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### MAGNETIC ORDERING AND SPIN WAVE DYNAMICS IN TRANSITION METAL ARSENIDES

### BY

### MANOHAR H. KARIGERASI

### **DISSERTATION**

Submitted in partial fulfillment of the requirements for the degree of Doctor of Philosophy in Materials Science and Engineering in the Graduate College of the University of Illinois at Urbana-Champaign, 2021

### Urbana, Illinois

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### **CHAPTER 6**

# TWO-STEP MAGNETIC ORDERING INTO A CANTED STATE IN FERRIMAGNETIC MONOCLINIC Mn<sub>3</sub>As<sub>2</sub>

Reprinted with permission from Manohar H Karigerasi, Bao H. Lam, Maxim Avdeev and Daniel P. Shoemaker, Journal of Solid State Chemistry 294, 121901 (2020). Copyright 2020 by the Elsevier. This manuscript version is made available under the CC-BY-NC-ND 4.0 license http://creativecommons.org/licenses/by-nc-nd/4.0/ In this work, I carried out SQUID and DSC measurements. I also carried out the magnetic structure refinement from NPD data and wrote the paper with help from coauthors. Bao Lam synthesized the samples and took SEM images.

### 6.1 Abstract

We report the magnetic structure of monoclinic Mn<sub>3</sub>As<sub>2</sub> at 3 K and 250 K using neutron powder diffraction measurements. From magnetometry data, the Curie temperature of Mn<sub>3</sub>As<sub>2</sub> was confirmed to be around 270 K. Calorimetry analysis showed the presence of another transition at 225 K. At 270 K, Mn<sub>3</sub>As<sub>2</sub> undergoes a k=0 ferrimagnetic ordering in the magnetic space group C2/m (#12.58) with Mn moments pointing along b. Below 225 K, there is a canting of Mn moments in the ac plane which produces a multi-k non-collinear magnetic structure in space group C2/c (#15.85). The components of Mn moments along b follow b ordering and the components along b and b have b propagation vector. The change in the magnetic ground state with temperature provides a deeper insight into the factors that govern magnetic ordering in Mn-As compounds.

### 6.2 Introduction

The Mn-As phase diagram contains a rich collection of phases with various magnetic structures [22,43, 72–75,103]. Most of the known compounds in this phase-space can be roughly divided into two groups. Compounds in one group are of the form  $Mn_{2+n}As_{1+n}$  where, starting with stripes of square-planar Mn-As units running along a at n = 0, every additional Mn-As involves adding an Mn-As octahedral

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