

MAGNETIC ORDERING AND SPIN WAVE DYNAMICS IN TRANSITION METAL
ARSENIDES

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BY

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DISSERTATION

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Abstract

Metallic antiferromagnets have gained interest in recent times due to the possibility of being useful as a memory device. Arsenic forms a large pool of magnetic metals in combination with other transition metals that have largely been ignored so far. In this report, we discover a new ternary metallic arsenide in the Cu-Mn-As phase space, identify its chemical and magnetic structure, and characterize its electrical and magnetic properties. We also carry out the magnetic structure refinement of Mn_3As_2 from neutron powder diffraction data at different temperatures to understand the magnetic ordering in Mn-As compounds. Using inelastic neutron scattering measurements, we determine exchange interactions in Fe_2As , which has the same structure as CuMnAs, showing a highly 2D magnon character although the phonons are 3D. Finally, we report a magnetic-structural coupled transition across 300 K in tetragonal CuMnAs and determine the correct magnetic structure of the compound.

Acknowledgments

This project would not be possible without many people. Firstly, thanks to my advisor, Prof. Daniel P. Shoemaker.

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Chapter 1

Introduction

1.1 Magnetic information storage

2 In information memory storage devices, there is typically a trade-off between the optimum
3 speed or response time and the complexity and size of memory storage [1]. Volatile memory
4 refers to temporary memory storage where the data is lost when the power is removed.
5 Volatile memory such as SRAM (static random access memory) and DRAM (dynamic random
6 access memory) are used as CPU caches and main memory respectively. SRAM has much
7 faster access times and does not require periodic refreshing. However, it requires four to six
8 transistors per bit as compared to one transistor and one capacitor in DRAM devices [2]. Non-
9 volatile memory (NVM) storage devices on the other hand, retain their data for a long period
10 of time until perturbed. Modern computers mostly use flash memory based solid state drives
11 (SSD) and magnetic hard disk drives (HDD) for storing large amounts of data permanently [2].
12 The first HDD was invented in 1956 by IBM and since then, has seen more than eight orders of
13 magnitude improvement in the storage density. However, the trilemma in magnetic recording
14 between poor thermal stability, coercive fields and signal-to-noise ratio has resulted in the
15 HDD reaching a saturation limit in their device performance [3]. Flash memory uses floating
16 gate MOSFETs (metal oxide semiconductor field effect transistors) to store memory and does
17 not contain any moving parts. Although SSD have dominated the NVM marketshare last few
18 years, there is an increasing need for alternative NVM technologies that are fast, low power
19 consuming and have high storage density [2].

20 One such emerging NVM is MRAM (magnetoresistive random access memory). Unlike
21 flash memory which uses electronic charge as a medium of memory storage, MRAM uses the
22 electronic spin degree of freedom to store information. Unlike charge based storage devices,
23 MRAM is stable against perturbations such as ionizing radiation [4]. MRAM devices consist
24 of cells with magnetic tunnel junctions (MTJ) that have two ferromagnet (FM) layers separated
25 by an insulating layer. One of the layer is pinned where the magnetization orientation is fixed
26 and acts as a reference layer. Depending on the orientation of the free layer, the tunneling
27 magnetoresistance (TMR) is high or low and hence, memory can be read out using electrical
28 currents [3]. Early MRAMs were written by induced fields from heavy currents passed on
29 the adjacent layer. With recent developments in spin transfer torque (STT) in ferromagnets,

30 it has become possible to write using electrical currents [5]. This has reduced the power
31 consumption significantly and made commercialization of MRAM devices possible [3, 6].

32 **1.2 Antiferromagnets for potential applications as a memory unit**

33 Historically, antiferromagnets (AFM) have been used as inactive components in MTJ, primarily
34 in exchange biasing the pinned FM layer. However in 2010, Gomonay *et al.* [7] proposed
35 electrical switching of AFMs using STT by passing a spin polarized current injected from
36 a fixed FM layer through the AFM layer. The electrical current gets spin polarized in the
37 FM layer and transfers its angular momentum to the AFM moments to switch it from one
38 orientation to another. There are advantages to using AFM over FM in MRAM devices. AFM
39 are not easily affected by external magnetic fields and do not produce stray fields of their
40 own. They have smaller domains which would allow for higher storage densities [4]. Since
41 the precession frequency of AFM moments is determined the geometric mean of exchange
42 and anisotropy energies, the dynamics in AFM materials occur at THz timescales which is
43 two orders of magnitude higher than in FM [8]. Although the AFM can be switched using
44 electrical currents from parallel to perpendicular orientation with respect to the FM mag-
45 netization direction, the reverse process cannot be obtained using electrical current. High
46 magnetic fields above the spin flop transition of the AFM needs to be applied in order to
47 switch back the AFM to its original state [7].

48 Unlike previously discussed STT MRAM devices, spin orbit torque based electrical switch-
49 ing in broken inversion symmetry FM does not require the presence of a FM polarizer [9].
50 This concept of the presence of relativistic fields is applicable to AFM as well provided the
51 local moments do not sit on centrosymmetric sites. If the two sublattices are related to each
52 other by a center of inversion, then the current induced spin polarized fields are staggered
53 across the two sublattices [4, 10, 11]. This results in a uniform fieldlike torque experienced by
54 the order parameter. This is possible in bulk materials that are globally centrosymmetric but
55 locally non-centrosymmetric and the two sublattices are related to each other by a center of
56 inversion. It was initially demonstrated in the case of epitaxially grown tetragonal CuMnAs
57 thin films on GaP substrate [4] and since then, it has also been shown in Mn₂Au and CuMnAs
58 sputtered films as well [12, 13]. Observation of electrical switching behavior in AFM requires
59 the presence of degenerate Néel vectors like in CuMnAs as shown in Fig. 1.1(a) as opposed to
60 compounds like MnF₂ where the Mn moments point along *c* in Fig. 1.1(b).

61 **1.3 Exploration of Cu-Mn-As phase space**

62 Compounds in Cu-Mn-As phase space have attracted a lot of attention in recent times mainly
63 due to the exotic properties of tetragonal and orthorhombic CuMnAs. As mentioned earlier,

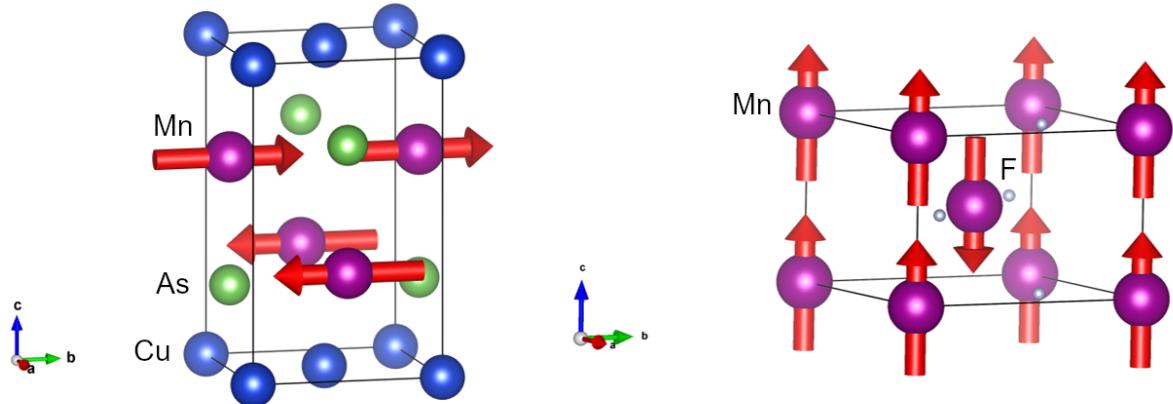


Figure 1.1: The magnetic structures of tetragonal CuMnAs and MnF₂ are shown in (a) and (b), respectively.

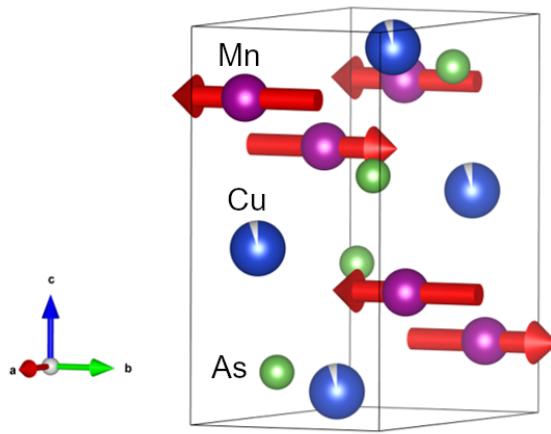


Figure 1.2: Magnetic structure of orthorhombic CuMnAs

tetragonal CuMnAs was the first antiferromagnet where electrical switching was supposedly demonstrated. Orthorhombic CuMnAs, shown in Fig. 1.2, was the first magnetic compound to be proposed as a Dirac semimetal. It is a special compound where the inversion and time reversal symmetry of the magnetic structure is broken but their combined symmetry (*PT* symmetry) is still preserved. Based on the orientation of the AFM order parameter, the compound changes from a conducting to an insulating phase. Hence, there are voltage based switching applications that have been proposed for this compound [14].

Despite the growing importance of the compounds in Cu-Mn-As system, the Cu-Mn-As ternary phase space has not been explored properly. There are four known ternary compounds including both the polymorphs of CuMnAs, orthorhombic CuMn₃As₂ and Cu₂Mn₄As₃ as shown in Fig. 1.3 [15–18]. Bulk orthorhombic CuMnAs can be grown us-

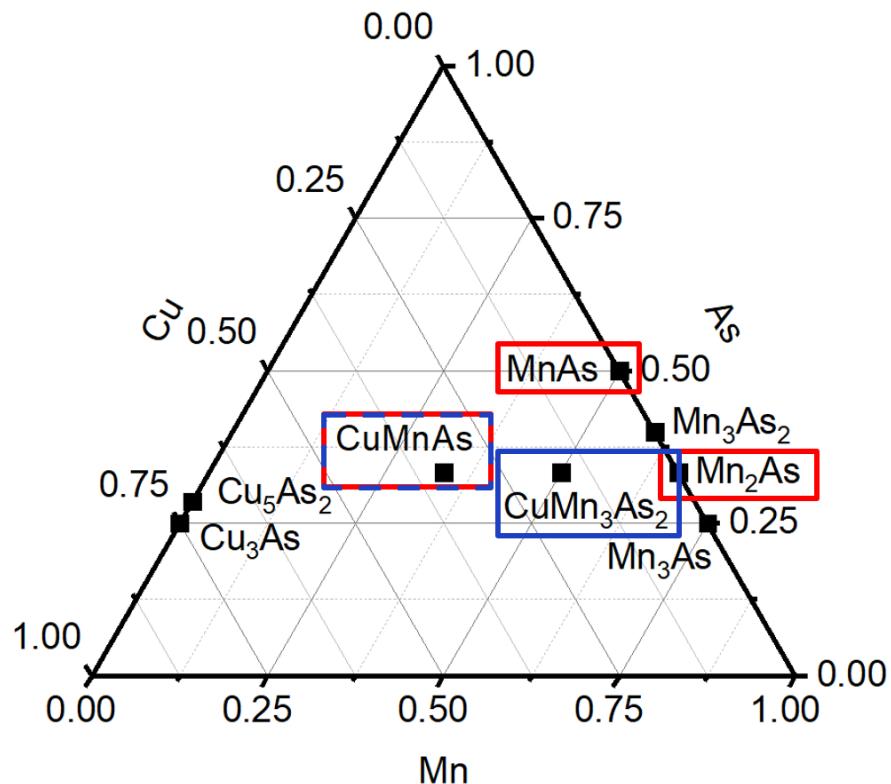


Figure 1.3: Cu-Mn-As ternary phase diagram highlighting the known ternary compounds in blue and known magnetic structures in red. Not all compounds in this system are shown here.

75 ing traditional solid state synthesis routes by Cu, Mn and As powders in stoichiometric ratio
76 and heating the powders to 1000°C. In order to synthesize pure bulk tetragonal CuMnAs,
77 we have to go off-stoichiometry and either substitute Mn with Cu or As [18]. Hence, it is
78 important to explore different regions in the Cu-Mn-As system and verify the stability of
79 different ternary compounds. The magnetic structures in the Cu-Mn-As system have also
80 not been identified for most of the compounds. Apart from the four Cu-Mn-As ternary
81 compounds, there are more than ten Mn-As binary compounds present in this system. The
82 magnetic structures are known only for the two previously mentioned CuMnAs compounds,
83 Mn₂As and MnAs as shown in Fig. 1.3 [16,17,19–22]. Since most, if not all, the binary Mn-As
84 compounds are metallic, there is a need to magnetically characterize the compounds and
85 identify their magnetic structures.

86 1.4 Exchange interactions in Cu₂Sb type structures

87 If we want to understand the electrical switching behavior in metallic antiferromagnets, we
88 should be able to quantify the fundamental energies such as magnetocrystalline anisotropy
89 and exchange interactions in materials like CuMnAs. CuMnAs has a Cu₂Sb structure type.
90 Other materials with this structure includes Mn₂As, Cr₂As, Fe₂As, CrMnAs, MnFeAs etc.
91 [23–25]. Although Mn₂As, Cr₂As and Fe₂As have the same structure, the magnetic ground
92 state is different in all three compounds. The strength and sign of direct exchange interactions
93 between two magnetic atoms is a result of the nature of orbital overlap between the two
94 magnetic atoms [24]. Since these materials are metallic, there are two contributions to indirect
95 exchange interactions. One contribution arises from superexchange interactions mediated by
96 As atoms and the other contribution comes from RKKY (Ruderman–Kittel–Kasuya–Yosida)
97 interactions [25]. It is important that we are able to determine what spin interactions are
98 relevant and how does it affect the magnetic ordering in these materials. It is also crucial that
99 we are able to verify the computational methods and the exchange coupling values obtained
100 from these methods so that we can use these methods for other systems as well.

Chapter 2

Theory of electrical switching in metallic antiferromagnets

101 **2.1 Hidden spin polarization in centrosymmetric crystals**

102 **2.2 Components of torque from non-equilibrium current induced
103 spin polarization**

104 **2.3 Torque in tetragonal CuMnAs**

105 **2.4 Torque in Fe₂As**

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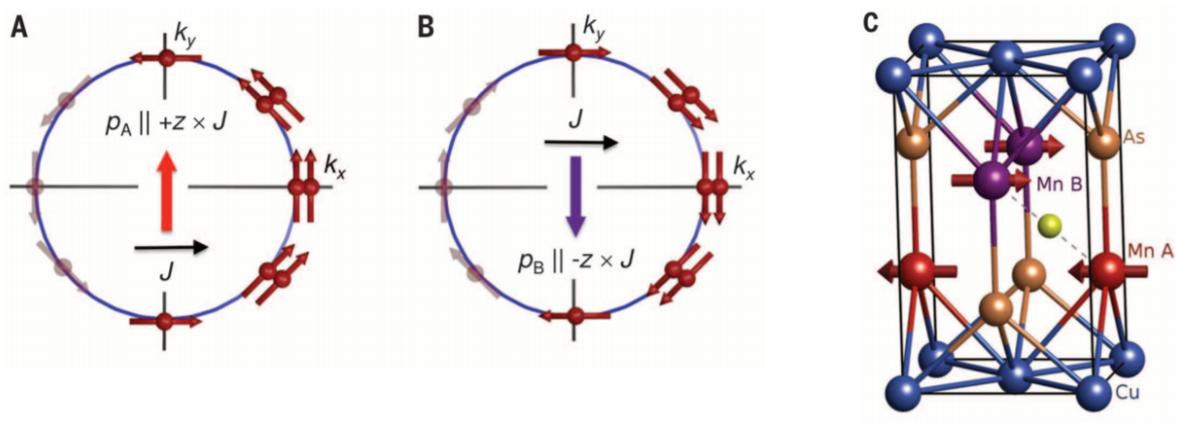


Figure 2.1: Electronic band structure

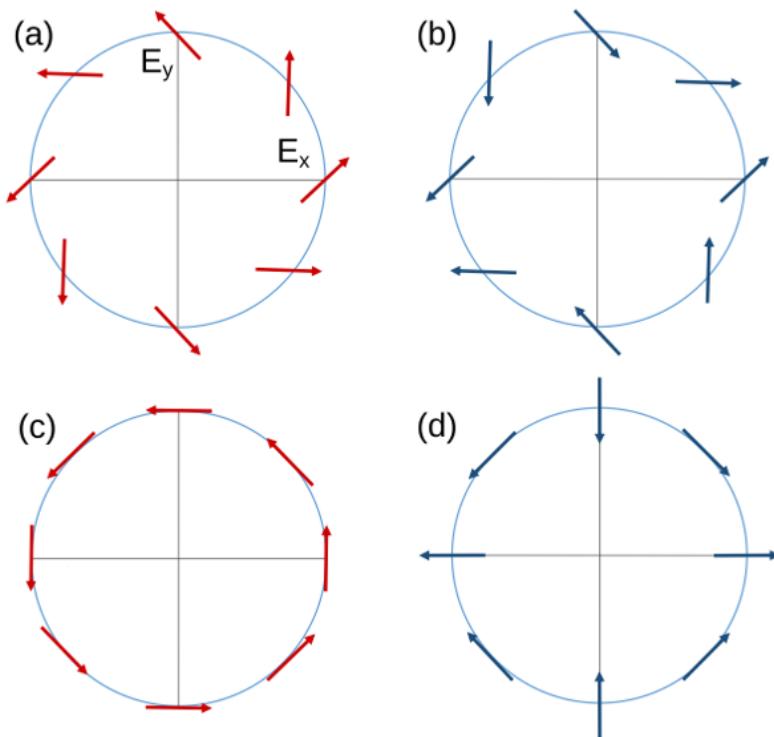


Figure 2.2: Electronic band structure

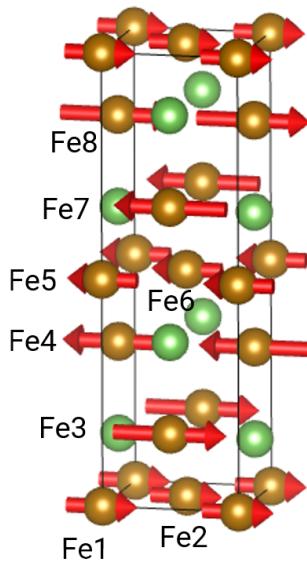


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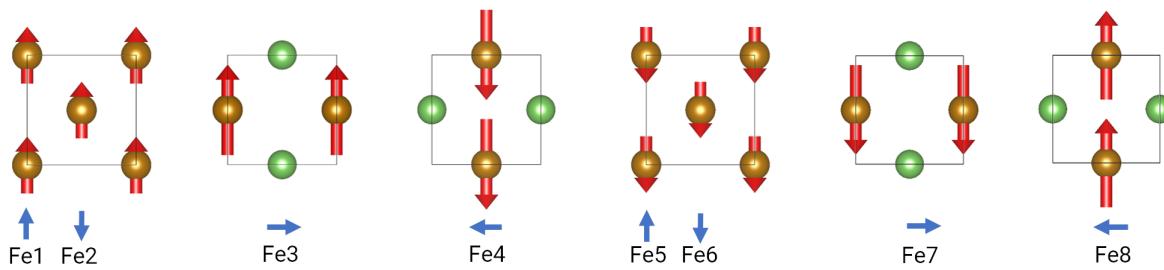


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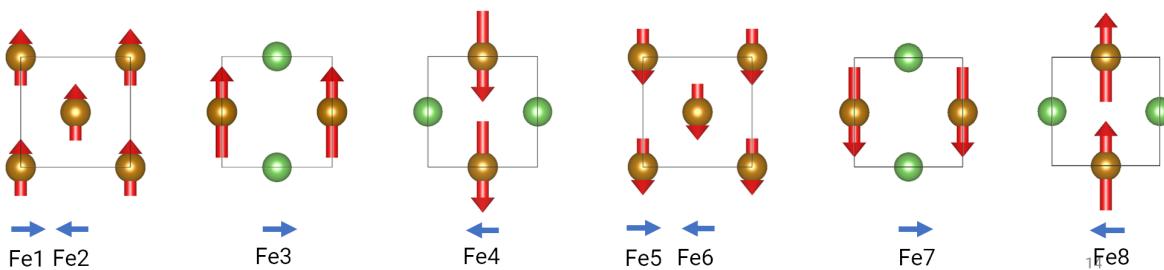


Figure 2.5: Electronic band structure

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Chapter 3

Magnetic structure refinement from neutron diffraction measurements

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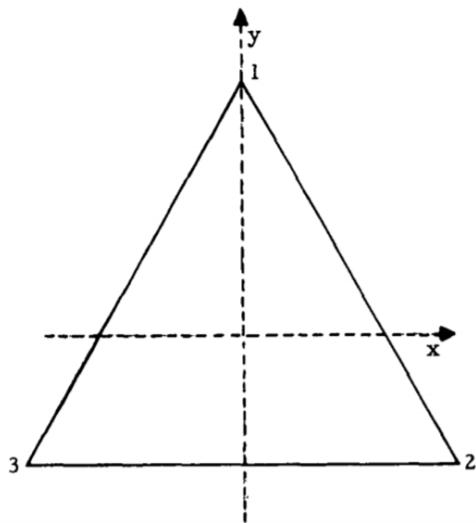


Figure 3.1: Electronic band structure

	E	C_3	C_3^2	σ_1	σ_2	σ_3
E	E	C_3	C_3^2	σ_1	σ_2	σ_3
C_3	C_3	C_3^2	E	σ_3	σ_1	σ_2
C_3^2	C_3^2	E	C_3	σ_2	σ_3	σ_1
σ_1	σ_1	σ_2	σ_3	E	C_3	C_3^2
σ_2	σ_2	σ_3	σ_1	C_3^2	E	C_3
σ_3	σ_3	σ_1	σ_2	C_3	C_3^2	E

Figure 3.2: Electronic band structure

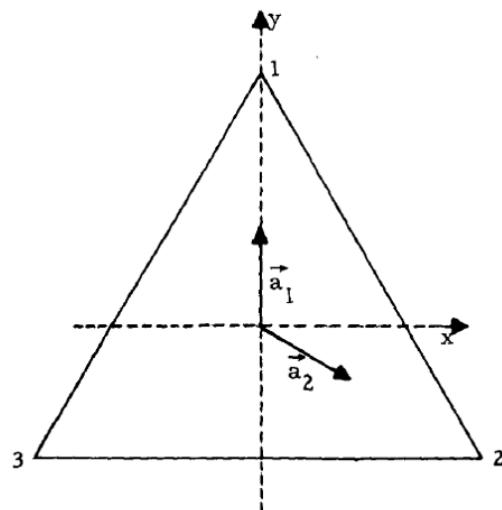


Figure 3.3: Electronic band structure

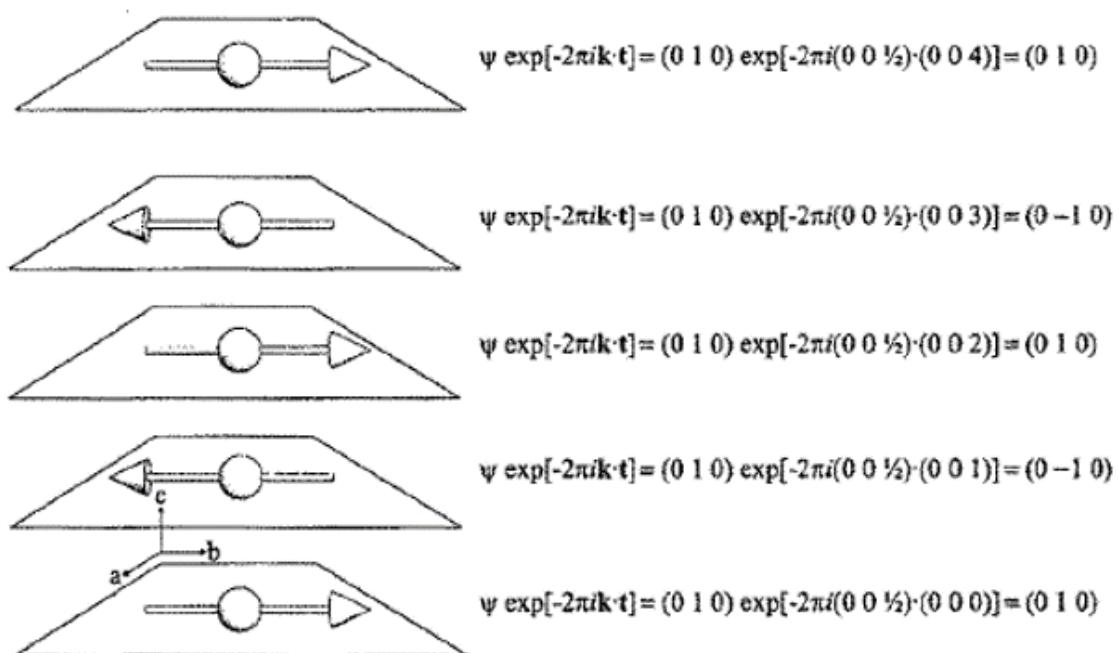


Figure 3.4: Electronic band structure

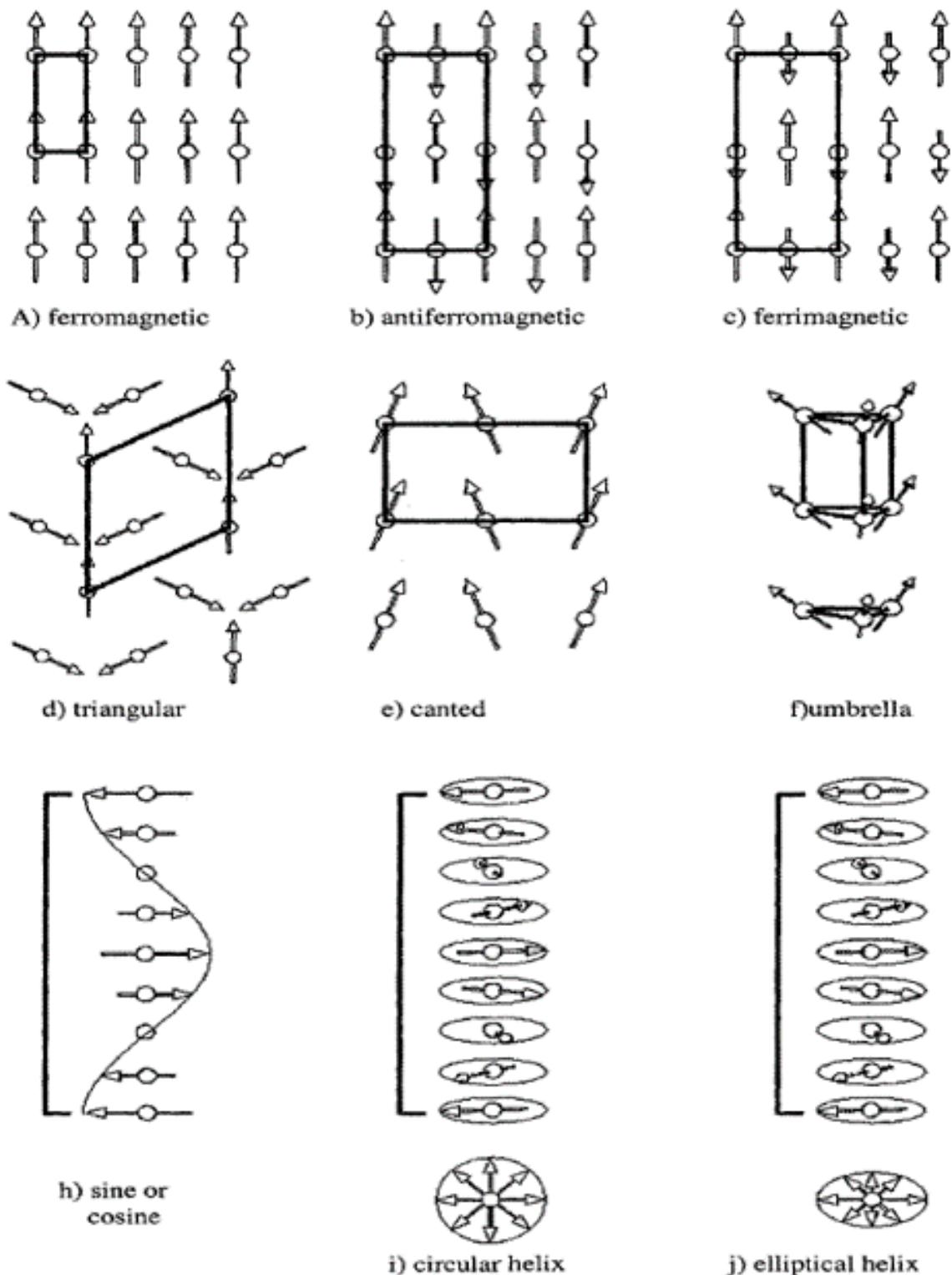


Figure 3.5: Electronic band structure

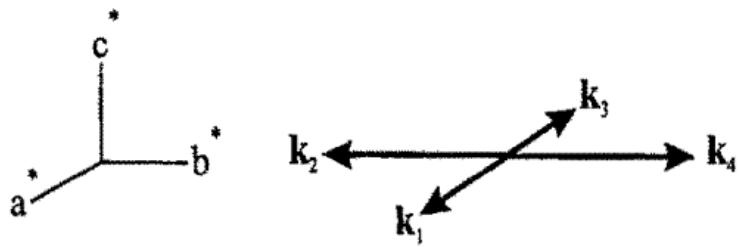


Figure 3.6: Electronic band structure

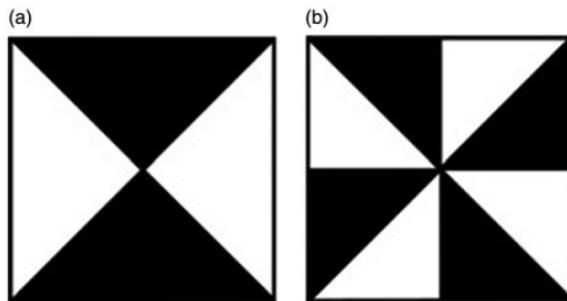


Figure 3.7: Electronic band structure

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Chapter 4

Materials synthesis and characterization

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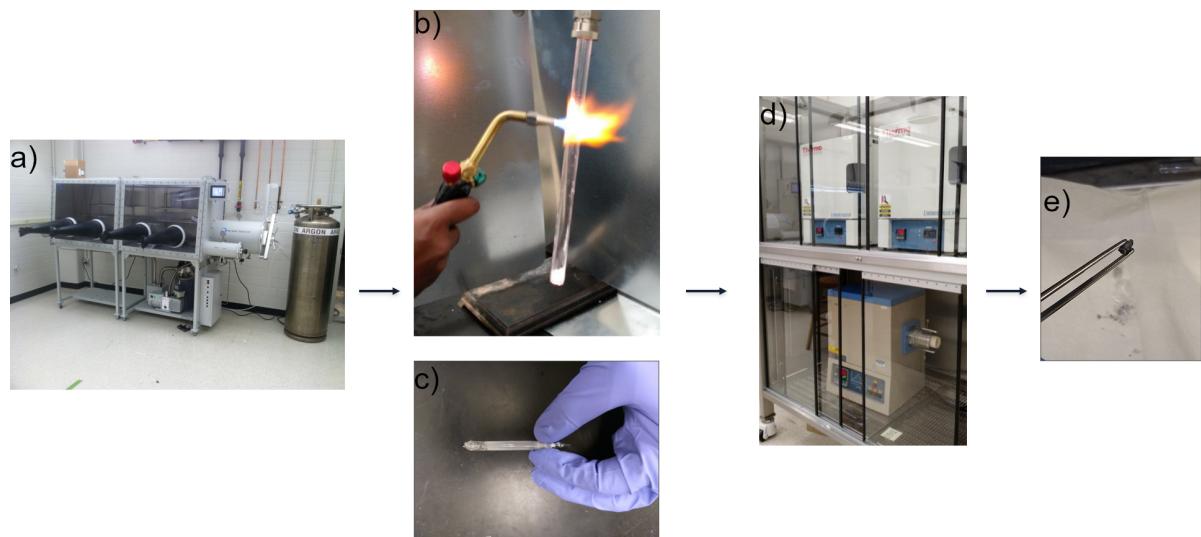


Figure 4.1: Electronic band structure

Chapter 5

Discovery and magnetic frustration of hexagonal Cu_{0.82}Mn_{1.18}As

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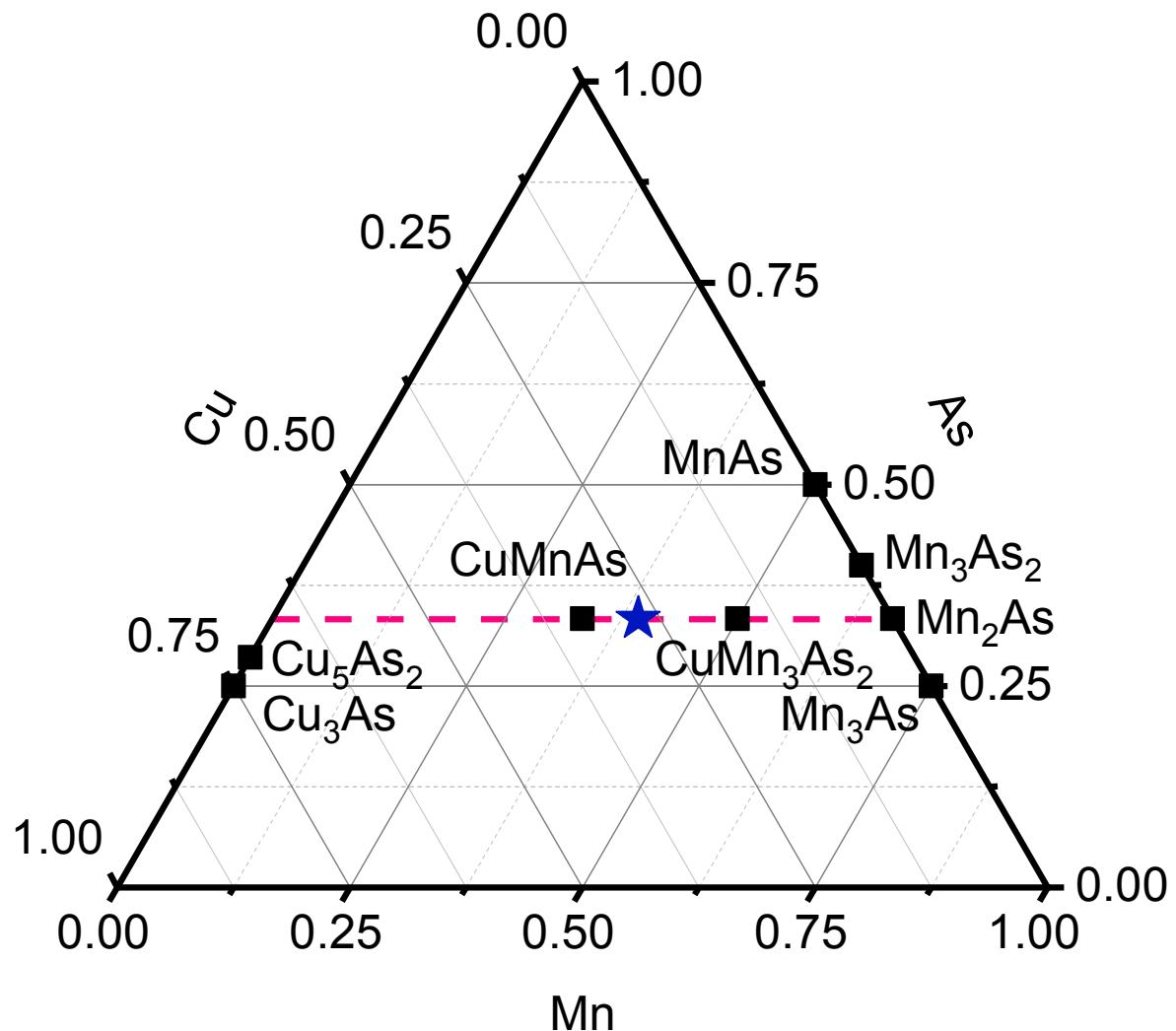


Figure 5.1: Electronic band structure

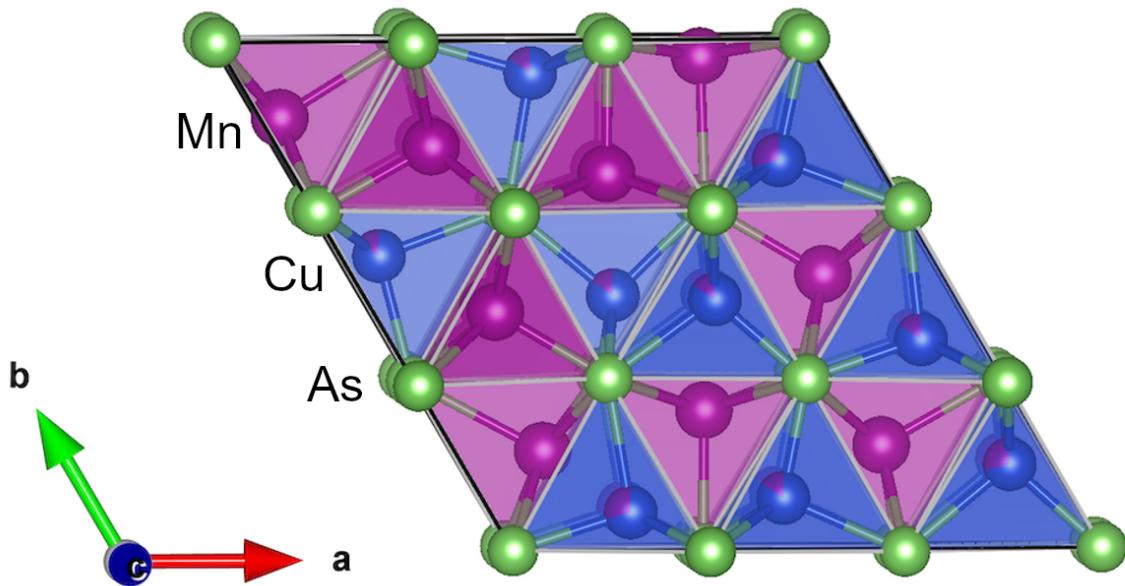


Figure 5.2: Electronic band structure

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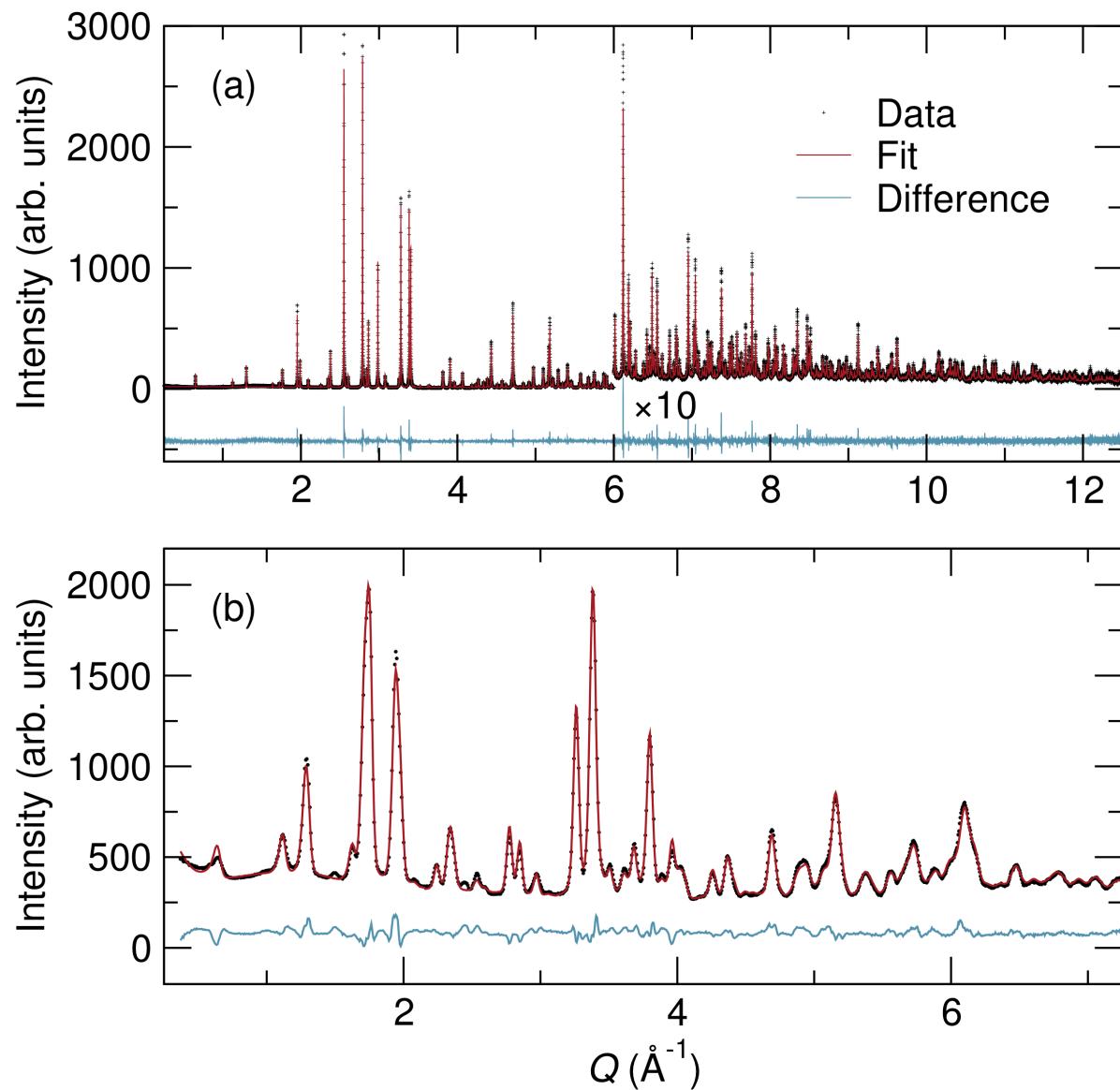


Figure 5.3: Electronic band structure

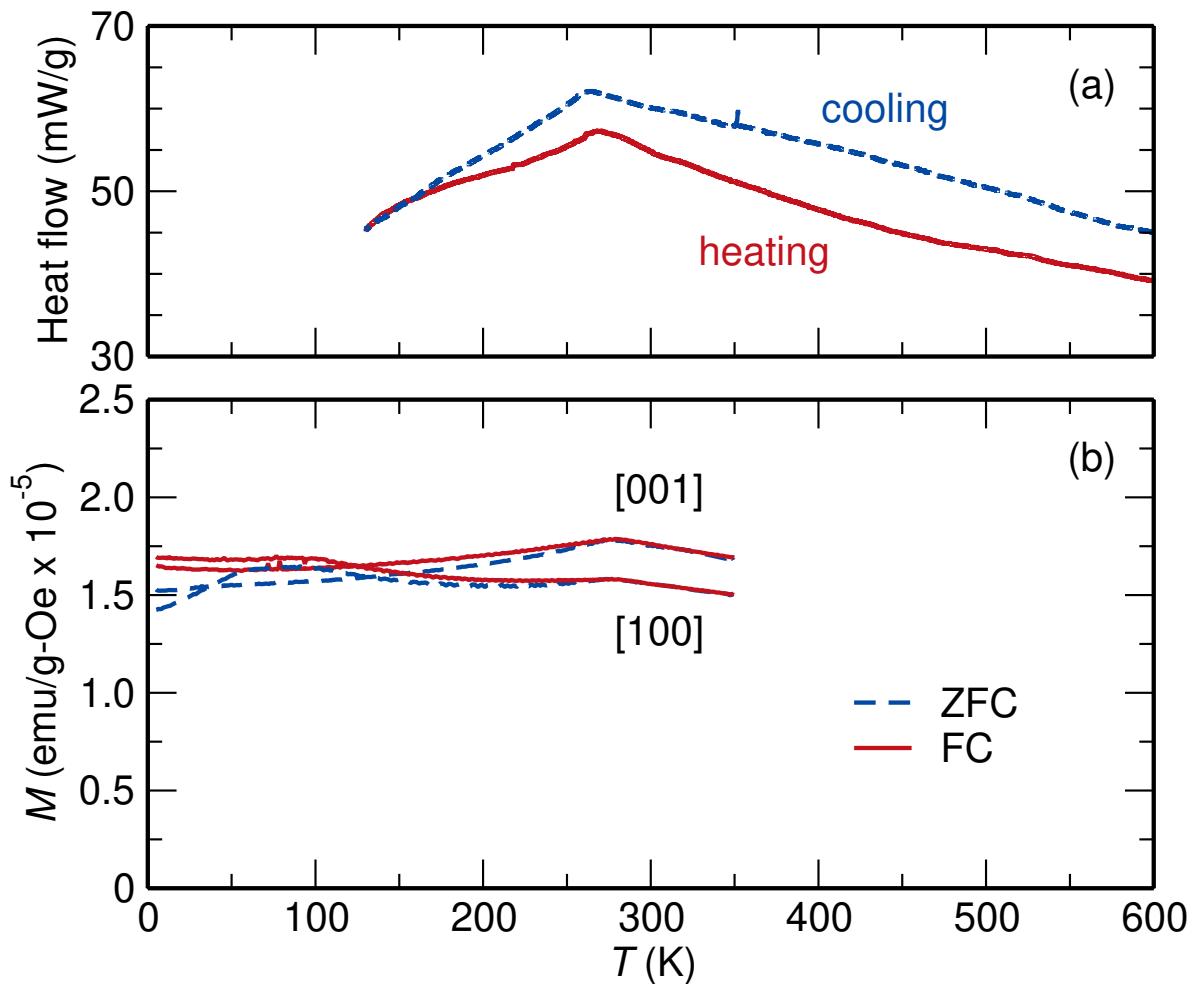


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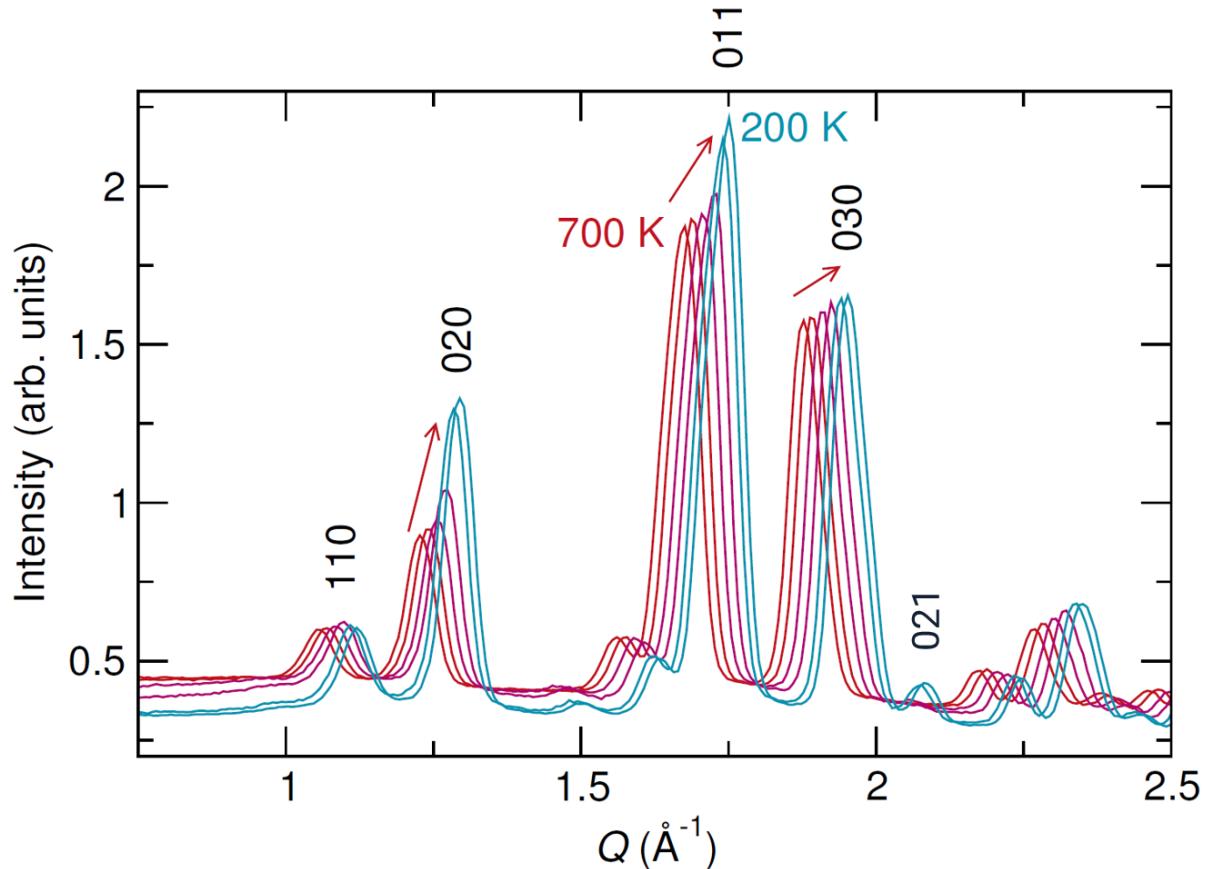


Figure 5.5: Electronic band structure

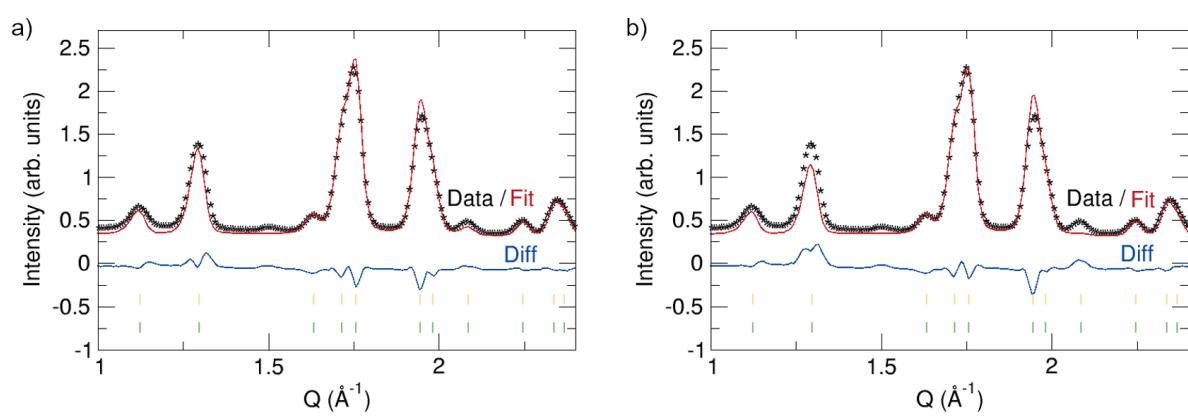


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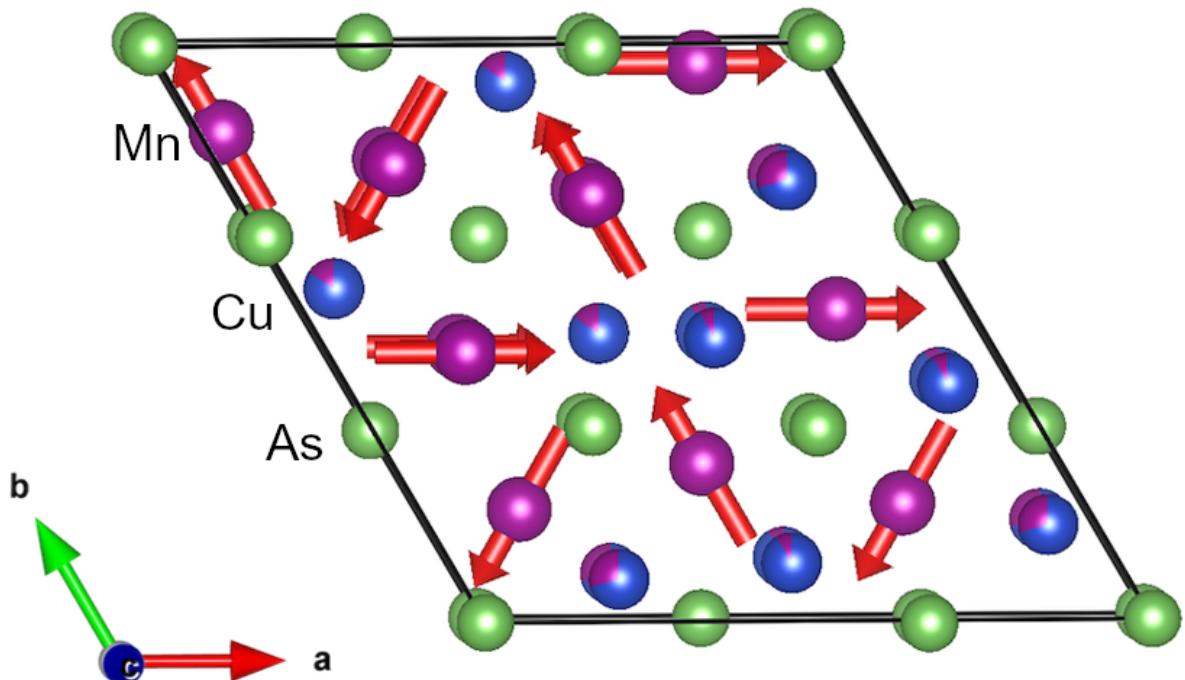


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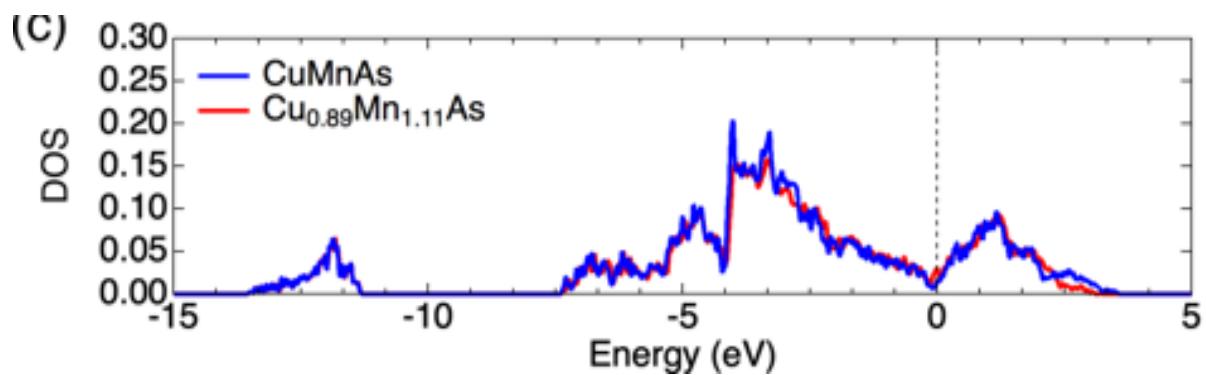


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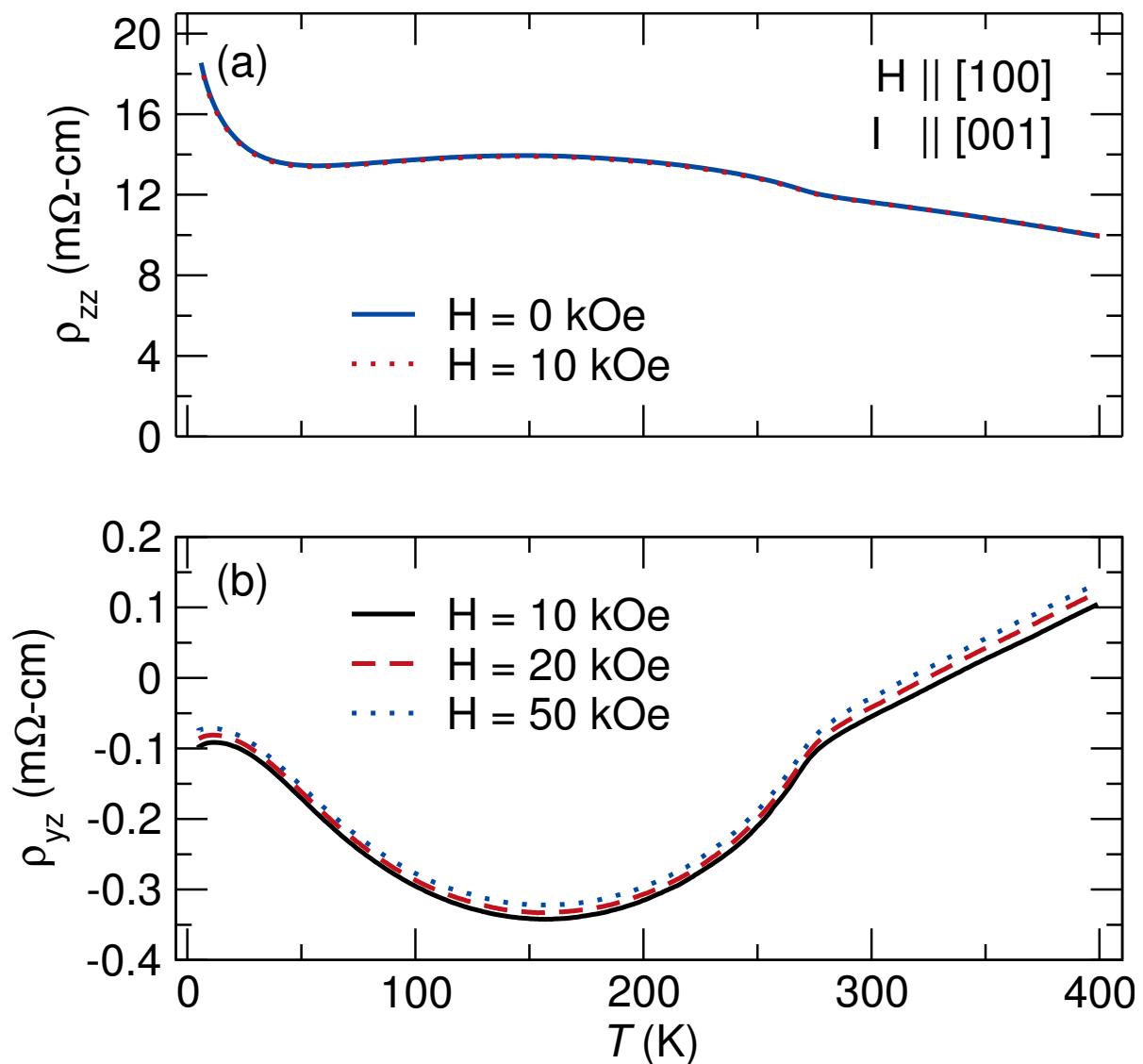


Figure 5.9: Electronic band structure

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Chapter 6

Two step magnetic ordering in monoclinic Mn₃As₂

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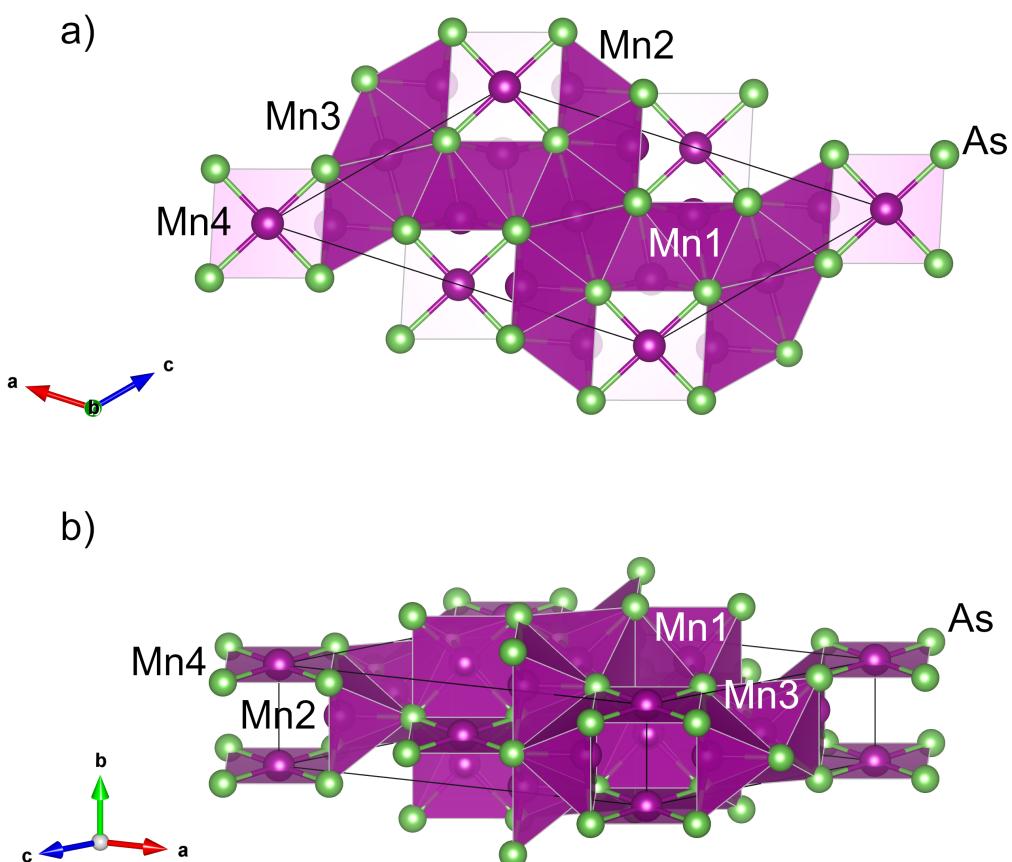


Figure 6.1: Electronic band structure

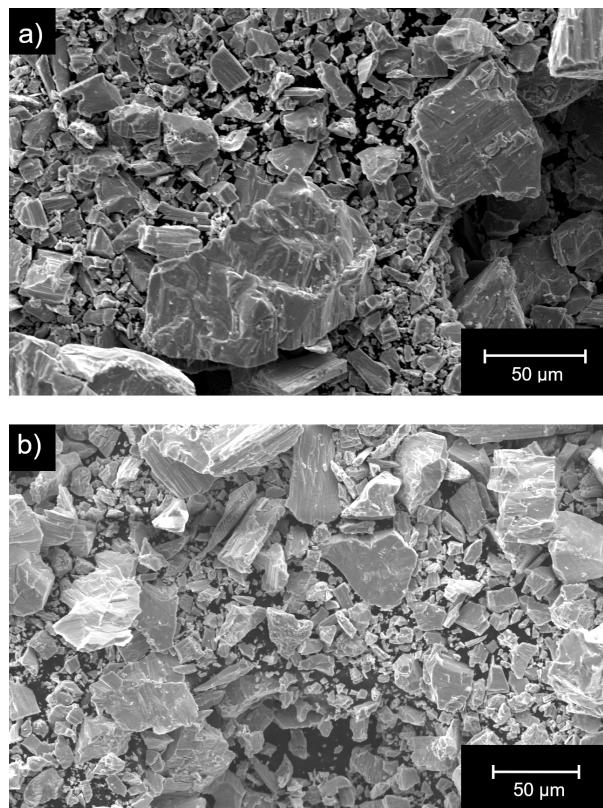


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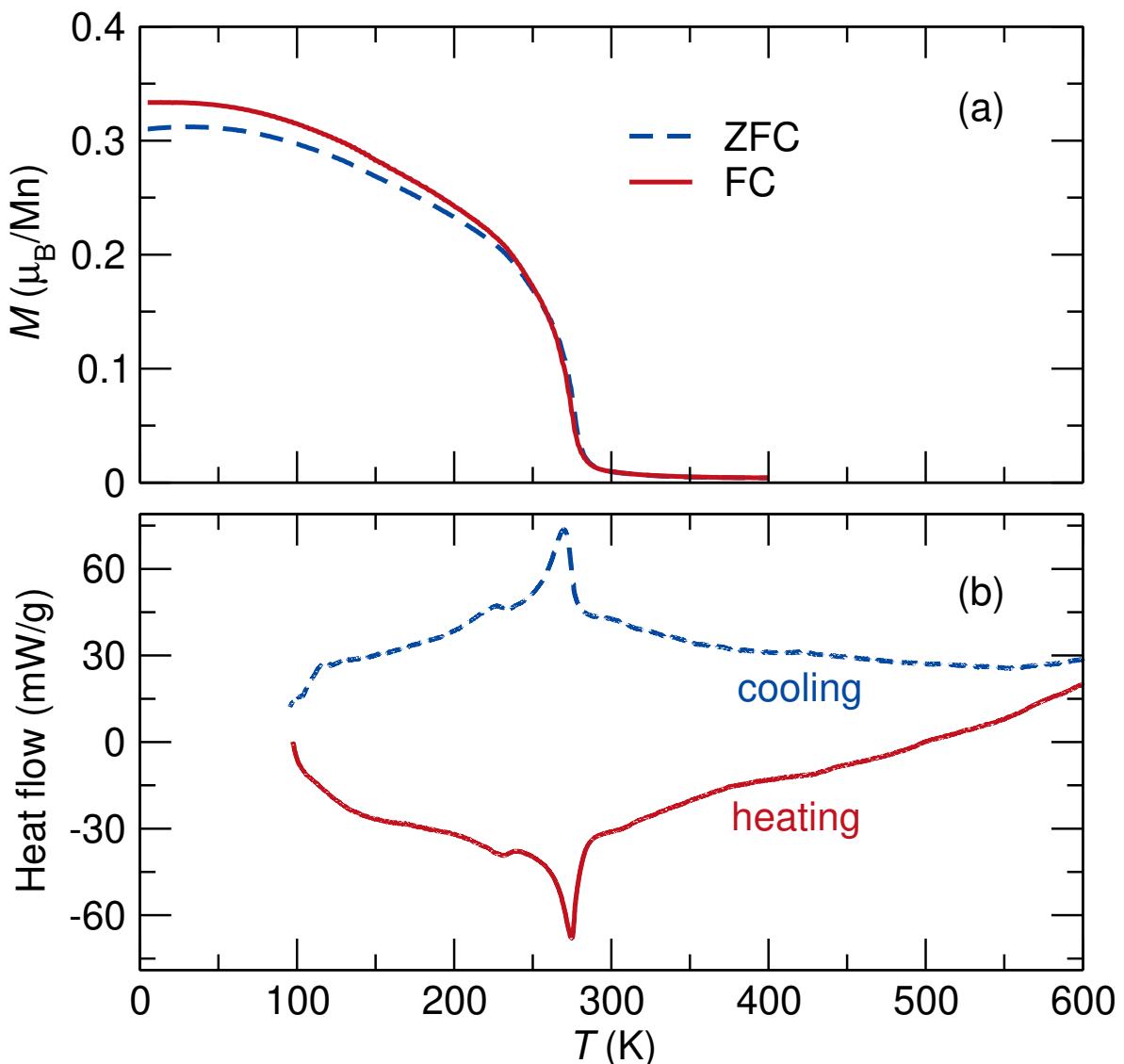


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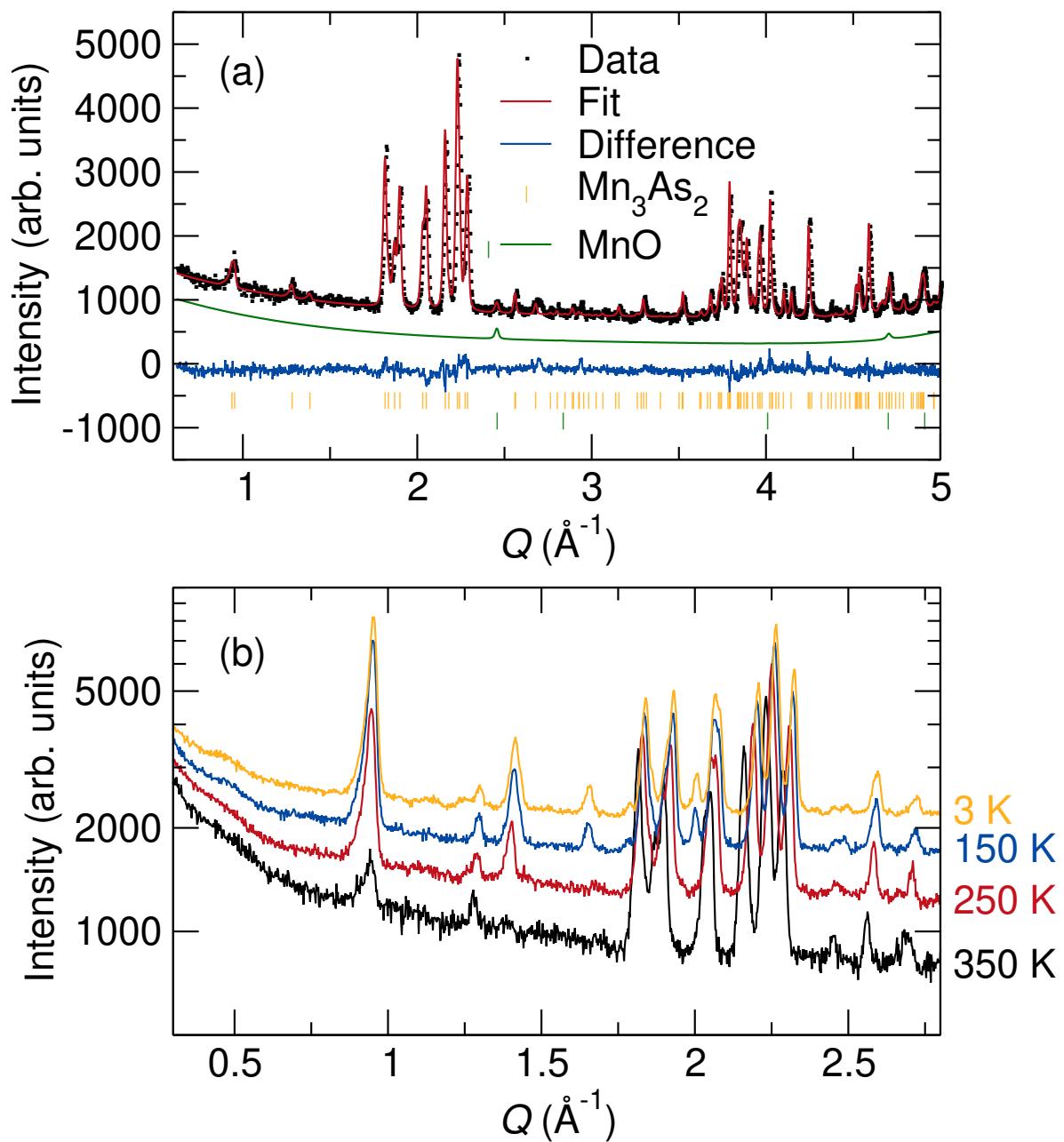


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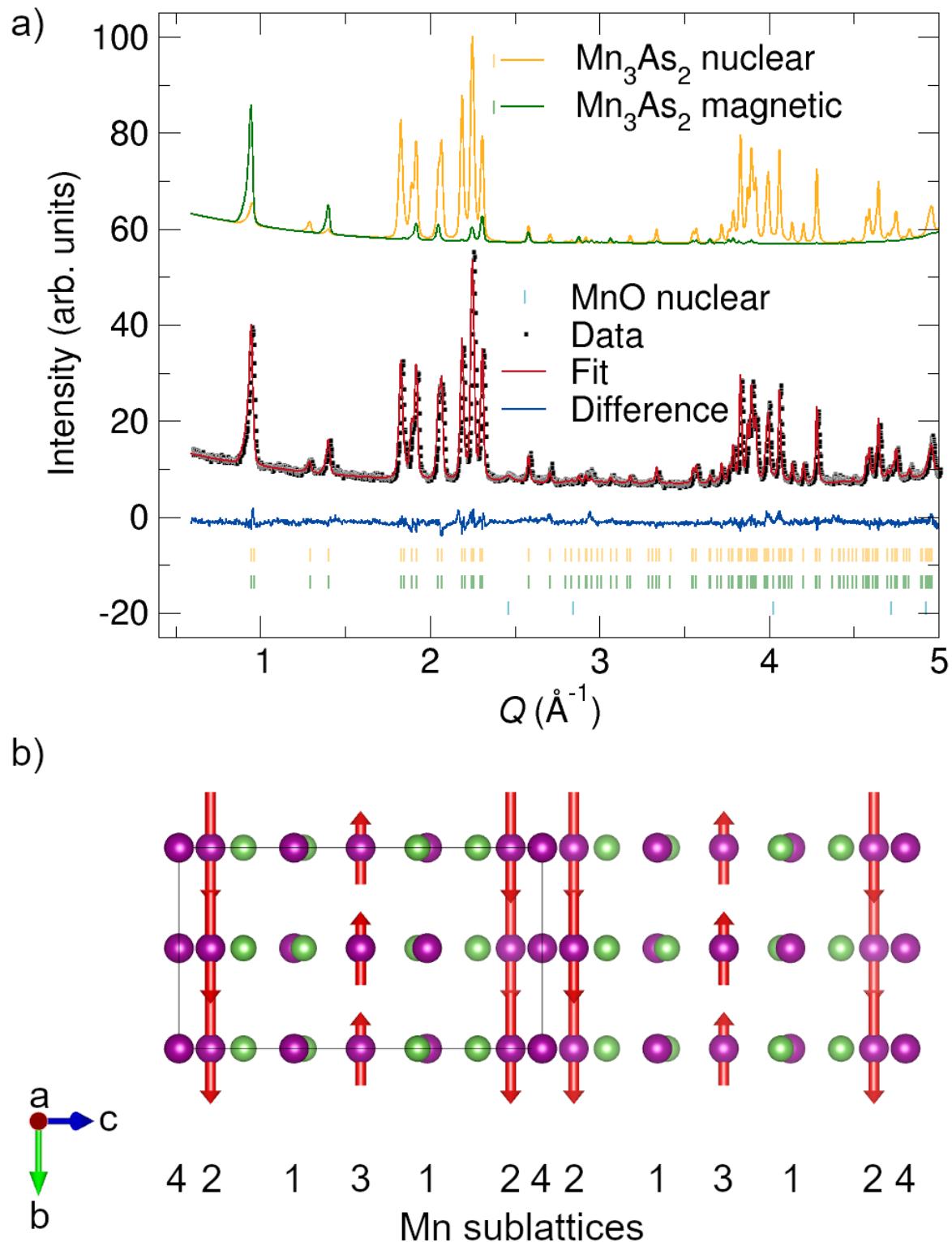


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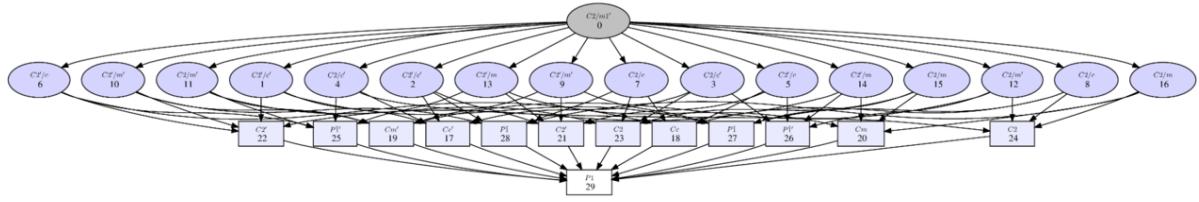


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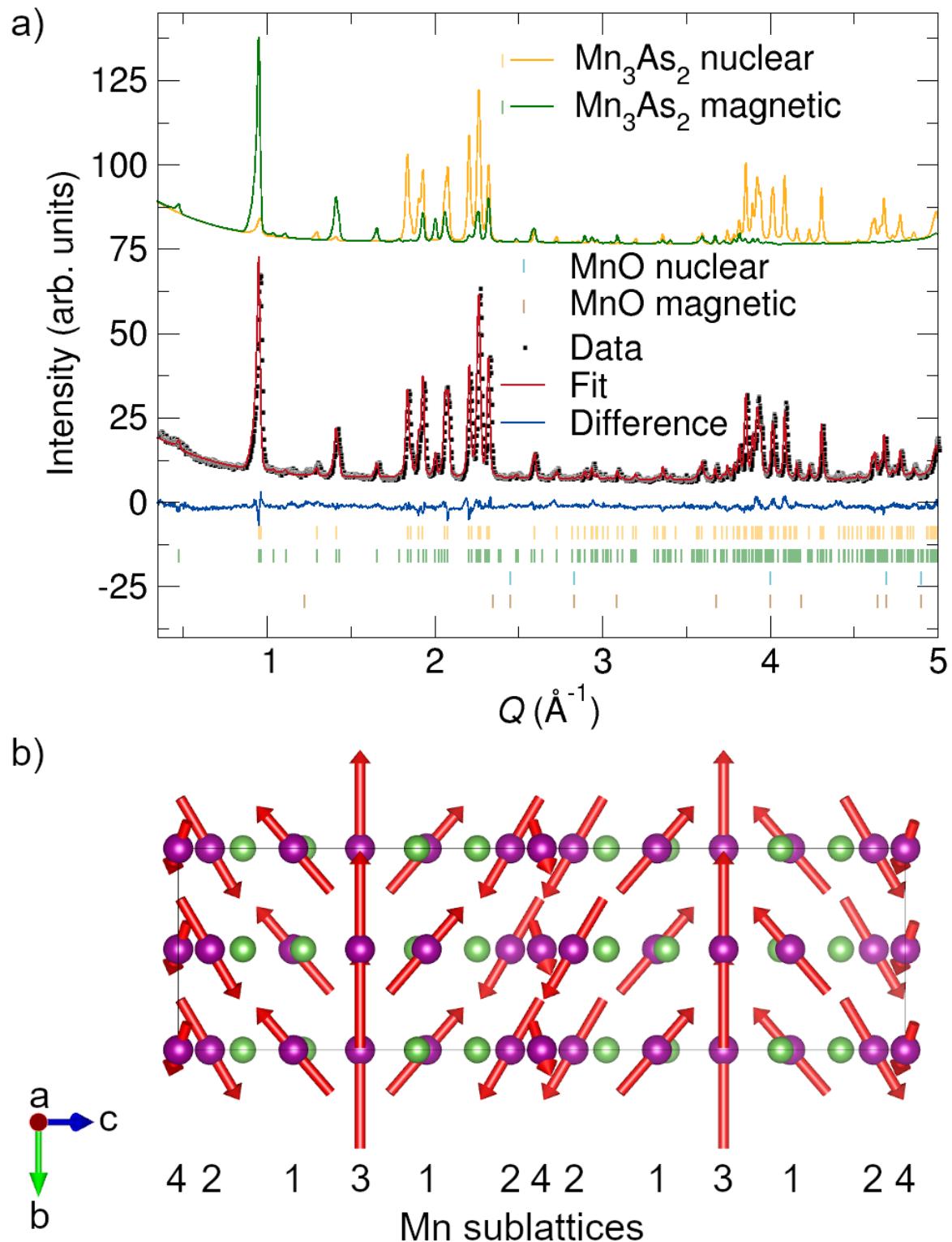


Figure 6.7: Electronic band structure

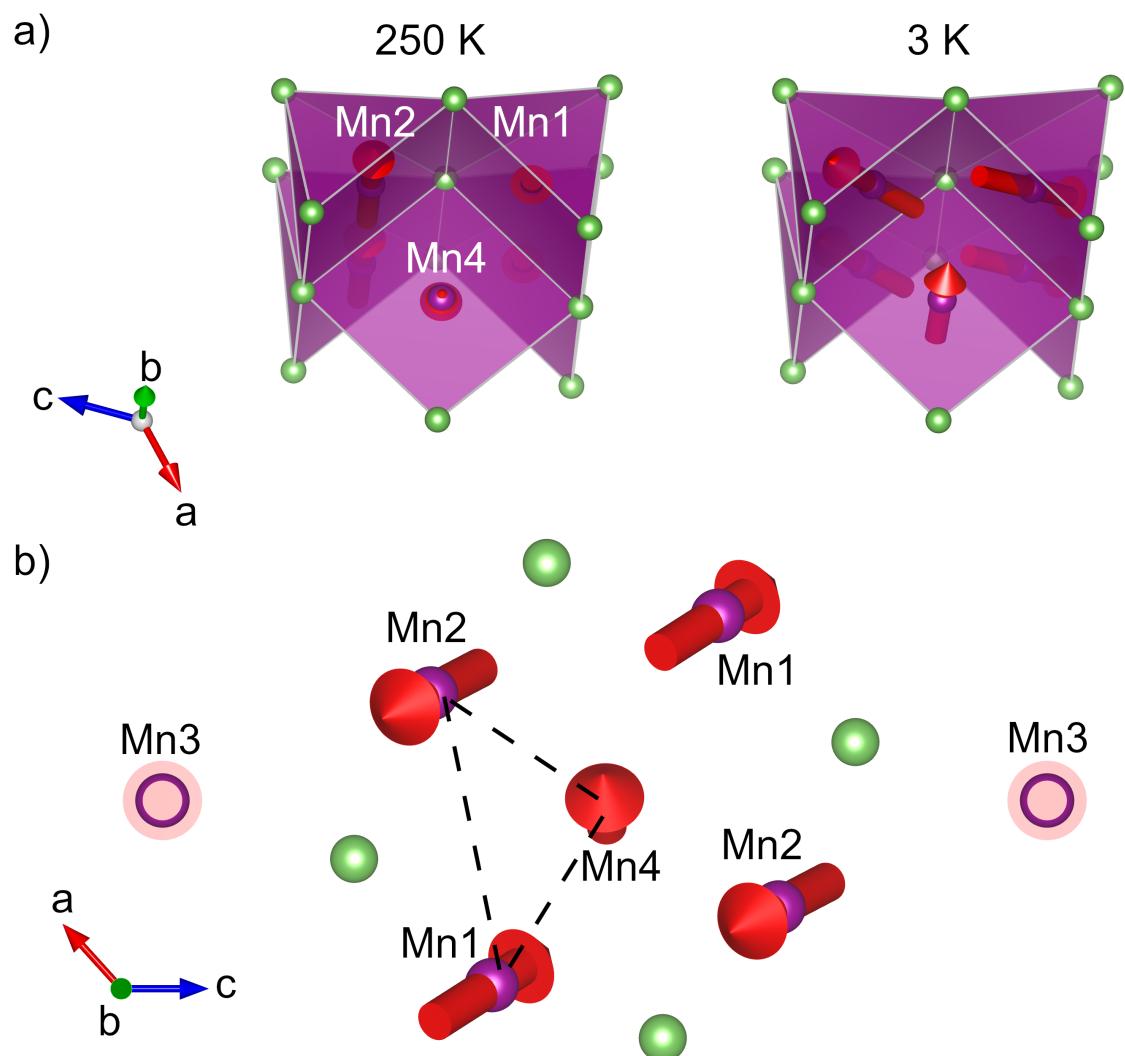


Figure 6.8: Electronic band structure

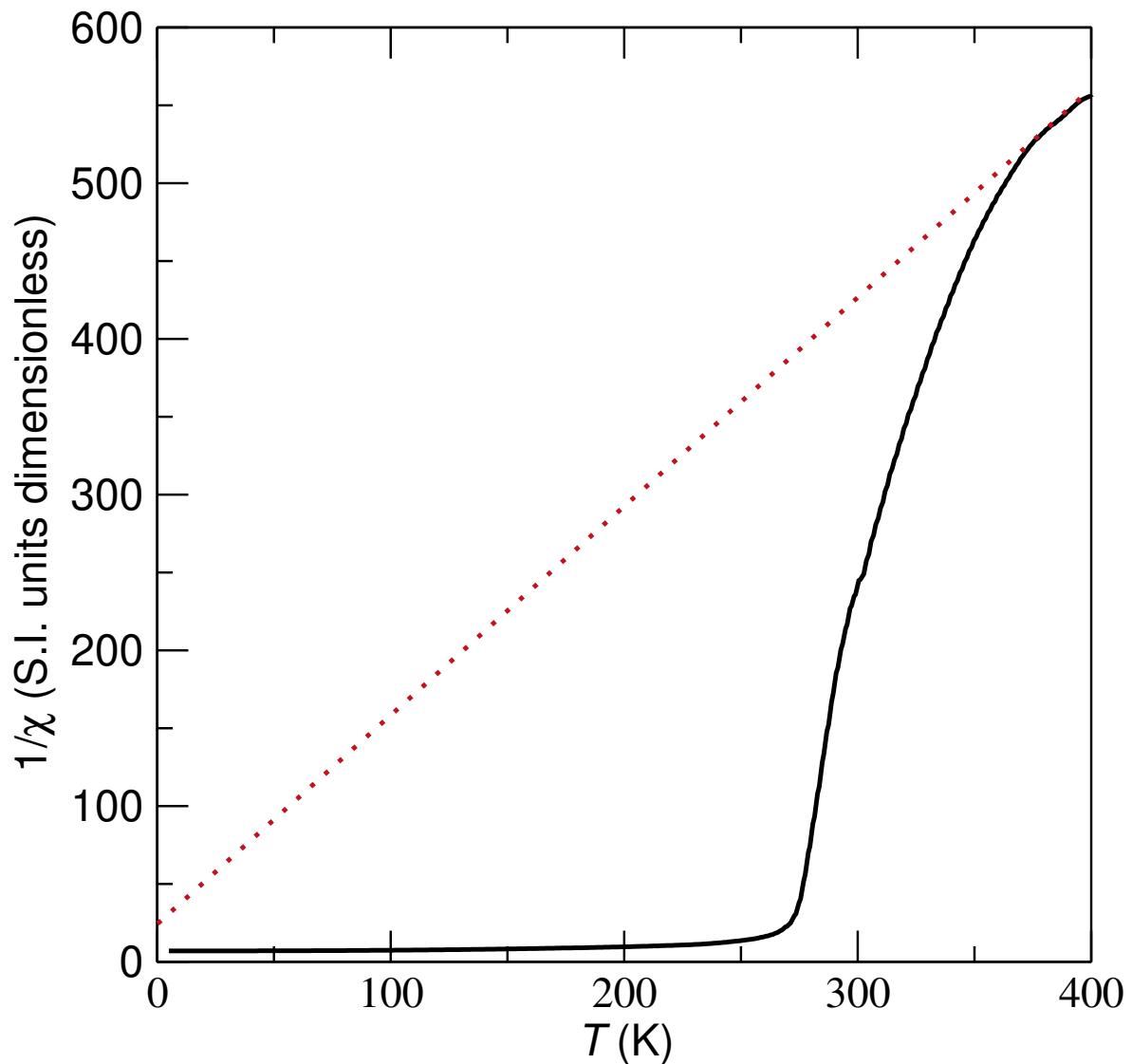


Figure 6.9: Electronic band structure

Chapter 7

Spin canting in tetragonal CuMnAs

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Chapter 8

Exchange interactions in Fe₂As probed by inelastic neutron scattering

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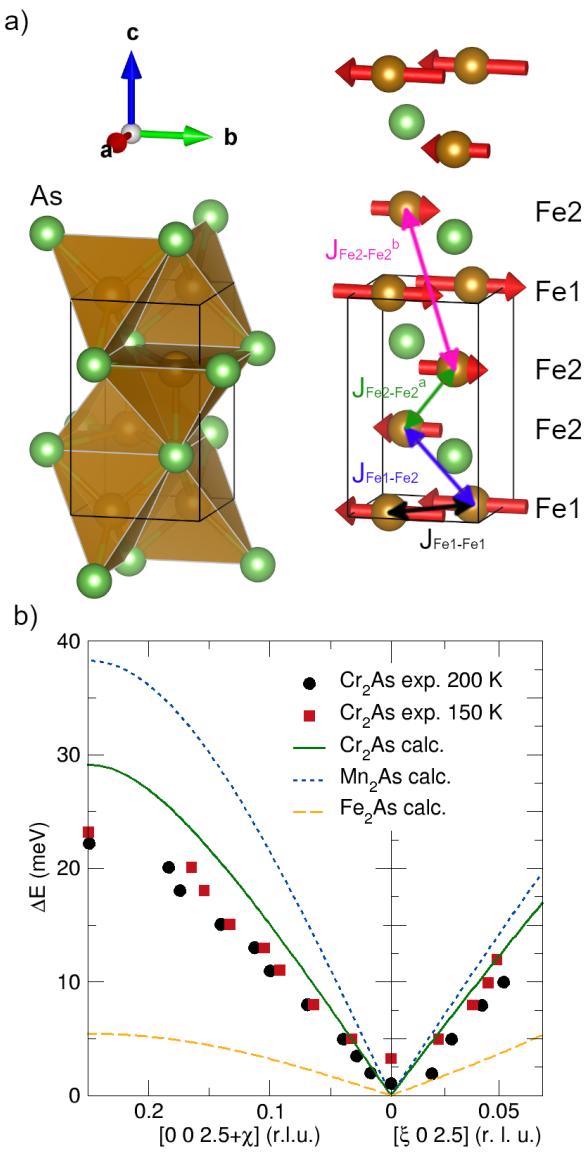


Figure 8.1: Electronic band structure



Figure 8.2: Electronic band structure

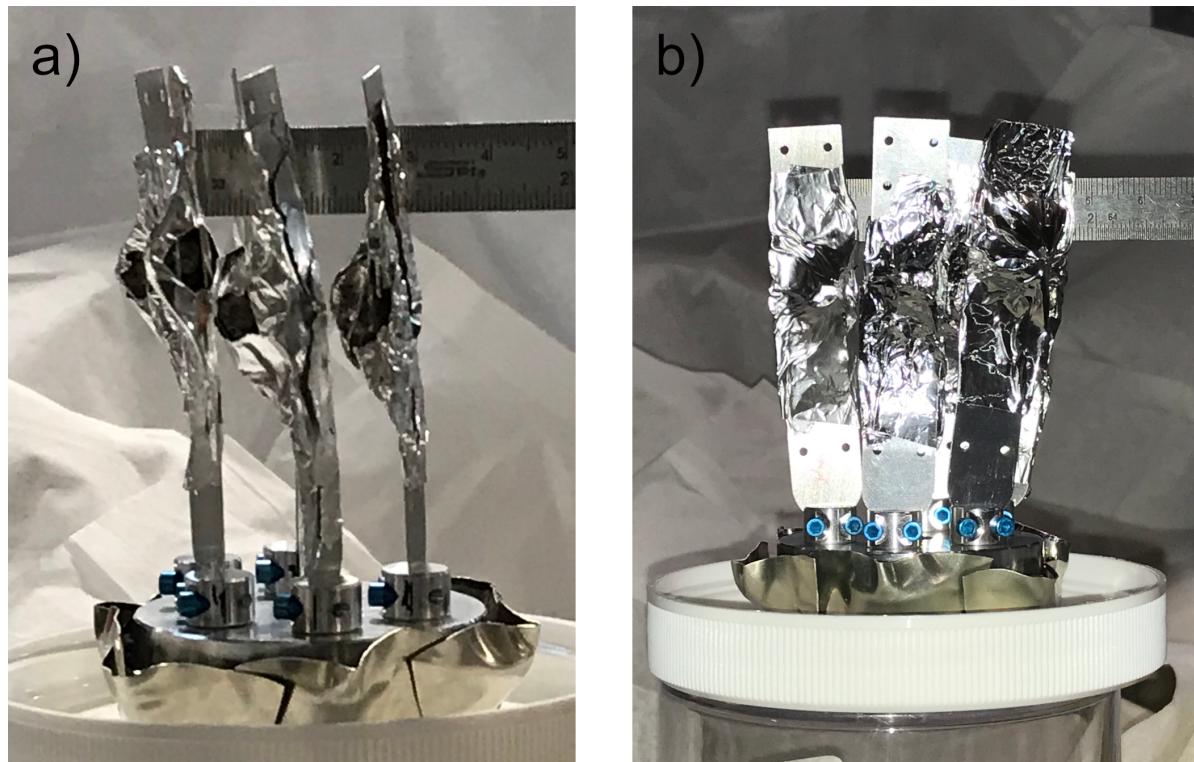


Figure 8.3: Electronic band structure

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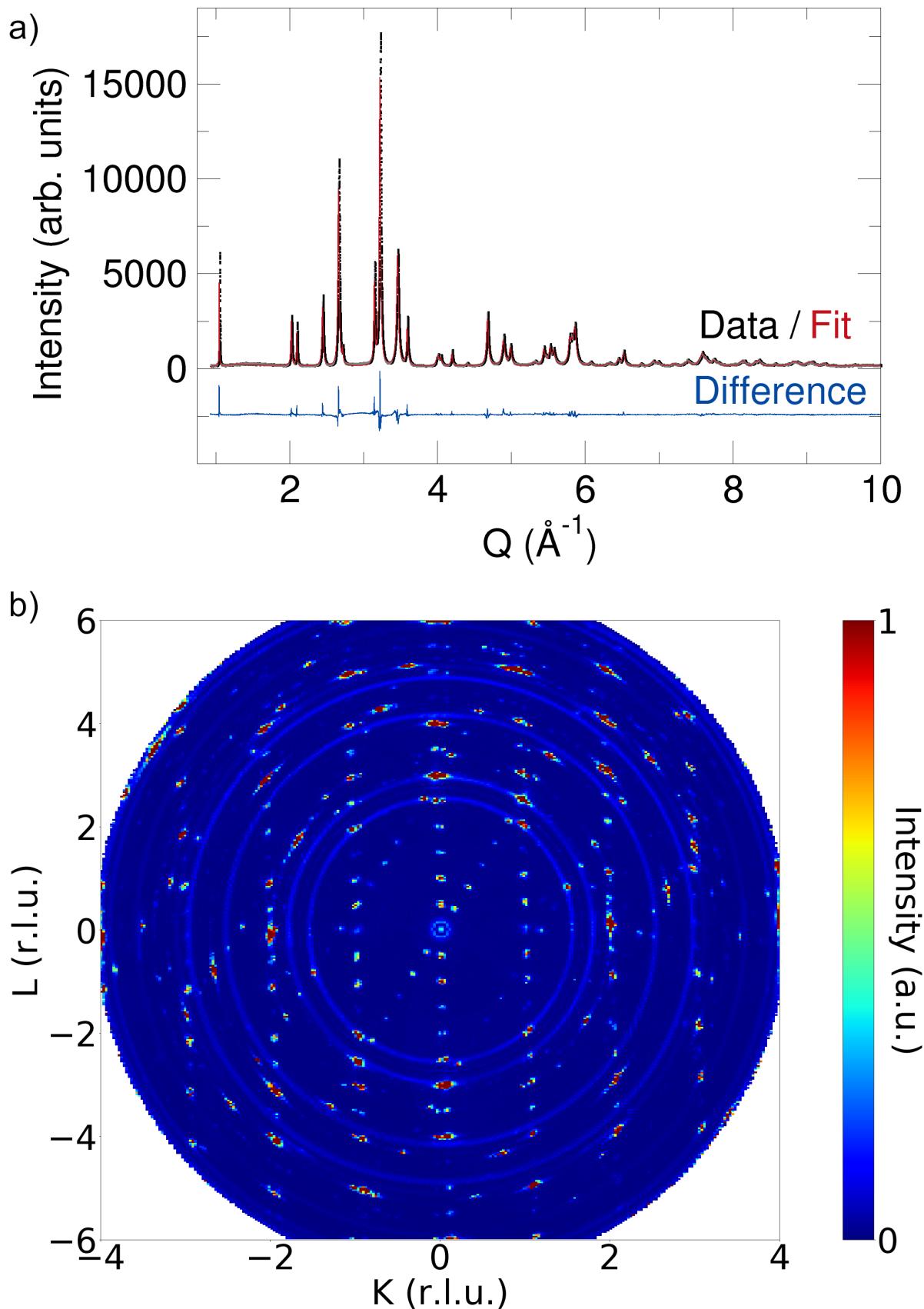


Figure 8.4: Electronic band structure

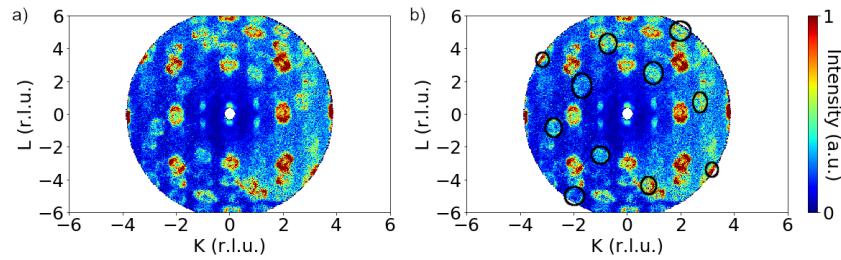


Figure 8.5: Electronic band structure

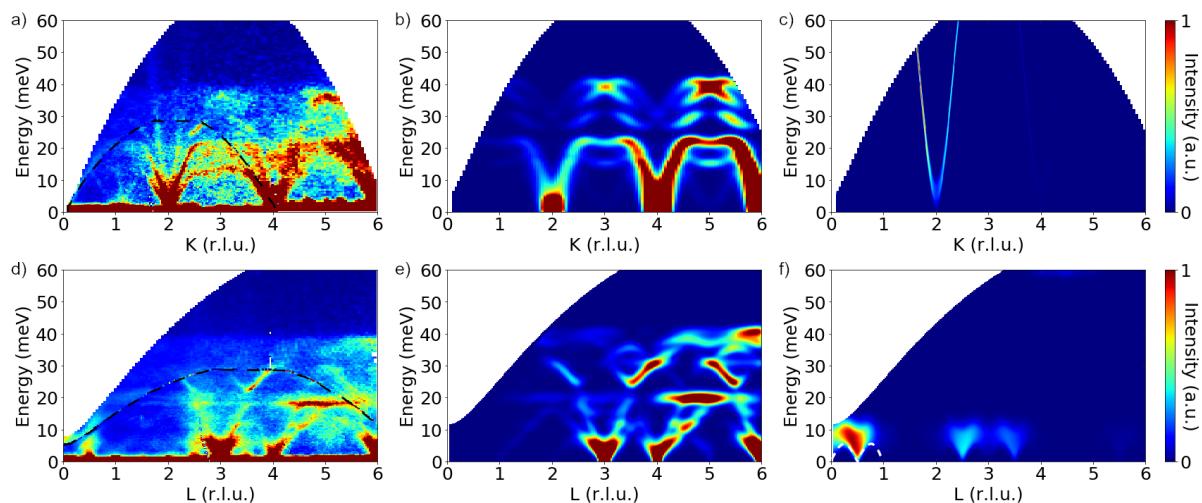


Figure 8.6: Electronic band structure

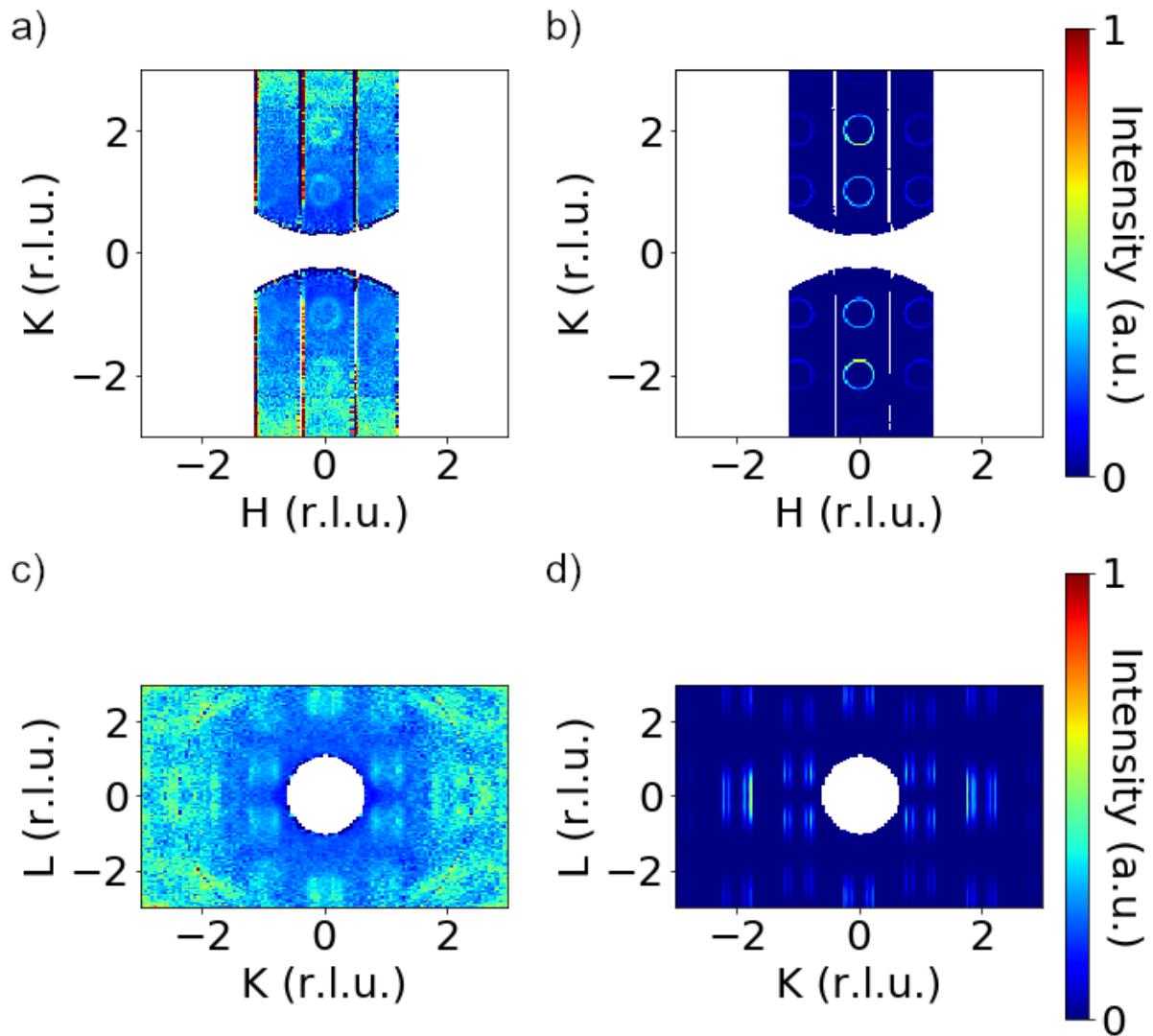


Figure 8.7: Electronic band structure

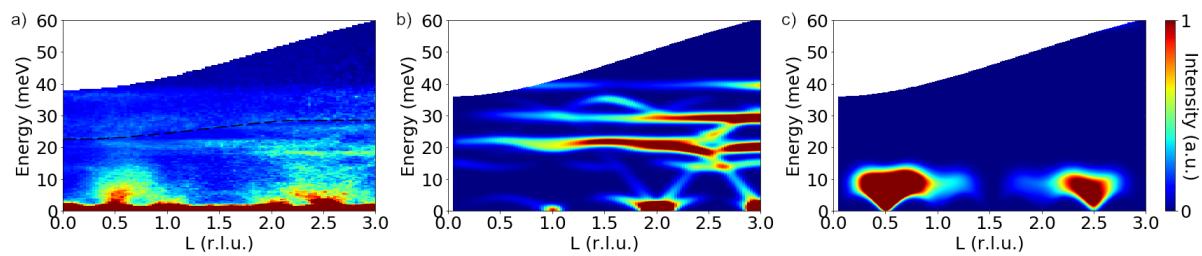


Figure 8.8: Electronic band structure

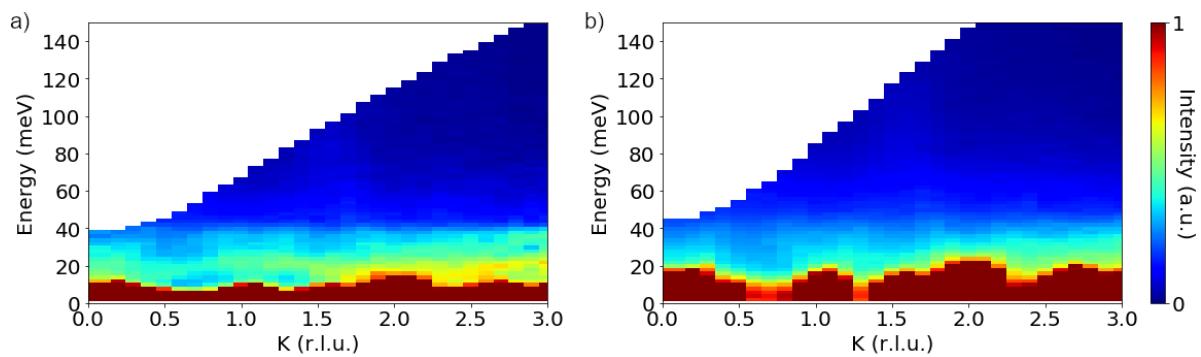


Figure 8.9: Electronic band structure

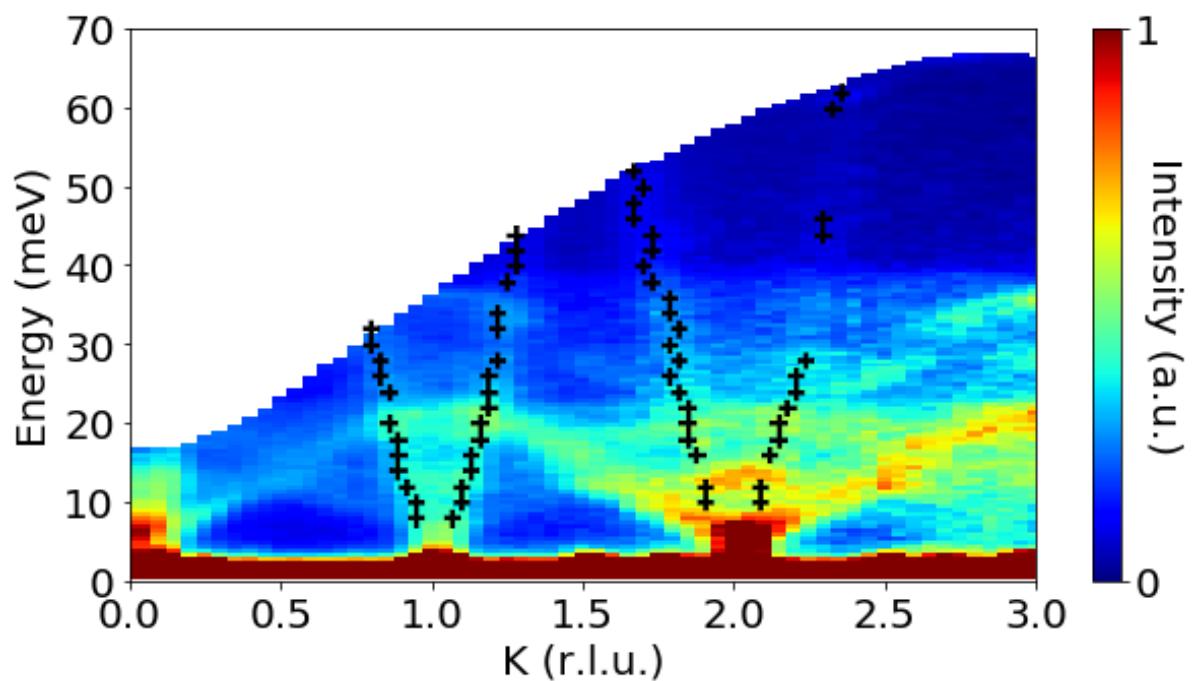


Figure 8.10: Electronic band structure

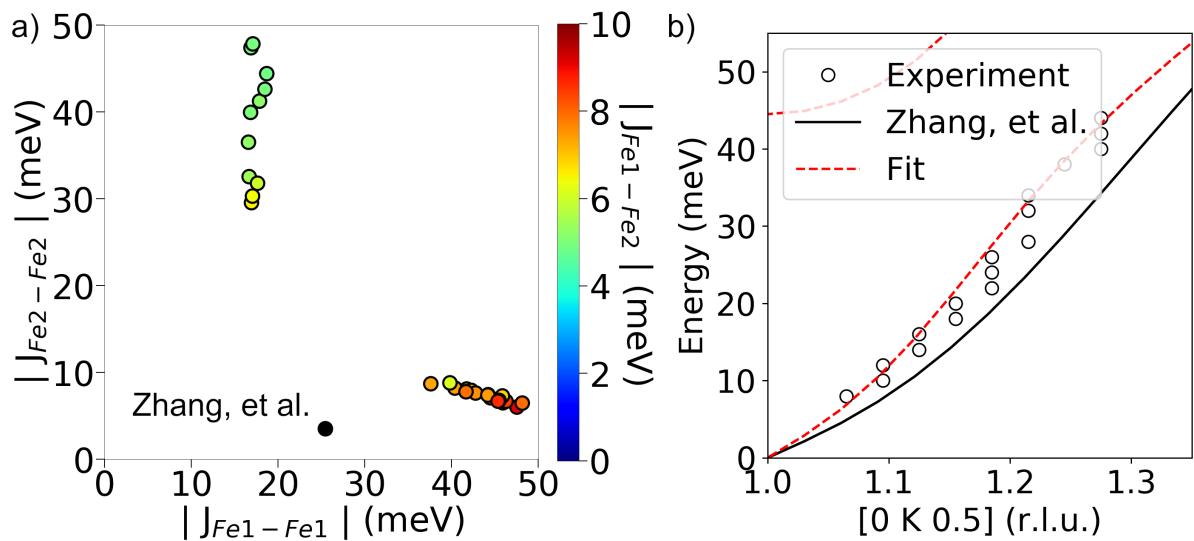


Figure 8.11: Electronic band structure

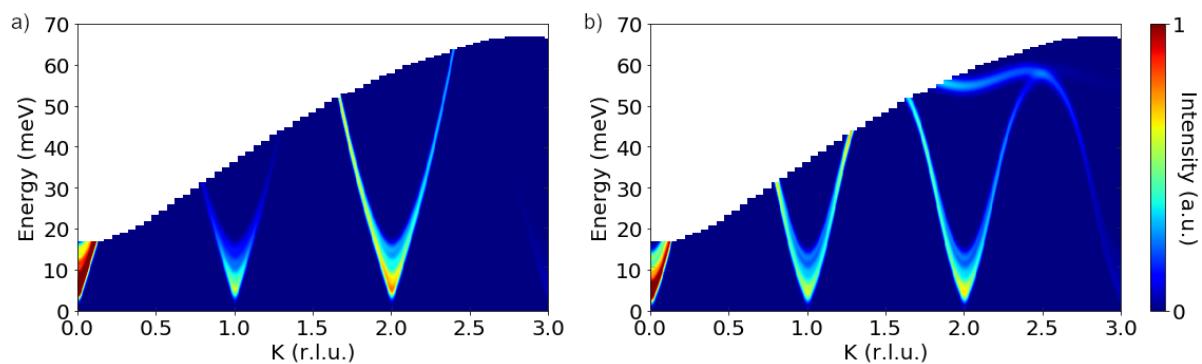


Figure 8.12: Electronic band structure

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Chapter 9

Conclusions

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