Proceedings of the European Conference "Physics of Magnetism '99", Poznań 1999

THE ELECTRONIC STRUCTURE OF THE RMn_2Ge_2 (R = Ca, Y, La, Ba) ANTIFERROMAGNETS

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The electronic structure of the tetragonal RMn_2Ge_2 (R = Ca, Y, La, Ba) antiferromagnets is presented using the self-consistent Korringa–Kohn–Rostoker method. According to the neutron refinements, two types of collinear antiferromagnetic structure are taken into account: AF_1 for YMn_2Ge_2 and AF_2 for the other compounds. The calculated magnetic moments on Mn: 2.17 μ_B (YMn_2Ge_2), 2.84 μ_B ($CaMn_2Ge_2$), 2.95 μ_B ($LaMn_2Ge_2$), and 3.47 μ_B ($BaMn_2Ge_2$) remain in good agreement with the neutron data (in μ_B) 2.20, 2.67, 3.05, and 3.66, respectively. As seen on antiferromagnetic density of states, all systems are metallic, however $BaMn_2Ge_2$ is found near semimetallic limit. The total energy Korringa–Kohn–Rostoker computations on $CaMn_2Ge_2$, performed in both antiferromagnetic phases, result in preferring of the AF_2 structure.

PACS numbers: 71.15.Mb, 71.15.Nc, 75.20.En, 75.20.Hr, 75.50.Ee

1. Introduction

The magnetic properties of the well-known ternary RMn_2Ge_2 compounds [1] with the tetragonal $ThCr_2Si_2$-type structure have attracted renewed interest in recent years. Detailed neutron diffraction investigations of many RMn_2Ge_2 have revised previously detected ferromagnetic (F) structures (R = La–Nd) [2, 3] and explored the magnetic behavior of new compounds ($CaMn_2Ge_2$ and $BaMn_2Ge_2$ [4]). In $LaMn_2Ge_2$, antiferromagnetic ordering (AF_2) occurs below $T_N \approx 400$ K whereas rather canted magnetic structure than c-axis collinear ferromagnetism appears below T_C, with the antiferromagnetic component markedly larger ($\mu_{AF} = 2.7\ \mu_B$) than the ferromagnetic one ($\mu_F = 1.5\ \mu_B$). These results have been later supported by the Mössbauer effect measurements [5]. The three other entitled compounds show purely AF ordering of the Mn magnetic moments. There is a critical distance between Mn atoms in the a–b plane ($d_{in} = a/\sqrt{2} \approx 2.86$ Å), where the magnetic properties change drastically (mostly due to the AF_1–AF_2 transition).

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Thus, in YMn_2Ge_2 (d_{in} = 2.82 Å) μ_{Mn} are arranged parallel in the Mn plane and coupled AF between the planes (AF_1-type) [6], while in $CaMn_2Ge_2$ (d_{in} = 2.94 Å) and $BaMn_2Ge_2$ (d_{in} = 3.14 Å) the neighboring μ_{Mn} are aligned antiparallel both in and between the manganese planes (AF_2-type) [4]. Ishida et al. [7] have calculated the electronic structure of the "ferromagnetic" $LaMn_2Ge_2$ by the Korringa–Kohn–Rostoker (KKR) method. However, the theoretical values of μ_{tot} and μ_{Mn} have been found rather in disagreement with experimental data available at this time. Furthermore, Kulatov et al. [8] have discussed magnetic ordering of RMn_2Si_2 and magnetic transition near $d_{in} \approx 2.86$ Å, using the non-polarized linear muffin-tin orbital (LMTO) results.

The purpose of this paper is to show the electronic structure and magnetism of RMn_2Ge_2 using experimentally detected magnetic ordering of μ_{Mn}.

2. Computational details

The spin-polarized calculations on the tetragonal body-centered (bct) RMn_2Ge_2 compounds are performed by the spin and charge self-consistent KKR method [9, 10] within the local spin density framework (the exchange-correlation potential given by the von Barth–Hedin formula [11]). In the RMn_2Ge_2 structure ($I4/mmm$) R, Mn, and Ge atoms occupy 2(a) [0,0,0], 4(d) [0,1/2,1/4] and 4(e) [0,0,z] Wyckoff sites, respectively. The crystallographic data measured at T = 2 K [2–4] and muffin-tin radii $r_R : r_{Mn} : r_{Ge} \approx 1.5 : 1 : 1$ (giving $\sum V^i_{mt}/V_{WS} \approx 67\%$) are used in the KKR computations. For the final potentials ($\Delta E_{tot} \approx 1$ mRy) the total densities of states (DOS), site-decomposed DOS and l-decomposed DOS (with $l_{max} = 2$ for R = Ca, Y, Ba and $l_{max} = 3$ for R = La) are computed. The relativistic effects are incorporated in calculations of core levels in $BaMn_2Ge_2$ and $LaMn_2Ge_2$. Note that in YMn_2Ge_2 (AF_1), no more bct symmetry is maintained, then the KKR calculations are performed within the simple tetragonal structure. Integration in the k-space is performed using 192 small tetrahedrals and 135 k-points in the irreducible part of the Brillouin zone.

3. Results and discussion

The main KKR results for RMn_2Ge_2, calculated both in F and AF states, are summarised in Table. The theoretical values of μ_{Mn} remain in close agreement with the neutron diffraction data, recently measured at T = 2 K [2–4]. The magnetisation (4.22 μ_B) as well as the Mn magnetic moments (2.29 μ_B) found in the ferromagnetic $LaMn_2Ge_2$ are also close to the earlier theoretical values (3.9 μ_B and 2.1 μ_B) [7]. From comparison of the F and AF results one notes that μ_{Mn} in the AF_2-type compounds are rather larger than these computed in the F state (particularly in $LaMn_2Ge_2$), while they are almost the same in YMn_2Ge_2 (AF_1-type ordering).

If plotting the theoretical μ_{Mn} versus d_{in} in the RMn_2Ge_2 antiferromagnets (Fig. 1) we observe that the Mn magnetic moment increases more or less proportionally to d_{in}, which well corresponds to the phenomenological relations established among d_{in}, μ_{Mn} and magnetic ordering type [12]. Nevertheless, one should take care using this simple model (Fig. 1), since in real samples when lattice constants increase, z_{Ge} also changes. A number of the KKR computations on

the RMn_2X_2 systems, carried out with the use of different a, c and z_X parameters, lead to conclusion that the z_X value (like d_{in}) has a strong influence on a magnitude of μ_{Mn} in RMn_2X_2. Moreover, both z_X and d_{in} seem crucial in appearing of magnetic properties in RT_2X_2 (with T $\neq$ Mn) [13].

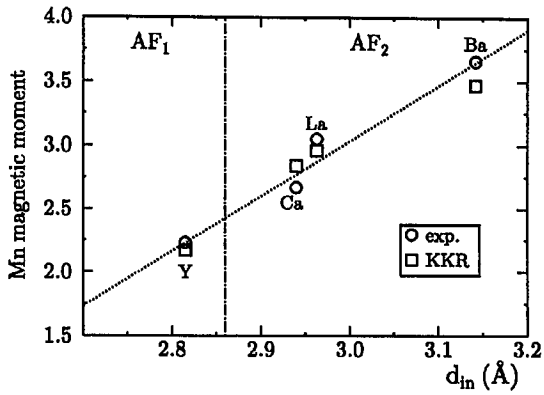

Fig. 1. The Mn magnetic moment (in μ_B) versus the Mn–Mn distance (d_{in}) in RMn_2Ge_2.

TABLE

The KKR results in RMn_2Ge_2 (magnetic moments in μ_B, DOS in states/Ry/spin).

	R = Y	R = Ca	R = La	R = Ba
F state				
μ_{tot}	4.02	4.99	4.22	6.78
μ_{Mn}	2.15	2.59	2.29	3.33
$n_\uparrow(E_F)$	18.79	18.69	20.16	27.55
$n_\downarrow(E_F)$	32.80	51.42	65.52	41.14
AF state				
KKR	2.17	2.84	2.95	3.48
exp.	2.20	2.67	3.05	3.66
$n_{\uparrow\downarrow}(E_F)$	47.51*	10.96	48.99	3.06

*per two formula units

The DOS of YMn_2Ge_2 (AF_1) (Fig. 2) consists of two broad peaks coming mostly from the Mn sites. Unlikely, the DOS of $CaMn_2Ge_2$, $LaMn_2Ge_2$, and $BaMn_2Ge_2$ (AF_2) presents complex structure with few narrow d-like peaks arising also on Mn atoms (note that the highest d-like peaks are localized well below E_F in contrast to d-DOS in YMn_2Ge_2). The conduction band in RMn_2Ge_2 is formed mostly by d-states on Mn and R with some admixture of p-states on Ge.

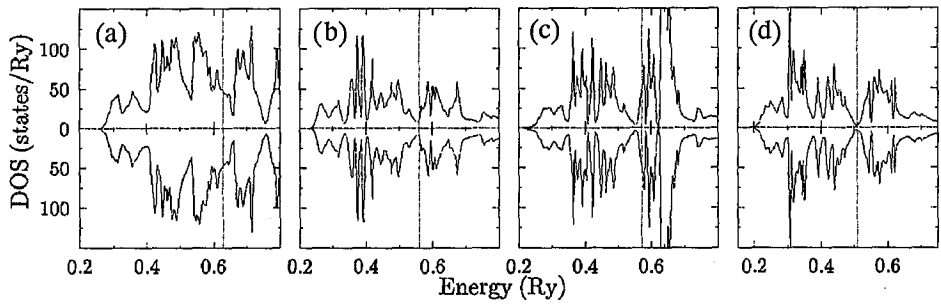

Fig. 2. The total DOS in the RMn_2Ge_2 antiferromagnets: (a) YMn_2Ge_2, (b) $CaMn_2Ge_2$, (c) $LaMn_2Ge_2$, (d) $BaMn_2Ge_2$.

The s-states (not plotted in Fig. 2) form two separate bands and are located 0.13–0.15 Ry below the conduction band bottom. When passing from R = Y to R = Ba (increasing d_{in}), less overlapping of d-like wave functions occurs, giving rise to narrow bands. Consequently, Mn atoms keeps successively larger magnetic moment reaching the 3.48 μ_B value (3.66 μ_B from experiment) in $BaMn_2Ge_2$. Interestingly, from the KKR calculations this compound is found near the semimetallic limit (Fig. 2). The $E(k)$ computations (not shown) result in two strongly dispersive bands crossing the Fermi level, while an energy gap occurs along most of k-vector directions. As $CaMn_2Ge_2$ and $BaMn_2Ge_2$ have the same number of valence electrons, the low DOS at E_F is also detected in $CaMn_2Ge_2$. In $LaMn_2Ge_2$ E_F is shifted into the DOS peak due to one more electron. To investigate a reason for the changing of the AF-type structure, near $d_{in} \approx 2.86$ Å the total energy KKR computations are done on $CaMn_2Ge_2$ applying AF_1 and AF_2. Indeed, E_{tot} has a slightly lower value when using the AF_2 structure. But the similar comparison in YMn_2Ge_2 does not give reliable preference for AF_1. Then, more detailed analysis $E_{tot} = f(a, c, z_{Ge})$ should be undertaken to clarify this point.

Acknowledgments

This work has been realized during the fellowship granted to J.T. from the Henri Poincaré University in Nancy. The computations have been partly performed in the Supercomputing Center-Cyfronet in Kraków.

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