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Magnetism of $L1_0$ compounds with the composition MT (M=Rh, Pd, Pt, Ir and T=Mn, Fe, Co, Ni)[a]

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The electronic band structure of ordered equiatomic compounds of $3d$ transition elements (Mn, Fe, Co, Ni) with nonmagnetic $4d$ and $5d$ elements (Rh, Pd, Pt, Ir) are investigated by linear muffin-tin orbital calculations. The systematic study considers $3d$ and $4d/5d$ spin moments and interatomic exchange interactions, with emphasis on the comparison between ferromagnetic and antiferromagnetic order. Total and site-resolved exchange interactions are calculated from first principles, and the obtained exchange constants are used to estimate ordering temperatures on a mean-field level. © *2004 American Institute of Physics.* [DOI: 10.1063/1.1687631]

I. INTRODUCTION

Equiatomic magnetic compounds crystallizing in the tetragonal $L1_0$ phase have attracted much attention as permanent magnets and in magnetic recording.[1–7] These compounds are of fundamental interest for their high uniaxial anisotropy to about 10^7 MJ/m³, which results from their natural superlattice structure, and CoPt was the first true permanent-magnet alloy. In addition, $L1_0$ nanoclusters have high potential as cluster-assembled granular films for 1 Tb/in.² perpendicular recording media. Exchange interactions in the compounds are important because they determine the zero- and finite-temperature properties such as magnetic order, finite-temperature anisotropy affecting, for example, domain formation and spin-flop transitions.

The tetragonal $L1_0$ intermetallics have the general structure MT, where M is an iron-series ($3d$) transition metal and T is a $4d$ or $5d$ transition metal. Many intermetallics where M = Fe or Co are ferromagnetic (FM), although antiferromagnetism (AFM) may be possible in some Fe-containing compounds.[8] $L1_0$ intermetallics where M = Mn are typically AFM, which reflects a general trend towards AFM due to half-filled $3d$ shells. Concerning first-principles calculations on $L1_0$ magnets, Ravindran *et al.*[9] calculated the magnetocrystalline anisotropy of some $L1_0$ compounds. This research was preceeded by earlier calculations on various $3d$-$4d/5d$ multilayers,[10] which contain the $L1_0$ structure as a special case. Moruzzi and Marcus,[11] discussed structural effects on magnetic properties of FeRh and FePd by first-principles total-energy calculations. Garcia *et al.*[12] obtained the magnetocrystalline anisotropy of FePd, and there are Curie temperature (T_C) calculations for FePt in a B_2O_3 matrix.[3] However, compared to Cu_3Au and other related structures,[13,14] little work has been done on $L1_0$ compounds made from elements other than Fe, Co, Pd, and Pt.

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[a]No proof corrections received from author prior to publication.

II. METHOD OF CALCULATIONS

To perform the self-consistent, spin-polarized electronic structure calculations, the linear muffin-tin orbital (LMTO) method within the atomic sphere approximation[15] is used. The calculations are performed for 288 k points, and the Barth-Hedin exchange-correlation potential was employed. The local spin-up and spin-down densities of states and magnetic moments are calculated using the linearized tetrahedron method. A minimal basis set consisting of of s, p, and d orbitals is used for both types of atoms and the spin-orbit coupling is not included in our calculations. Due to the different sizes of the constituent atoms it is necessary to assume different Wigner-Seitz (WS) cell radii, but the ratio of WS radii is kept to be the same as in their elemental form. Experimental data for the lattice parameters for all the compounds are taken from Ref. 16. One example is that, for FePt compound, WS values are 2.709 a.u. and 2.947 a.u. for Fe and Pt, respecively. The $L1_0$ structure can be understood as alternate layers of M (Mn, Fe, Co, Ni) and T (Rh, Pd, Ir, Pt) atoms with each M atom surrounded by eight T atoms and four M atoms as nearest neighbors. The unit cell contains four atoms but can be considered as simple tetragonal cell with two atoms. The interatomic exchange interactions (J_{ij}) are calculated in the Heisenberg approximation. Details of the method can be found elsewhere.[17] The total exchange parameter (J_0) reflects the energy change due to small-angle rotation of the moment at one site. In contrast to the J_{ij}, it is given by site-diagonal scattering matrix (or Green function), where $i = j$. Using J_0, transition temperatures are estimated within mean-field theory, using the relation $T_C = 2/3k_B \langle J_0 \rangle$ ($T_N = -T_C$).

III. RESULTS AND DISCUSSION

Table I shows self-consistent magnetic moments for all investigated compounds. The results are in agreement with all other calculations and with the available experimental data. Table II shows the calculated J_0 and calculated mean-field ordering temperature along with some available experimental data.[18] As expected, the T_C is overestimated by the

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TABLE I. Magnetic moments (μ_B/f.u.) in the form of $m_{total}=m_T+m_M$. Experimental values are quoted in parentheses.

	Mn	Fe	Co	Ni
Rh	3.010=2.978+0.022	2.238=2.214+0.022	1.321=1.119+0.102	0
Pd	4.138=3.845+0.293	3.299=2.952+0.347	2.281=1.902+0.379	1.013=0.732+0.281
	(4.4+0.0)	(2.85+0.35)	(1.97+0.35)	(m_{avg}/atom=0.55)
Ir	1.116=1.222−0.006	1.909=1.924−0.015	0.898=0.800+0.098	0
Pt	4.022=3.667+0.355	3.265=2.916+0.349	2.254=1.837+0.417	0
	(4.3+0.0)	(2.80+(−0.25))	(1.60+0.25)	

TABLE II. Total exchange parameter J_0 (mRy= 156 K) for M and T sites and Curie temperature (K) along with experimental values given in parentheses.

		J_0 Mn	Fe	Co	Ni	T_C Mn	Fe	Co	Ni
Rh	M	−17.883	3.024	2.863		1850(200)	347	355	
	T	0.277	0.273	0.512					
Pd	M	−14.256	9.543	9.696	1.974	1285(810)	1270(700)	1284	310
	T	2.045	2.529	2.506	0.956				
Ir	M	−2.367	3.436	1.861		240	377	217	
	T	0.092	0.149	0.199					
Pt	M	−15.008	8.338	8.766		1318(970)	1128(750)	1186(850)	
	T	2.482	2.371	2.506					

TABLE III. Site-resolved exchange interaction parameters (mRy) for the FM and AFM configurations. The table shows the M-M (J_{11},J_{13}) and M-T (J_{12},J_{14}) pairs and the number of neighbors with increasing interatomic distance.

	J (FM)	Mn	Fe	Co	Ni	J (AFM)	Mn	Fe	Co	Ni
Rh	$J_{12}(8)$	0.041	0.034	0.051		$J_{12}(4)$	0	0.003	0.001	
	$J_{11}(4)$	−4.550	0.657	0.481		$J_{14}(4)$	0	0	0	
	$J_{11}(2)$	0.694	0.792	0.148		$J_{11}(4)$	−3.322	−0.157	0.806	
	$J_{11}(4)$	−0.293	0.054	−0.043		$J_{13}(2)$	0.010	−0.008	0.002	
	$J_{11}(8)$	−0.400	−0.036	0.133		$J_{11}(4)$	−0.587	0.253	−0.096	
	$J_{12}(8)$	0.041	0.034	0.051		$J_{13}(8)$	0.001	0.004	0.003	
	$J_{11}(8)$	0.468	−0.124	−0.155		$J_{14}(4)$	0	0	0	
						$J_{12}(4)$	0	0.003	0.014	
						$J_{13}(8)$	−0.246	0.333	0.134	
Pd	$J_{12}(8)$	0.227	0.234	0.205	0.073	$J_{12}(4)$	−0.093	0.004	0.012	0.030
	$J_{11}(4)$	−4.331	0.533	1.605	0.241	$J_{14}(4)$	0	0	0	0
	$J_{11}(2)$	1.691	0.090	−0.127	−0.064	$J_{11}(4)$	−4.951	1.361	1.775	0.172
	$J_{11}(4)$	0.042	0.952	0.120	0.038	$J_{13}(2)$	0	0.010	0.004	0
	$J_{11}(8)$	−0.223	0.205	0.234	0	$J_{11}(4)$	0.015	0.401	0.220	0.030
	$J_{12}(8)$	0.277	−0.234	0.205	0.073	$J_{13}(8)$	0	0	−0.005	0
	$J_{11}(8)$	−0.230	0.234	−0.125		$J_{14}(4)$	0	0	0	0
						$J_{12}(4)$	−0.093	0.004	0.012	0.030
						$J_{13}(8)$	0.098	0.318	0.174	−0.026
Ir	$J_{12}(8)$	0.005	0.0156	0.014		$J_{12}(4)$	0.001	0	0.003	
	$J_{11}(4)$	−1.129	0.748	0.319		$J_{14}(4)$	0	0	0	
	$J_{11}(2)$	0.080	0.502	0.095		$J_{11}(4)$	−1.035	0.071	0.404	
	$J_{11}(4)$	0.204	−0.116	−0.038		$J_{13}(2)$	0.003	−0.010	0.002	
	$J_{11}(8)$	−0.109	0.067	0.084		$J_{11}(4)$	0.291	−0.065	−0.068	
	$J_{12}(8)$	0.006	0.016	0.014		$J_{13}(8)$	0.001	−0.001	0.001	
	$J_{11}(8)$	0.097	−0.048	−0.087		$J_{14}(4)$	0	0	0	
						$J_{12}(4)$	0.001	0	0.003	
						$J_{13}(8)$	−0.133	0.235	0.057	
Pt	$J_{12}(8)$	0.264	0.239	0.226		$J_{12}(4)$	0.013	0.009	0	
	$J_{11}(4)$	−3.188	0.787	1.305		$J_{14}(4)$	0	0	0	
	$J_{11}(2)$	2.650	0.056	−0.053		$J_{11}(4)$	−4.847	1.884	1.312	
	$J_{11}(4)$	−0.602	0.711	0.038		$J_{13}(2)$	−0.025	0.017	0.001	
	$J_{11}(8)$	−0.357	0.253	0.267		$J_{11}(4)$	0.126	−0.052	0.113	
	$J_{12}(8)$	0.263	0.239	0.226		$J_{13}(8)$	0.004	−0.001	−0.004	
	$J_{11}(8)$	−0.552	−0.400	−0.235		$J_{14}(4)$	0	0	0	
						$J_{12}(4)$	0.013	0.008	0	
						$J_{13}(8)$	0.377	0.491	0.302	

above simple mean-field expression. Our J_0 values are negative for Mn compounds suggesting the AFM order to be more stable in MnRh, MnPd, MnIr, and MnPt. For the compounds of Fe and Co, J_0 calculations suggest FM ordering at equilibrium lattice parameters. Compounds of Ni with Rh, Ir, and Pt are paramagnetic (PM) at equilibrium lattice parameters, in accordance with the experimental data.[18] However, the strain dependence of the spin structure may yield transitions to a FM state, for example due to embbeding of nanoparticles in a matrix. Further experimental complications are coexistent phases, such as disordered fcc. Table III shows site-resolved exchange interaction parameters $[(J_{ij})s]$ for all for the FM and AFM configurations. Nearest neighbors around the magnetic $3d$ transition element (Mn, Fe, Co, Ni) are also given upto a distance of nearly twice the lattice parameter. J_{11} and J_{13} indicate exchange between M atoms and J_{12} and J_{14} indicate exchange between M-T atoms. As evident from the data in Table III, all Mn compounds exhibit pronounced negative exchange interactions between four Mn-Mn first nearest pairs, $J_{11}(4)$, indicating that the Mn spins in a given plane would prefer to align antiferromagnetically, trying to form an intraplane checkerboard pattern in addition to the AFM interlayer coupling. In all Fe and Co compounds, FM ordering is more favorable, whereas the Ni compounds are PM, with the exception of the ferromagnetic NiPd.

IV. CONCLUSIONS

We have performed first-principles calculations for $L1_0$ compounds of Mn, Fe, Co with Rh, Pd, Pt, Ir. The obtained moments and exchange coupling constants are then used to discuss the spin structure of the intermetallics and to estimate the ordering temperature. As expected from band-filling arguments, there is a trend from ferromagnetism to antiferromagnetism when going from late $3d$ elements towards the middle of the series. However, for some compounds the energy differences are small and strain or doping may stabilize the FM phase.

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