3 is a two-dimensional ferromagnet based on honeycomb vanadium layers. Several x-ray diffraction experiments were performed of both powder and single-crystals. Most of the studies show a structural transition at 79K followed by a ferromagnetic transition near 50K. However, there are still some discussions on the space-group of the compound, on whether it is monoclinic [1] or trigonal [2] at room temperature, and after the structural transition. Some studies also point out an additionnal structural and magnetic transition around 32K [3, 4]. So far neutron diffraction experiments were only performed on powder sample, and single-crystal neutron diffraction is the next step for refining magnetic structure. This is the purpose of this experiment on D9.

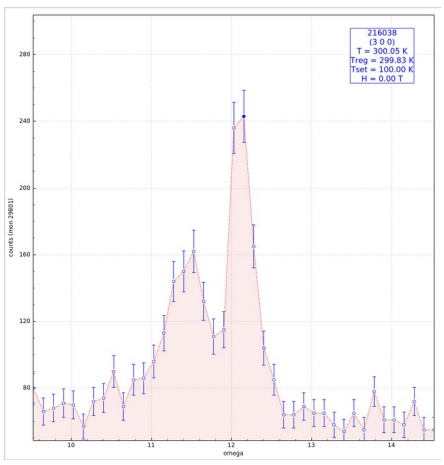
Experiment

VI_3 single-crystals were sealed in quartz ampoules, which were broken within a glovebag filled with helium. In the same glovebag, the crystal was glued onto the sample holder. Then, it was carefully moved (~5 seconds in the air) to the instrument. Helium was projected onto the sample while checking the centering of the sample, then it was protected inside the Displex. We noticed that the sample was slighty bended on the borders, because of the thickness of the crystal, or some degradation. We actually changed the sample twice to get proper reflections to work with.

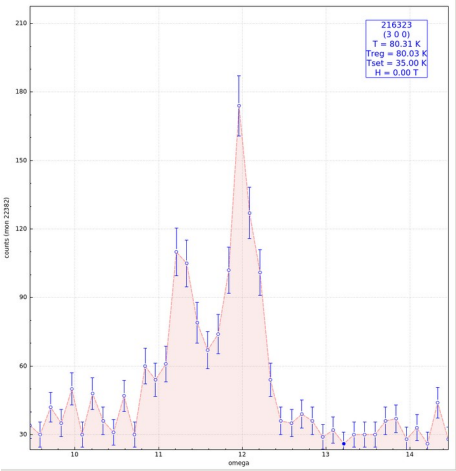
Bragg peak collection

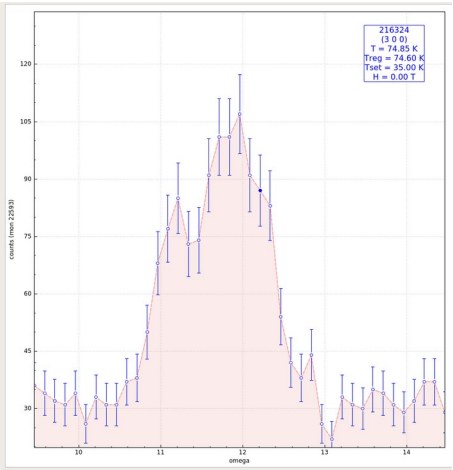
First of all, the reflections are quite broad along the c-axis (stacking faults), and two domains can be seen in the (ab) plane.





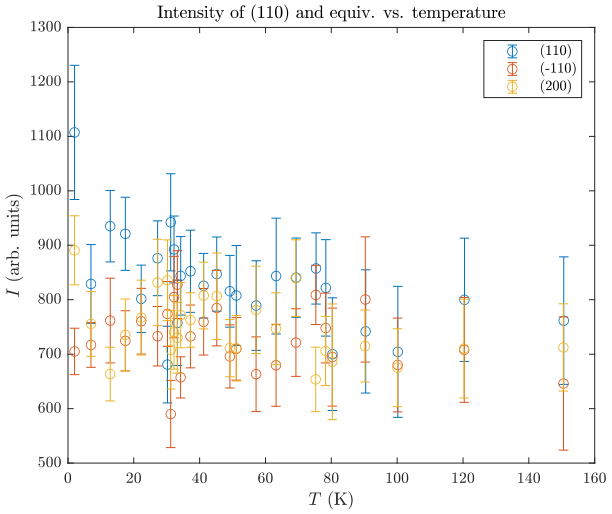
Bragg peaks were collected at 100K, 65K and 35K. The shapes of the peaks make the intensities integration more difficult, therefore data reduction was not successful ($R_{\text{int}} = 30\%$ at 100K). In addition, the comparison between 65K (paramagnetic phase) and 35K (ferromagnetic phase) did not give significant evidence of magnetic ordering. The intensities were integrated using RACER software, taking into account the ellipsoid of resolution, but further studies are necessary to take into account the different shapes of the Bragg peaks and get a proper evaluation of the intensities. The shapes of some peaks evolve with temperature. This is the case for reflection (300) which shows two peaks at room temperature, which merge at the structural transition (below 80K). Similar behavior was not found at lower temperature.

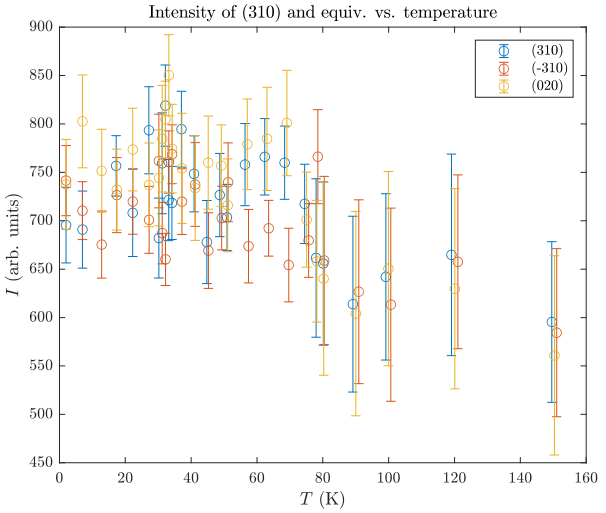




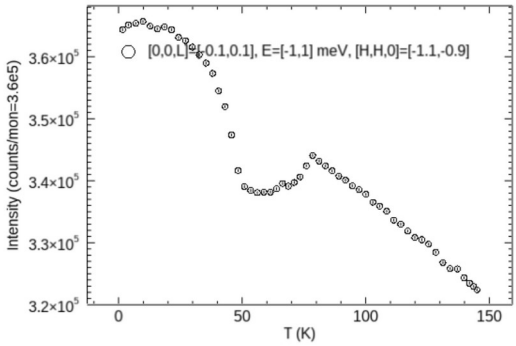
Temperature dependence

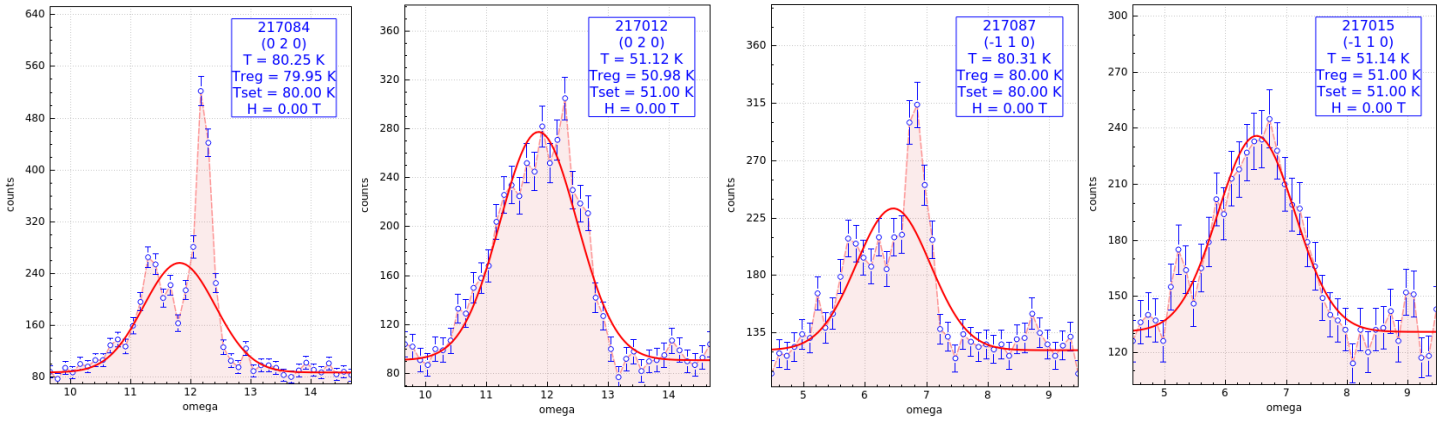
Finally, we measured “good” peaks, in the sense that they were well-resolved, as a function of temperature. These 12 peaks formed 4 groups of 3 equivalent reflections in R-3 space-group, which are no longer equivalent (only a pair of them are) in C2/m space-group. Thus the intensities are allowed to be different at the structural transition at 79K. In-plane and out-of-plane reflections were measured to track any ferromagnetic component below 50K.





Within the uncertainties, one can not conclude a shift from equivalent to non-equivalent reflections at the structural transition. Moreover, no track of magnetic order was found in these peaks. But this can be due to the low magnetic structure factor for these peaks, but measurements on MACS showed the magnetic order in reflection (110). The merging of the two peaks at the structural transition is still visible in most of the (HK0) reflections below the structural transition temperature.





[1] S. Tian, J.-F. Zhang, C. Li, T. Ying, S. Li, X. Zhang, K. Liu, and H. Lei, J. Am. Chem. Soc. **141**, 5326 (2019).
[2] S. Son, M. J. Coak, N. Lee, J. Kim, T. Y. Kim, H. Hamidov, H. Cho, C. Liu, D. M. Jarvis, P. A. C. Brown, J. H. Kim, C.-H. Park, D. I. Khomskii, S. S. Saxena, and J.-G. Park, Phys. Rev. B **99**, 041402 (2019).
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[4] T. Marchandier, N. Dubouis, F. Fauth, M. Avdeev, A. Grimaud, J.-M. Tarascon, and G. Rousse, Phys. Rev. B **104**, 014105 (2021).