Available online at www.sciencedirect.com
ScienceDirect
Journal of Magnetism and Magnetic Materials 307 (2006) 165–169
Journal of magnetism and magnetic materials
www.elsevier.com/locate/jmmm

Magnetic states of Gd_2Co_2Al and Gd_2Co_2Ga with the W_2Co_2B-type structure

X.B. Liu*, Z. Altounian

Center for the Physics of Materials and Department of Physics, McGill University, Rutherford Physics Building, 3600 University St., Montreal, Que., Canada H3A 2T8

Received 12 September 2005; received in revised form 10 January 2006
Available online 2 May 2006

Abstract

Electronic band structures of the W_2Co_2B-type Gd_2Co_2Al and Gd_2Co_2Ga compounds are calculated by self-consistent spin-polarized linear muffin-tin orbital (LMTO) method, within a local spin density approximation (LSDA) frame. The calculation results indicate that the magnetic moments of Gd and Co are antiferromagnetic coupled via the 3d–5d interaction in these compounds. The Co magnetic moment is induced by the exchange field of the ferromagnetically ordered Gd^{3+} in these compounds. All the ground states are shown to be ferrimagnetic by the total electronic energy calculations. Very good agreement between the calculated and experimental values for the lattice constants and magnetic moments is obtained for Gd_2Co_2Al and Gd_2Co_2Ga compounds.
© 2006 Elsevier B.V. All rights reserved.

PACS: 75.10.Lp; 75.50.Cc

Keywords: Magnetic state; Electronic structure; LMTO; Magnetic moment

1. Introduction

Since the giant magnetocaloric effect (MCE) was discovered in $Gd_5Si_xGe_{4-x}$ compounds [1], gadolinium intermetallic compounds such as Gd_2MgGe_2, Gd_2Co_2Al, Gd_5Sn_4, GdFeSi, $GdCo_3$ and $(Gd, Er)Co_2$ are widely studied for their possible application as refrigerants in magnetic refrigeration device [2–7]. Gd_2Co_2Al and Gd_2Co_2Ga crystallize in the W_2Co_2B-type structure with a space group *Immm* [3,8]. According to Canepa et al. [3], the compounds Gd_2Co_2Al and Gd_2Co_2Ga order ferromagnetically below 78.2 and 76 K, respectively. The authors ascribe the magnetic ordering solely to the contribution of the Gd atoms. They give experimental value for the magnetic moment as about $6.6\,\mu_B$/Gd. Kervan [9] studied these compounds by fitting the measured thermomagnetic curves [3] using molecular field theory based on the two-sublattice model and found that the interaction between Gd and Co moments is antiferromagnetic.

In order to clarify the magnetic ground states, a first principle density functional theory (DFT) calculations are performed using the linear muffin-tin orbital method (LMTO) with the atomic sphere approximation (ASA) [10–13]. The most stable lattice volume and magnetic states are discussed.

2. Results and discussion

The standard approximation to the DFT like local spin density approximation (LSDA) or generalized gradient approximation (GGA) fail to describe the on-site Coulomb interaction and correlation among highly localized electrons such as 4f electrons in rare earth (RE) and RE-based compounds. One simple method is that the localized 4f orbitals are not contained in the valence basis set but are treated as part of the atomic like core [15]. Although fixing the 4f occupation number to an integer value often violates the Kohn–Sham rule to occupy the lowest one electron states in constructing the charge and spin density [15], the

*Corresponding author. Tel.: +1 514 398 2863; fax: +1 514 398 6526.
E-mail address: liux@physics.mcgill.ca (X.B. Liu).

0304-8853/$ - see front matter © 2006 Elsevier B.V. All rights reserved.
doi:10.1016/j.jmmm.2006.03.064

'core state' treatment has been successfully applied in some magnetic RE metal and RE compounds [16,17]. More complex methods include a LDA + U approach [18–20] and a self-interaction corrected (SIC) LSDA scheme [15].

The first principle band structure calculations for Gd_2Co_2Al and Gd_2Co_2Ga are performed by LMTO method with the ASA. All relativistic effects are included except the spin–orbit interaction. Exchange and correlation effects are treated with LSDA [14]. The localized Gd 4f states are treated as fractional core electrons. Here, Gd has three electrons entering valence band, which conform to the fact that RE element often displays a R^{3+} state in RE-based compounds.

Gd_2Co_2Al and Gd_2Co_2Ga crystallize in the W_2Co_2B-type structure with a space group *Immm*. In the unit cell, Gd, Co, and Al (Ga) atoms occupy the sites 4*j*, 4*h*, and 2*a*, respectively. Published experimental structural data [3] are used for the electron structure calculations (Table 1).

Fig. 1 shows the total and partial density of states (DOS) for Gd_2Co_2Al at the experimental lattice constants. The vertical line indicates the position of the Fermi level. The total DOS for Gd_2Co_2Al consists of the lower part (below −0.4 Ry), which contains mainly Al s-states, and the higher part (above −0.35 Ry), where the d-states of Co and Gd atoms and the Al p-states occupy. Comparing the corresponding local DOS of Gd, Co, and Al, it is clear that the DOS of Co contributes mostly to the total DOS near the Fermi level. Fig. 2 displays the spin projected local DOS curves for Gd-5d, Co-3d and Al-p. It shows clearly the hybridization between the Co-3d state and Gd-5d states. The majority subbands of Gd 5d prefer to mix with the Co 3d minority subbands, indicating the antiferromagnetic coupling between Co 3d and Gd 5d. A similar situation is observed in the Gd_2Co_2Ga compounds. The spin-projected DOS for Gd_2Co_2Ga are presented in Fig. 3.

Fig. 4 displays the calculated total energy as a function of lattice volume (i.e. unit cell volume) for Gd_2Co_2Al. The ratio between the lattice constants is fixed in the calculations. The calculated energies for spin unpolarized state (paramagnetic state or nonmagnetic state) and spin polarization state (magnetic state) are denoted by PM and FM, respectively. The minimum energy is obtained, within an uncertainty of 1.0%, at the room temperature experimental volume, V_{exp}, for the magnetic states. The total energy for the paramagnetic state is much higher than that for magnetically ordering state. These calculations are in very good agreement with the experimental results.

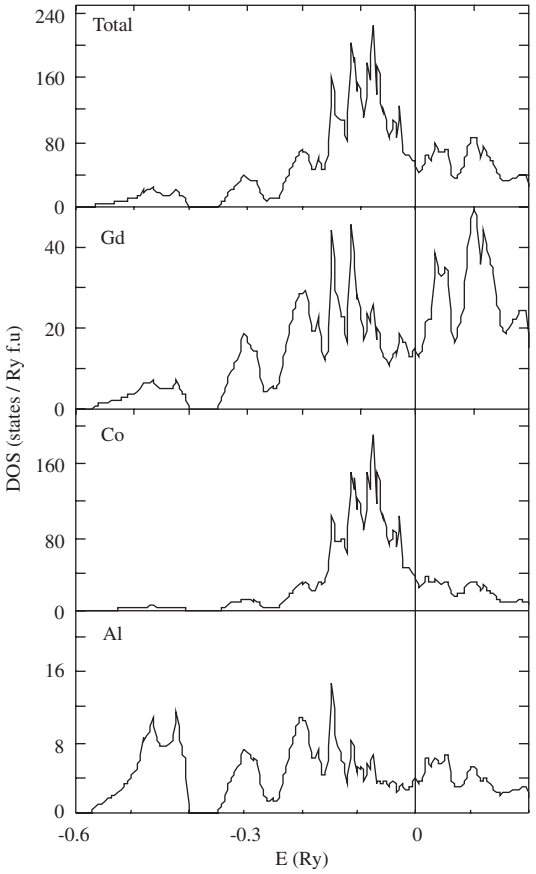

Fig. 1. Total and partial DOS for nonmagnetic Gd_2Co_2Al. The Fermi level is at $E = 0$.

Table 1
Experimental lattice constants, atomic positions and magnetic moments [3], and calculated magnetic moments for Gd_2Co_2Al and Gd_2Co_2Ga compounds

	Gd_2Co_2Al	Gd_2Co_2Ga
Lattice constants (Å)	$a = 4.103$	$a = 4.127$
	$b = 5.501,\ c = 8.535$	$b = 5.448,\ c = 8.495$
Atom position	(x, y, z)	(x, y, z)
Gd (4*j*)	(0.5, 0, 0.2961)	(0.5, 0, 0.2946)
Co (4*h*)	(0, 0.2243, 0.5)	(0, 0.2210, 0.5)
Al (Ga) (2*a*)	(0, 0, 0)	(0, 0, 0)
Exp. moment (μ_B/f.u)	13.2	13.2
Cal. moment (μ_B/f.u)	14.0	13.8
Cal. moment (μ_B/atom)		
Gd (4*j*)	7.35	7.37
Co (4*h*)	−0.35	−0.43
Al (Ga) (2*a*)	0.02	0.02

The calculated magnetic moments for Gd_2Co_2Al and that of Gd, Co, and Al as a function of the lattice volume are shown in Fig. 5. At V_{exp}, the local moments of Gd, Co, and Al are 7.349, −0.352, and 0.022 μ_B, respectively (Table 1). The calculated moment is 14.0 μ_B per formula unit, in good agreement with the observed value (13.2 μ_B at 5 K, Table 1). With increasing lattice volume, the local moment of Co, mainly from 3d electrons, increases due to the volume effect. The local moment of Gd originates from 4f and 5d electrons. The Gd 4f moment is 7 μ_B and remain constant because the Gd 4f electrons are treated as core electrons. The 5d moment of Gd increases with lattice volume, but less than that of Co 3d. As the magnetic coupling between Gd and Co is antiferromagnetic, the net magnetic moment of Gd_2Co_2Al decreases with increasing

Download English Version:

https://daneshyari.com/en/article/1804639

Download Persian Version:

https://daneshyari.com/article/1804639

Daneshyari.com