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1992 J. Phys.: Condens. Matter 4 L33

(http://iopscience.iop.org/0953-8984/4/1/008)

J. Phys.: Condens. Matter 4 (1992) L33–L36. Printed in the UK

LETTER TO THE EDITOR

Electronic band structure in the ferrimagnetic state of Mn_2Sb

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Received 10 October 1991

Abstract. The electronic band structure for the ferrimagnetic state of Mn_2Sb has been calculated using the augmented plane-wave (APW) method. The energy splitting between the spin-up and spin-down bands cannot be described by a rigid splitting of the non-magnetic band. It has been found that the width of 3d band of Mn(II) atoms is narrower than that of Mn(I) atoms and the hybridization between the 3d states of Mn(I) and Mn(II) is relatively weak compared with that in the non-magnetic state. The calculated magnetic moments are in good agreement with the observed values. The density of states for the ferrimagnetic state of Mn_2Sb as well as that for the ferromagnetic state of MnAlGe are compared with recent photoemission measurements.

Recently, Cu_2Sb-type intermetallic compounds have attracted much attention due to their rich variety of magnetic structures (Adachi and Ogawa 1988). In the Cu_2Sb-type crystal structure, the unit cell contains four metallic atoms, 1, 2, 3 and 4, and two anions, 5 and 6 (see figure 1(*a*)). Due to the difference of site symmetry, two kinds of metallic atom sites are distinguished from each other; they are usually called M(I) and M(II) sites.

Of the various kinds of Cu_2Sb-type compounds, such as Mn_2As, Cr_2As, Fe_2As etc, Mn_2Sb becomes a ferrimagnet below $T_N = 550$ K. The magnetic moment of the Mn(II)-site is antiparallel to that of the Mn(I)-site and the value of the former is much larger than that of the latter. The magnetic unit cell is the same as the crystallographic unit cell shown in figure 1(*a*).

Previously, we have performed the band-structure calculation by using a self-consistent augmented plane wave (APW) method for non-magnetic states of the Cu_2Sb-type compounds, Cr_2As, CrMnAs, Mn_2As, MnFeAs, Fe_2As and Mn_2Sb (Chônan *et al* 1991). For all of these compounds, it was found that the itinerant character of 3d electrons of M(II) atoms is weaker than that of M(I) atoms, because the width of the mixed d bands arising from d orbitals of M(II) atoms is narrow compared with that arising from d orbitals of M(I) atoms. This is consistent with the observed result that the magnetic moments of the M(II) atoms are large compared with those of the M(I) atoms. For quantitative argument, however, it is necessary to study the electronic band structures in the magnetic ordered state.

In this letter, we have performed the band calculation for the ferrimagnetic state of Mn_2Sb by using the self-consistent APW method. We adopt the muffin-tin approximation

0953-8984/92/010033 + 04 $03.50

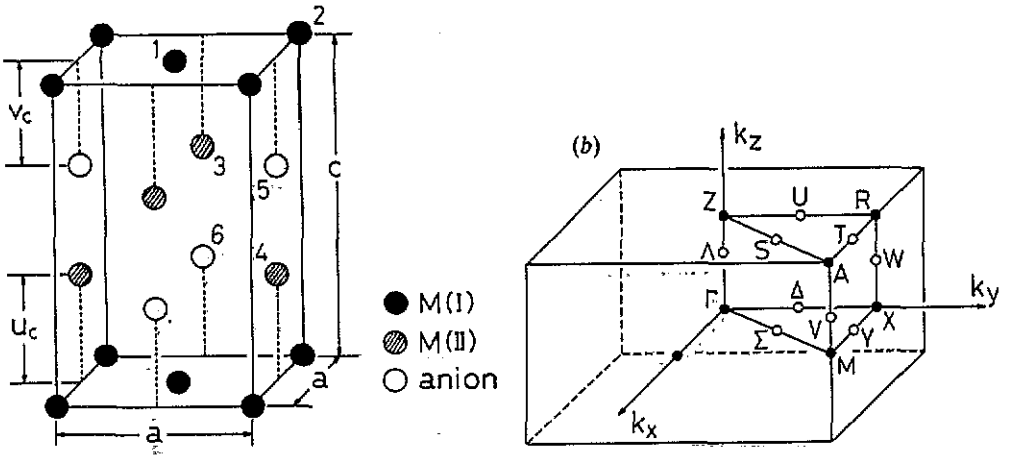

Figure 1. (*a*) The crystal structure and (*b*) the first Brillouin zone for the Cu_2Sb-type intermetallic compounds.

Table 1. The lattice parameters of Mn_2Sb (Heaton and Gingrich 1955) and the muffin-tin radii of Mn(I), Mn(II) and Sb spheres used in the present calculation.

Lattice parameters				Muffin-tin radii		
a	*c*	u_c	v_c	Mn(I)	Mn(II)	Sb
4.078 Å	6.557 Å	0.295*c*	0.280*c*	0.3535*a*	0.3200*a*	0.3193*a*

to the crystal potential. The local density approximation is used to construct the exchange–correlation terms of the one-electron potential (Gunnarsson and Lundqvist 1976). We use the criteria that $l_{max} = 7$ and $|k + G|_{max} = (2\pi/a) \times 3.8$, with k being a wavevector in the first Brillouin zone of figure 1(*b*), and G a reciprocal lattice vector. The latter criterion yields a set of about 360 basis functions. We have determined self-consistently the charge density of the crystal using a set of four special k points in the iteration process. The starting configurations of valence electrons for Mn(I) and Mn(II) atoms are taken to be different for spin-up and spin-down states as follows: $(3d)^{3.5}\,(4s)^1$ and $(3d)^{1.5}\,(4s)^1$ for spin-up and spin-down states of Mn(I); $(3d)^{0.5}\,(4s)^1$ and $(3d)^{4.5}\,(4s)^1$ for spin-up and spin-down states of Mn(II), while those for Sb atoms are taken to be the same for both spin-up and spin-down states, i.e. $(5s)^1\,(5p)^{1.5}$. The core electrons are assumed to be frozen. We have calculated the energy band within an accuracy of 1 mRyd. In table 1, the lattice parameters and the muffin-tin radii of Mn(I), Mn(II) and Sb spheres used in the present calculation are listed. The density of states (DOS) has been calculated with the linearly energy-interpolated tetrahedron method (Lehmann and Taut 1972) using the energy eigenvalues at 60 points in the 1/16 irreducible Brillouin zone.

The DOS for the spin-up and spin-down bands are shown in figure 2. Contributions arising from the Mn(I) 3d, Mn(II) 3d and Sb 5p states, inside each muffin-tin sphere, are also shown separately in figure 2. Comparing the DOS for the ferrimagnetic state with that for the non-magnetic state shown in our previous paper (Chônan *et al* 1991), we

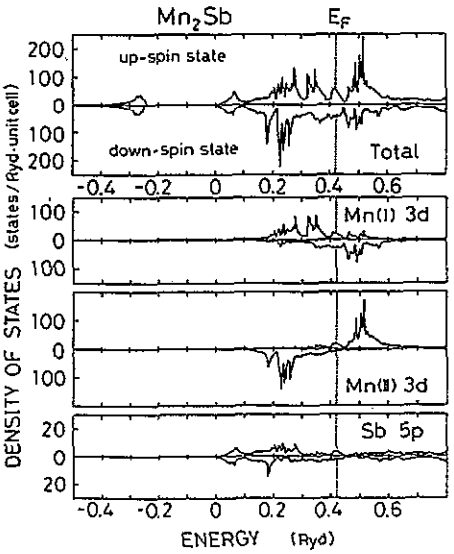

Figure 2. The electronic density of states (DOS) of Mn_2Sb in the ferrimagnetic state. Contributions arising from the Mn(I) 3d, Mn(II) 3d and Sb 5p states, inside each muffin-tin sphere, are also shown separately.

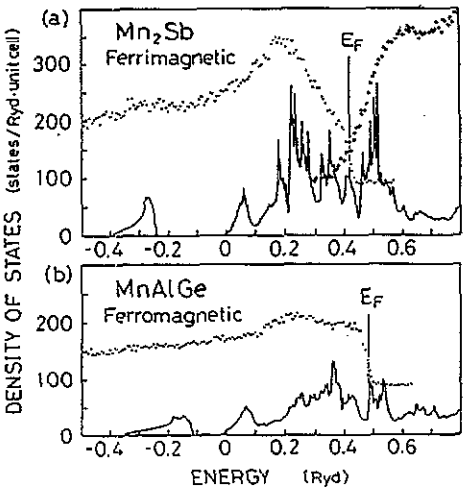

Figure 3. The total DOS for (*a*) Mn_2Sb and (*b*) MnAlGe (Motizuki *et al* 1991) obtained by the band calculation (full curves). The photoemission (dots) and inverse photoemission (crosses) spectra are also shown for comparison (Kimura *et al* 1991).

have found that the energy splitting between the spin-up and spin-down bands cannot be described by a rigid splitting of the non-magnetic band. In the ferrimagnetic state, the hybridization between the 3d states of Mn(I) and Mn(II) becomes considerably weak compared with that in the non-magnetic state. Due to this effect, the itinerant character of 3d electrons of Mn(II) atoms is weakened further in the ferrimagnetic state.

The energy splitting of the spin-up and spin-down bands of the Mn(II) is much larger than that of the Mn(I) as seen in figure 2. The energy splitting can be roughly estimated by evaluating the difference between the centre of gravities for the DOS of both spin states. They have been estimated to be 0.11 and 0.23 Ryd for Mn(I) and Mn(II), respectively. This difference between the energy splittings is directly reflected in the difference in the magnitude of magnetic moments between Mn(I) and Mn(II). The total magnetic moment and the magnetic moments inside the muffin-tin spheres at the Mn(I), Mn(II) and Sb sites are obtained as shown in table 2. The observed values (Wilkinson *et al* 1957) are also listed in table 2. The agreement between the calculated and the observed values are extremely good.

Recently, photoemission and inverse photoemission measurements have been performed for Mn_2Sb and photoemission measurement for MnAlGe (Kimura *et al* 1991). The observed spectra are shown in figure 3. The most prominent feature of the observed spectra of Mn_2Sb is the presence of a broad peak structure at about 0.2 Ryd below the Fermi level (E_F) and a peak structure at about 0.15 Ryd above E_F. On the other hand, in MnAlGe, no prominent peak structure is found below E_F, but the presence of valence band states with a width of about 0.4 Ryd is appreciable below E_F. To compare the observed spectra with the results of our band calculation, we have plotted the sum of

Table 2. The total magnetic moment and the magnetic moments inside the muffin-tin spheres at Mn(I), Mn(II) and Sb sites. The observed values are also listed (Wilkinson *et al* 1957). The unit of magnetic moments is μ_B.

Magnetic moments	Total	Mn(I)	Mn(II)	Sb
Calculated	1.76	−2.11	3.65	0.01
Observed	1.74	−2.13	3.87	—

the DOS of spin-up and spin-down bands for Mn_2Sb and MnAlGe (Motizuki *et al* 1991) in figures 3(*a*) and (*b*), respectively. The characteristic feature of the observed spectra is found to be in good correspondence with the calculated total DOS for both compounds. In particular, the prominent peak structures observed in Mn_2Sb can be attributed to the narrow 3d bands of the Mn(II) atoms. On the other hand, the absence of the peak structure in the photoemission spectra for MnAlGe is due to the fact that the DOS near the E_F arises from only the broad 3d bands of the Mn(I) atoms (Motizuki *et al* 1991). In fact, as for MnAlGe, Mn atoms occupy only the M(I) sites.

In conclusion, the results of the electron-band calculation for the ferrimagnetic state of Mn_2Sb, such as the magnitude of magnetic moments and the overall aspect of the DOS, are in good agreement with the experimental results. For more detailed comparison, further experimental studies are desirous.

This work is supported by Grant-in-Aid for Scientific Research from the Ministry of Education, Science and Culture, Japan. We are grateful to Mr A Kimura and Professor S Suga for useful discussion about their experimental results. Thanks are also due to Professor A Yanase for providing us with a computer program developed by him.

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