Effects of geometry in itinerant electron magnets

2007 J. Phys.: Condens. Matter 19 145285

(http://iopscience.iop.org/0953-8984/19/14/145285)

IOP PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **19** (2007) 145285 (6pp) doi:10.1088/0953-8984/19/14/145285

Effects of geometry in itinerant electron magnets

H Nakamura[1], Y Muro[1], T Kohara[1] and M Shiga[2]

[1] Graduate School of Material Science, University of Hyogo, Kamigori, Ako-gun, Hyogo 678-1297, Japan
[2] Department of Materials Science and Engineering, Kyoto University, Kyoto 606-8501, Japan

Received 16 October 2006
Published 23 March 2007
Online at stacks.iop.org/JPhysCM/19/145285

Abstract

The magnetism of quasi-one-dimensional itinerant electron magnets RMn_4Al_8 is compared with that of the typical frustrated itinerant electron magnet YMn_2. The possible formation and observation of the spin pseudogap are discussed in connection with the spin-liquid state in strongly correlated itinerant electron systems.

1. Introduction

The cubic Laves phase compound YMn_2 is a typical three-dimensional (3D) frustrated itinerant electron antiferromagnet with the Néel temperature $T_N \simeq 100$ K [1]. The spin-liquid or spin-singlet-like state in YMn_2 (at $T > T_N$) has been understood as a consequence of the frustration of the nearest-neighbour antiferromagnetic interaction in the pyrochlore lattice. Ballou *et al* [2] reported that the magnetic neutron scattering signal of $Y(Sc)Mn_2$ (the antiferromagnetic ordering in YMn_2 is suppressed by the chemical pressure) is broader in the [001] than in the [110] direction; the correlation length in the [001] direction is smaller than the interatomic distance. Such spatially anisotropic spin fluctuations have also been confirmed by nuclear magnetic relaxation measurements [3]. Noting that three different ⟨110⟩ atomic chains cross at each atomic site in the pyrochlore lattice (figure 1(a)), it is expected that the strong nearest-neighbour antiferromagnetic interactions in the ⟨110⟩ chains inevitably decouple the interaction between {001} planes. In other words, the spin-liquid state in YMn_2, as a nearest-neighbour antiferromagnetic pyrochlore lattice, is dynamically coupled quasi-one-dimensional (1D) antiferromagnetic chains in the 3D manner. This interpretation may be related to the spin–Peierls-type instability in the pyrochlore lattice [4]. Below the Néel temperature, a small tetragonal distortion lifts the degeneracies of three ⟨110⟩ axes to realize long-period helical spin chains [5]. Hence, it is expected that the magnetism of YMn_2 is similar to that of the itinerant electron 1D magnet.

On the other hand, there have been few studies on itinerant electrons in asymmetric crystals, stressing the anisotropy in magnetic interaction. Motivated by the comparison with YMn_2, we tried to find a prototype of the 1D magnet in itinerant electron systems.

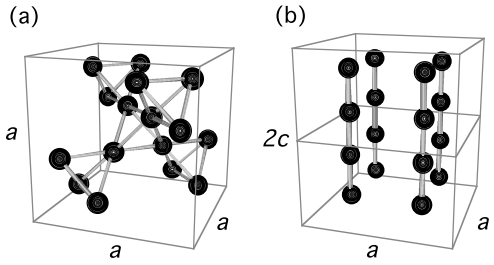

Figure 1. Mn sublattices in YMn_2 (a) and RMn_4Al_8 (b).

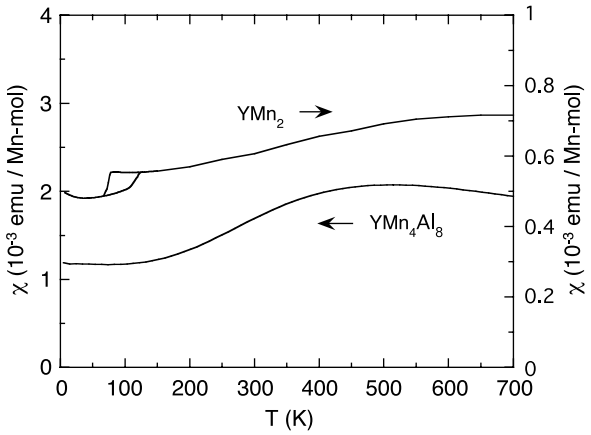

Figure 2. The magnetic susceptibility of YMn_2 and YMn_4Al_8. The Curie–Weiss-type upturn at low temperatures was subtracted from the susceptibility for YMn_4Al_8.

2. Comparison with quasi-1D itinerant electron magnets RMn_4Al_8

As typical itinerant electron magnets with a 1D atomic arrangement [6], we select RMn_4Al_8 (R = La, Y, Lu and Sc) with the tetragonal $ThMn_{12}$-derived $CeMn_4Al_8$-type structure, where Mn atoms form 1D chains along the *c* axis with the in-chain Mn–Mn distance of 2.6 Å, while having the large interchain distance of 4.4 Å (see figure 1(b)) [7, 8]. The in-chain distance is comparable to the Mn–Mn distance in YMn_2, 2.7 Å. The RMn_4Al_8 compounds show metallic conductivity [9–11] as YMn_2. The magnetic ground state of Mn in RMn_4Al_8 is basically nonmagnetic (except the partial spin freezing in $LaMn_4Al_8$ [11–15]). The reported values of the electronic specific heat coefficient γ are 265 and 83 mJ K^{-2} mol^{-1}/f.u. (formula unit) (66 and 21 mJ K^{-2} /Mn mol) for $LaMn_4Al_8$ and YMn_4Al_8, respectively [16], which are comparable to that for Y(Sc)Mn_2 ($\gamma \simeq 80$ mJ K^{-2}/Mn mol [17]). For both the systems, the resistivity shows a Fermi-liquid-like T^2 dependence at low temperatures [18, 19]. No appreciable anisotropy has been observed in the resistivity of single-crystalline YMn_4Al_8 [20]; RMn_4Al_8 is *not* a 1D conductor. The specific heat and resistivity results of both Y(Sc)Mn_2 and YMn_4Al_8 satisfy the Kadowaki–Woods relation. These results indicate that both Y(Sc)Mn_2 and RMn_4Al_8 are strongly correlated electron systems with nonmagnetic ground states. In RMn_4Al_8, the electron correlation mainly depends on the lattice volume, i.e. on the Mn–Mn bond length.

The temperature dependence of the susceptibility measured for YMn_4Al_8 [21] is shown in figure 2 together with that of YMn_2. The susceptibility of YMn_4Al_8 shows a broad

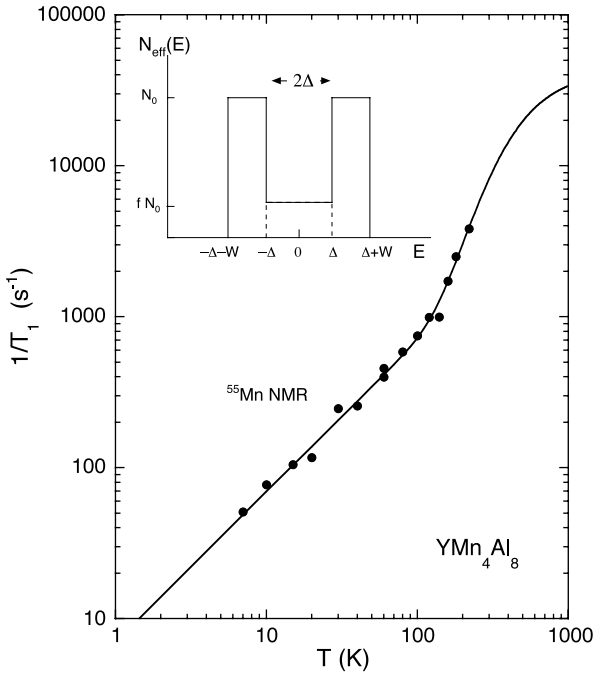

Figure 3. The temperature dependence of ^{55}Mn—$1/T_1$ for YMn_4Al_8 [21]. A similar temperature dependence has also been obtained for ^{27}Al—$1/T_1$. The inset shows the model of the spin excitation density of states with the Fermi energy at $E = 0$. The solid curve was calculated with $W = 720$ K, $\Delta = 620$ K and $f = 25\%$.

maximum at $T_{max} \simeq 500$ K (the broad maximum was originally found by Coldea *et al* [22]). At high temperatures, a Curie–Weiss-like temperature dependence with a negative Weiss temperature appears, suggesting antiferromagnetic interaction between Mn atoms [19]. YMn_2 shows a similar weak temperature dependence, possibly, with a broad peak at a higher temperature. (The low-temperature anomaly due to the antiferromagnetic ordering is easily suppressed in Y(Sc)Mn_2 or under pressure.) As the origin of the characteristic temperature dependence of YMn_4Al_8, the Mn atomic arrangement in RMn_4Al_8 reminds us of the Bonner–Fisher susceptibility expected for the 1D Heisenberg antiferromagnet, where T_{max} is in proportion to the exchange integral J between nearest-neighbour spins [23]. In the RMn_4Al_8 system, however, T_{max} is negatively correlated with the lattice volume, and hence the electron correlation, as revealed in studies of (La–Y–Lu–Sc)Mn_4Al_8 [19, 24]. Therefore, we ascribed the behaviour to the energy gap existing in the spin excitation spectrum as in well-known nearly ferromagnetic semiconductor FeSi [25]. For a fully opened gap as in FeSi, we expect zero susceptibility at 0 K, while the susceptibility of YMn_4Al_8 retains a finite value at 0 K. Thus we assumed a partially filled gap as shown in the inset of figure 3 (see [21] for details). Within this model, we expect continuous spin excitations at low temperature and activation beyond the gap at high temperature, which are actually observed in the nuclear spin–lattice relaxation rate $1/T_1$ for YMn_4Al_8; metallic T_1T = constant behaviour at low temperatures and exponential increase at high temperatures as seen in figure 3 [21]. Overall temperature dependences of both susceptibility and $1/T_1$ can be reproduced well with common band parameters by applying the model ($W = 720$ K, $\Delta = 620$ K and $f = 25\%$ for YMn_4Al_8). Within this model T_{max} gives a rough estimate of the gap width Δ, although

the parameters obtained may be less significant in comparing with actual physical quantities because of the extreme simplification. For $LaMn_4Al_8$, a broad inelastic neutron scattering centred near 30 meV has been reported [14], suggesting a spin excitation spectrum similar to that of Y(Sc)Mn_2 [26, 27] and the presence of spin-liquid-like state in both RMn_4Al_8 and YMn_2. On the other hand, the dimensionality seems to be of importance for explaining the quantitative features of the susceptibility. Although we expect smaller T_{max} ($\simeq\Delta$) for stronger electron correlation as known from the correlation between T_{max} and γ in (La–Y)Mn_4Al_8, the potential T_{max} for YMn_2 with larger γ is much higher than that of YMn_4Al_8 with smaller γ. This is probably because thermally excited spins are compensated better in the 3D tetrahedral lattice than in 1D spin chains.

Since the gap width, and also overall magnetism, strongly depends on the Mn–Mn bond length [19, 24], it is suggested that 1D nature of the Mn atomic arrangement is one of the principal origins of the gap formation in RMn_4Al_8. In insulating magnets, generally, both frustration and low dimensionality suppress long-range spin ordering. The present results evidenced similar effects in the strongly correlated itinerant electron system. The similar magnetisms of 3D frustrated YMn_2 and frustration-free quasi-1D RMn_4Al_8 support the idea discussed in section 1: the lowering of the magnetic dimensionality in the 3D frustrated lattice.

3. A spin gap in itinerant electron frustrated magnets

The above results can be interpreted as the frustration and the one dimensionality suppressing the long-range ordering and stabilizing the spin-liquid or spin-singlet-like state in the itinerant electron magnets. In general, one may expect the presence of a spin gap in the spin-liquid state. Neutron scattering experiments, however, were not successful in monitoring the narrow spin gap in frustrated itinerant electron magnets such as YMn_2. On the other hand for RMn_4Al_8, the spin excitation density of states formally introduced to explain the susceptibility and $1/T_1$ can be interpreted as an evidence of the gap formation. The partial filling in the gap is probably due to the interaction with conduction electrons originating from other metallic sites and/or the renormalization of the gap to a pseudogap, where in-gap states show heavy electron mass. As seen above, a nuclear magnetic resonance experiment, as a probe with good energy resolution, particularly at high temperature, is appropriate for detecting such a narrow pseudogap. Recently, an activation-type temperature dependence of $1/T_1$ has been reported for another itinerant electron 3D frustrated system β-Mn [28]; $1/T_1$ increases divergently above $\sim$150 K deviating from the $\sqrt{T}$ dependence expected for the nearly antiferromagnetic metal. This result may be evidence of the spin-gap formation in the frustrated itinerant electron magnet. It is interesting to note that, if we observe the susceptibility of β-Mn in detail, it exhibits a characteristic temperature dependence expected for the system with a spin gap as shown in figure 4. Since the susceptibility of β-Mn has a large positive temperature independent term, it is not realistic to analyse with the same model as was used for RMn_4Al_8. The rather low T_{max} ($\simeq$150 K), however, suggests a very narrow gap width. In β-Mn, there are two different crystallographic sites, 12d (Mn II) and 8c (Mn I) sites. We have proposed that the Mn II site is strongly correlated and geometrically frustrated, and that the Mn I site contributes to weakly correlated wide 3d bands [29]. The large temperature independent background of the susceptibility, especially in β-Mn, is probably ascribable to marked renormalization of the gap due to the strong Mn II–Mn I coupling, as suggested from recent electronic state calculations [30].

In conclusion, we have pointed out the qualitative similarity between the magnetism of frustrated YMn_2 and that of quasi-1D RMn_4Al_8. In both the systems, long-range ordering is suppressed in spite of strong electron correlation, and the spin-liquid-like state is realized. We

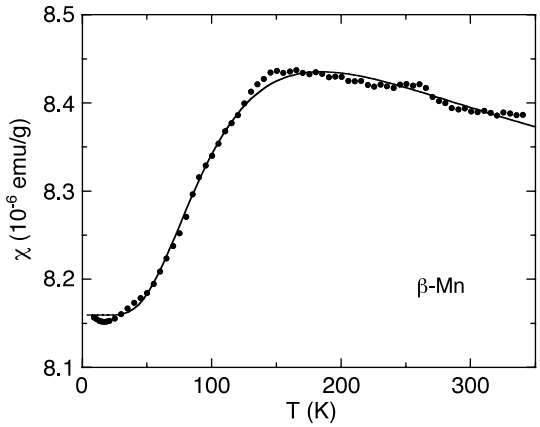

Figure 4. The magnetic susceptibility of β-Mn. The small Curie–Weiss-type upturn at low temperatures was subtracted. The solid curve was calculated for the background subtracted susceptibility by assuming square bands separated by a gap with $2\Delta = 540$ K.

have discussed the formation of a spin pseudogap as part of the nature of the spin liquid in itinerant electron magnets, and possible observation via nuclear magnetic relaxation.

Acknowledgment

This work was supported by Grants-in-Aid for Scientific Research from the Ministry of Education, Culture, Sports, Science and Technology, Japan.

References

[1] Shiga M 1994 *J. Magn. Magn. Mater.* **129** 17
[2] Ballou R, Lelievre-Berna E and Fak B 1996 *Phys. Rev. Lett.* **76** 2125
[3] Nakamura H and Shiga M 2001 *J. Alloys Compounds* **326** 157
[4] Tchernyshyov O, Moessner R and Sondhi S L 2002 *Phys. Rev.* B **66** 064403
[5] Nakamura H, Metoki N, Suzuki S, Takayanagi F and Shiga M 2001 *J. Phys.: Condens. Matter* **13** 475
[6] Yamasaki T, Matsui K, Nakamura H and Shiga M 2001 *Solid State Commun.* **119** 415
[7] Buschow K H J, van Vucht J H N and van den Hoogenhof W W 1976 *J. Less-Common Met.* **50** 145
[8] Felner I and Nowik I 1979 *J. Phys. Chem. Solids* **40** 1035
[9] Palasyuk A M, Kotur B Ya, Bauer E, Michor H and Hilscher G 2004 *J. Alloys Compounds* **367** 205
[10] Kotur B Ya, Myakush O R and Palasyuk A M 2004 *J. Alloys Compounds* **367** 211
[11] Muro Y, Giri S, Motoyama G, Nakamura H and Kohara T 2005 *J. Phys. Soc. Japan* **74** 1135
[12] Rainford B D, Leavey C J, Hillier A D and Stewart J R 2005 *Physica* B **359–361** 929
[13] Muro Y, Motoyama G, Nakamura H and Kohara T 2006 *Physica* B **378–380** 673
[14] Leavey C J, Rainford B D, Stewart J R and Adroja D T 2006 *Physica* B **348–380** 675
[15] Muro Y, Nakamura H and Kohara T 2006 *Proc. ICM2006*
[16] Hagmusa I H, Klaasse J C P, Brück E, de Boer F R and Buschow K H J 2000 *J. Alloys Compounds* **297** 21
[17] Wada H, Shiga M and Nakamura Y 1989 *Physica* B **161** 197
[18] Shiga M, Fujisawa K and Wada H 1993 *J. Phys. Soc. Japan* **62** 1329
[19] Nakamura H, Muro Y, Giri S, Uemura J, Kobayashi H, Koyama T and Kohara T 2005 *J. Phys. Soc. Japan* **74** 2421
[20] Muro Y, Motoyama G, Nakamura H and Kohara T, unpublished
[21] Nakamura H, Giri S and Kohara T 2004 *J. Phys. Soc. Japan* **73** 2971
[22] Coldea M, Neumann M, Lükehoff St, Mähl S and Coldea R 1998 *J. Alloys Compounds* **278** 72
[23] Bonner J C and Fisher M E 1964 *Phys. Rev.* **165** A640

[24] Muro Y, Nakamura H and Kohara T 2006 *J. Phys.: Condens. Matter* **18** 3931
[25] Jaccarino V, Wertheim G K, Wernick J H, Walker L R and Arajs S 1976 *Phys. Rev.* **160** 476
[26] Shiga M, Wada H, Nakamura Y, Deportes J, Ouladdiaf B and Ziebeck R A 1988 *J. Phys. Soc. Japan* **57** 3141
[27] Nakamura H, Takayanagi F, Shiga M, Nishi M and Kakurai K 1996 *J. Phys. Soc. Japan* **65** 2779
[28] Hama T, Matsumura M, Kato H, Yamagata H, Kohori Y, Kohara T and Iwamoto Y 2004 *J. Phys. Soc. Japan* **73** 2305
[29] Nakamura H, Yoshimoto K, Shiga M, Nishi M and Kakurai K 1997 *J. Phys.: Condens. Matter* **9** 4701
[30] Hafner J and Hobbs D 2003 *Phys. Rev.* B **68** 014408