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Electronic structure and magnetic properties of $Ce_2Pd_{1-x}Co_xSi_3$ and $Ce_2Pd_{1-x}Fe_xSi$*

G. KOZŁOWSKI[1], A. JEZIERSKI[2**]

[1]Faculty of Technical Physics, Poznań University of Technology,
ul. Nieszawska 13A, 60-965 Poznań, Poland

[2]Institute of Molecular Physics, Polish Academy of Sciences,
M. Smoluchowskiego 17, 60-179 Poznań, Poland

Electronic and magnetic properties of $Ce_2Pd_{1-x}Co_xSi_3$ and $Ce_2Pd_{1-x}Fe_xSi_3$ alloys have been calculated by the TB LMTO-ASA method. The spin polarized calculations indicate that the magnetic moment of $Ce_2Pd_{1-x}Co_xSi_3$ decreases upon the increase of the concentration x but for $Ce_2Pd_{0.5}Fe_{0.5}Si_3$ the value of the magnetic moment has a minimum.

Key words: *electronic structure; LMTO; magnetic moment; intermetallic compounds*

1. Introduction

The intermetallic Ce_2TSi_3 compounds have been studied experimentally in the last years [1–5]. Ce_2TSi_3 systems with T = (Co, Fe) crystallizes into hexagonal crystal structures described by the space group *P*6/*mmm* (No. 191). The positions of atoms are listed in Table 1. Magnetic measurements [1] suggested a reduction of the cerium moment for systems with high Co content. Ce_2PdSi_3 was reported to exhibit the Kondo effect [2] and classified as a heavy fermion system with a strong anisotropic magnetic behaviour [3]. In this work, we present the effect of substitution of Co and Fe in Ce_2PdSi_3 on its electronic and magnetic properties.

2. Method of calculations

The electronic and magnetic properties were calculated by ab-initio spin-polarized Tight Binding Linear Muffin Tin Orbital (TB LMTO) method in the atomic spheres

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**Corresponding author, e-mail: andrzej.jezierski@ifmpan.poznan.pl

approximation (ASA) [6]. The exchange correlation potential was assumed according to von Barth and Hedin [7]. We apply a scalar-relativistic approximation for the band electrons and the fully-relativistic approximation for the frozen core electrons. The values of the atomic spheres' radii were chosen in such a way that the sum of volumes of all atomic spheres was the same as the volume of the unit cell. The calculations were performed for the experimental lattice parameters [1] listed in Table 2. The number of *k*-points in the irreducible Brillouin zone changed from 340 to 370 depending on the symmetry of the system.

Table 1. Nonequivalent atomic positions in Ce_2PdSi_3

Atom	Wyckoff site	*x*	*y*	*z*
Ce(1)	1a	0	0	0
Ce(2)	3f	0.5	0	0
Pd	2d	0.3333	0.6666	0.5
Si	6m	0.1702	0.3403	0.5

Table 2. Lattice parameters and the space groups of $Ce_2Co_xPd_{1-x}Si_3$ and $Ce_2Fe_xPd_{1-x}Si_3$

Material	*a* [Å]	*c* [Å]	Space group
Ce_2PdSi_3	8.2631	4.2830	*P*6/*mmm*
$Ce_2Co_xPd_{1-x}Si_3$			
$x = 0.25$	8.2241	4.2678	*P*2*mm*
$x = 0.5$	8.1869	4.2494	*Pcmm*
$x = 0.75$	8.1460	4.2297	*P*2*mm*
$x = 1$	8.1040	4.1970	*P*6/*mmm*
$Ce_2Fe_xPd_{1-x}Si_3$			
$x = 0.25$	8.2414	4.2674	*P*2*mm*
$x = 0.5$	8.2229	4.2520	*P*2*mm*
$x = 0.75$	8.1956	4.2431	*P*6/*mmm*
$x = 1$	8.1240	4.2120	*P*6/*mmm*

The calculations were performed for the supercell model ($Ce_8T_4Si_{12}$). In the supercell model, we had four positions of transition metal and we changed the concentration of impurity with the step of 0.25. The electronic and magnetic properties were calculated for the various distributions of Pd and Co(Fe) atoms in the supercell. From all possible arrangements of Pd and Co(Fe) atoms, one with a minimum total energy was chosen for further calculations.

3. Results and discussion

In Figure 1a, we present the total density of states (DOS) and the partial density of states (PDOS) of ferromagnetic Ce_2PdSi_3. The density of states consists of three regions. The bands situated at the bottom of the energy scale were occupied by 3s states

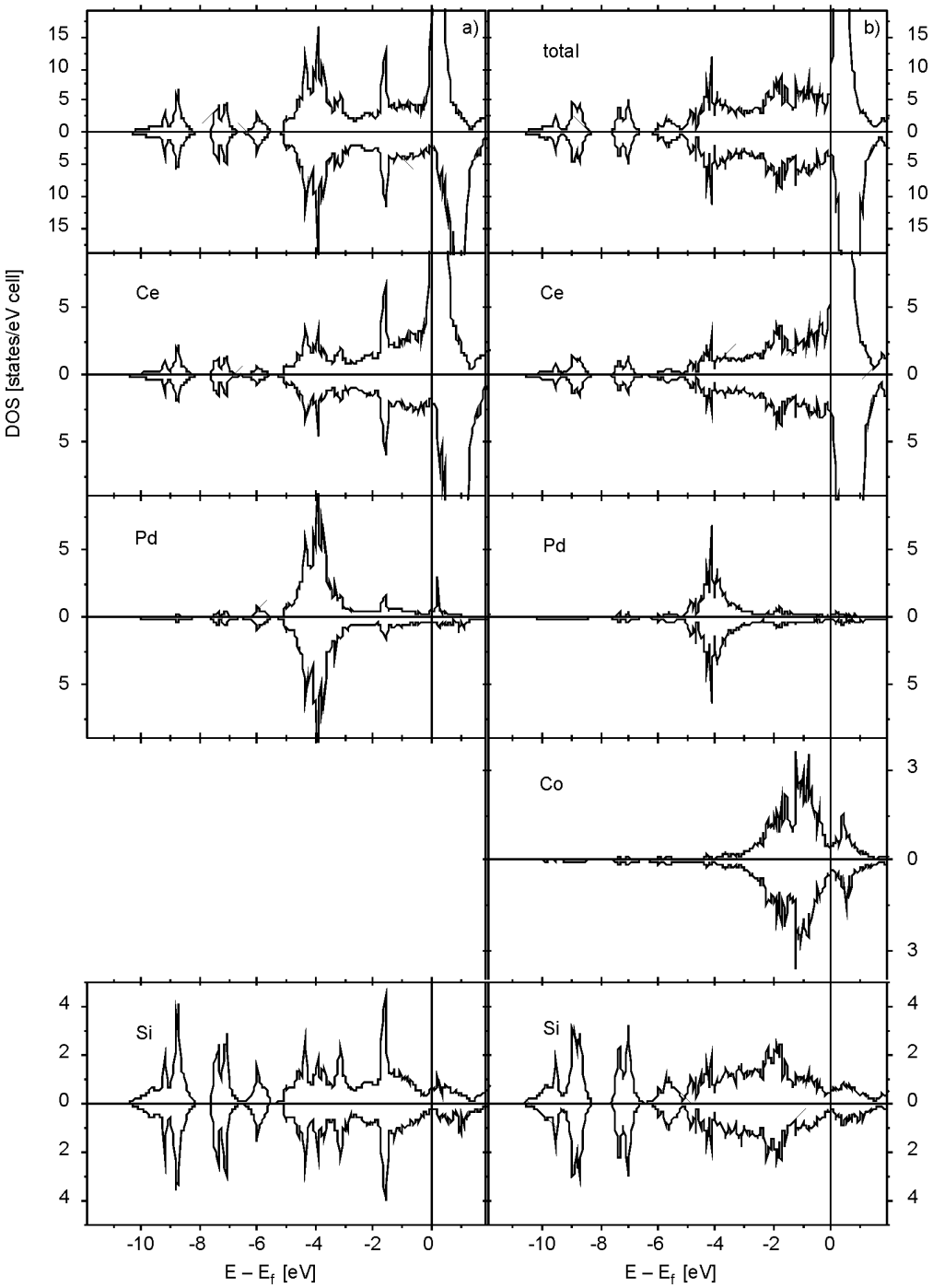

Fig. 1. Total and partial densities of states of: a) Ce_2PdSi_3, b) $Ce_2Pd_{0.5}Co_{0.5}Si_3$

of Si. The bottom of valence band is dominated by 4d states of Pd which hybridize with 3p states of Si. 4f states of Ce are located near the Fermi level and give the main contribution to the density at the Fermi level. The total DOS at Fermi energy is estimated as 19.19 (states/eV f.u.) The magnetic moment of Ce_2PdSi_3 obtained from the spin-polarized self-consistent calculations is $2.12\mu_B$ (Table 3).

Table 3. Magnetic moment on cerium [μ_B] in $Ce_2Pd_{1-x}Co_xSi_3$ and $Ce_2Pd_{1-x}Fe_xSi_3$

Material	Magnetic moment on cerium [μ_B]				Total magnetic moment [μ_B]
	Ce	Ce1	Ce2	Ce3	
Ce_2PdSi_3	0.58	–	–	0.48	2.12
$Ce_2Pd_{1-x}Co_xSi_3$					
$x = 0.25$	0.58/0.6	0.41/0.42	–	0.14/0.15	1.46
$x = 0.5$	0.57	0.33	0.11	0.03	0.91
$x = 0.75$	–	0.22/0.3	0.05/0.08	–0.06/–0.09	0.31
$x = 1$	–	–	0	0	0
$Ce_2Pd_{1-x}Fe_xSi_3$					
$x = 0.25$	0.6/0.56	–0.62	–	0.11/0.13	0.85
$x = 0.5$	0.39	0.39	0.39	0.39	0.15
$x = 0.75$	–	–0.5/–0.52	–0.54	0.3/0.41	0.71
$x = 1$	–	–	0	0	0

In Figure1b we plotted the total and partial densities of states of $Ce_2Pd_{0.5}Co_{0.5}Si_3$. The substitution of Co by Pd leads to a change of the total density of states particularly near the Fermi energy. Co 3d states are located below the Fermi energy and we observed a strong hybridization between 4f Ce and 3d Co states. The local disorder and hybridization lead to the change of the shape of cerium density of states.

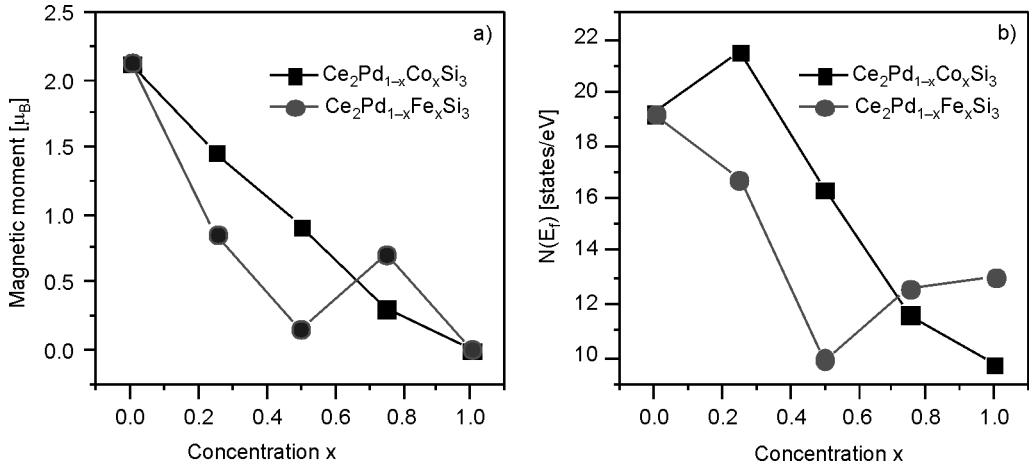

Fig. 2. Dependence of the total magnetic moment (a) and the density of states at the Fermi level $N(E_f)$ (b) on the concentration x for $Ce_2Pd_{1-x}Co_xSi_3$ and $Ce_2Pd_{1-x}Fe_xSi_3$

The dependences of the total magnetic moment on concentration in $Ce_2Pd_{1-x}Co_xSi_3$ and $Ce_2Pd_{1-x}Fe_xSi_3$ are presented in Fig. 2a. The total magnetic moment in $Ce_2Pd_{1-x}Co_xSi_3$ decreases upon increase of cobalt concentration. In $Ce_2Pd_{1-x}Fe_xSi_3$ we observe a similar dependence up to $x = 0.5$ but for $x = 0.75$ the total magnetic moment increases. In Table 3, we presented the values of the total magnetic moment on cerium atoms, that strongly depended on the local environment. The dependences of the densities of states at the Fermi level $N(E_f)$ on concentration x for $Ce_2Pd_{1-x}Co_xSi_3$ and $Ce_2Pd_{1-x}Fe_xSi_3$ alloys is presented in Fig. 2b. We observe different dependences of $N(E_f)$ on concentration x for both systems. The value of $N(E_f)$ in $Ce_2Pd_{1-x}Co_xSi_3$ decreases in the range $0.25 < x < 1.0$, however in $Ce_2Pd_{1-x}Fe_xSi_3$ alloys the value of $N(E_f)$ decreases up to $x = 0.5$ and then increases.

4. Conclusion

Ab-intio spin-polarized TB LMTO calculations have shown that substitution of palladium atom by cobalt or iron atoms changed electronic and magnetic properties of $Ce_2Pd_{1-x}Co_xSi_3$ and $Ce_2Pd_{1-x}Fe_xSi_3$ systems. Our calculations indicated that the total magnetic moment decreased upon increasing the Co and Fe concentration up to 50%, what was in a good agreement with experimental observation. For $Ce_2Pd_{1-x}Fe_xSi_3$, the density of states at the Fermi level $N(E_f)$ decreases till $x = 0.5$ and then increases up to Ce_2FeSi_3.

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