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Magnetic properties of MoPd₃

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Abstract. The Stoner model of ferromagnetism in combination with self-consistent linear muffin-tin orbital paramagnetic calculations are used for the investigation of magnetic properties of MoPd₃. Our calculations predict magnetic ordering in MoPd₃ and we calculate the magnetic moment per unit cell of this compound.

1. Introduction

Recently Nautiyal and Auluck (1989) have studied the possibility of magnetic order in MoPd₃, which is a transition-metal compound with non-magnetic constituents. They performed paramagnetic self-consistent band calculations using the linear muffin-tin orbital (LMTO) method (Andersen 1975, Skriver 1984). Applying the Stoner criterion for magnetism in compounds (Jarlborg and Freeman 1980, Stoner 1939), they predicted magnetic ordering in the compound MoPd₃.

So, Nautiyal and Auluck (1989) performed a qualitative analysis of the magnetic properties of MoPd₃. In this work we report the results of a quantitative analysis of the magnetic properties of MoPd₃, i.e. we calculate the magnetic moment per cell of this compound.

There is a method which enables one to calculate values of magnetic moments on different atomic sites in compounds, in the framework of the Stoner model of itinerant ferromagnetism (Krasko 1987). Using this method it was possible to find all magnetic phases of FCC iron, in an easier and cheaper way than in the framework of self-consistent spin-polarised calculations by means of a new fixed spin-moment method (Moruzzi *et al* 1986).

The fixed spin-moment method requires much more computational work than do standard spin-polarised calculations, for a detailed explanation of magnetic properties.

In our previous paper we investigated the magnetic properties of Pd₃Fe using the Stoner (1939) model of magnetism and the method suggested by Krasko (1987), and we obtained very good agreement with the experimental value for the magnetic moment of this compound (Maksimović *et al* 1989). So, here we decided to use the same method for the investigation of MoPd₃, in order to obtain a quantitative picture of the magnetic properties of this compound.

In section 2 we present details of the Stoner approach in combination with self-consistent non-spin-polarised LMTO calculations for compounds. Section 3 gives the results for magnetic properties of MoPd₃.

2. Method of calculation

The Stoner (1939) model for ferromagnetism gives the change ΔE_m in the total energy of the crystal per atom, when the magnetic moment changes from zero to m . This change in energy for MoPd₃ can be obtained from paramagnetic calculations and the change ΔE_m in energy per unit cell is given by (Krasko 1987, Marcus and Moruzzi 1988)

$$\Delta E_m = -\frac{1}{4}Im^2 + \frac{1}{2} \int_0^m \frac{m' dm'}{N(m', s)} \quad (1)$$

The first term in this equation is the exchange energy contribution upon forming a ferromagnetic state with magnetic moment per cell equal to m , and I is the Stoner exchange parameter. The second term is produced by changing the occupation of the single-particle states at $m = 0$, when forming a ferromagnetic state with magnetic moment per cell equal to m . This is achieved by flipping $m/2$ spin-down d electrons from just below the non-magnetic Fermi level ϵ_F , into the non-occupied spin-up states just above the Fermi level. $\overline{N(m, s)}$ is the average density of states (DOS) defined over an energy interval around ϵ_F containing m electrons ($m/2$ above and $m/2$ below ϵ_F). The quantities required for 'constructing' $\overline{N(m, s)}$ are found from the paramagnetic band calculation at the given Wigner-Seitz radius s . The procedure for constructing $\overline{N(m, s)}$ is described in the paper by Marcus and Moruzzi (1988) and in our previous work (Maksimović *et al* 1989), while the condition for the existence of a magnetic phase is obtained by minimising ΔE_m with respect to m :

$$\overline{N(m, s)} = 1/I. \quad (2)$$

The generalised Stoner parameter I for compounds is given by Jaswal (1984):

$$I \approx \sum_t I_t \left(\frac{N_t(\epsilon_F)}{N(\epsilon_F)} \right)^2 \quad (3)$$

where $N_t(\epsilon_F)$ is the non-magnetic DOS at the Fermi energy ϵ_F on site t , and $N(\epsilon_F)$ is the total DOS at ϵ_F .

I_t is given by Gunnarsson (1976):

$$I_t = -\frac{1}{6\pi} \int_0^s \mu^x(r_s) \delta(r_s) \frac{\varphi_d^4(r, \epsilon_F) r^2}{\rho(r)} dr \quad (4)$$

and this is the Stoner parameter for d orbitals $\varphi_d(r, \epsilon_F)$, which dominate the DOS at the Fermi level at site t . $\rho(r)$ is the electron density at site t .

$\mu^x(r_s)$ and $\delta(r_s)$ are expressed in terms of the electron parameter r_s :

$$\delta(r_s) = 1 - 0.036r_s - 1.36r_s/(1 + 10r_s)$$

$$\mu^x(r_s) = -2/\pi\alpha r_s \quad \alpha = (4/9\pi)^{1/3}$$

with

$$4\pi r_s^3 a_B^3 / 3 = 1/\rho(r).$$

As we mentioned above, the magnetisation satisfies the Stoner equation (2). Intersection

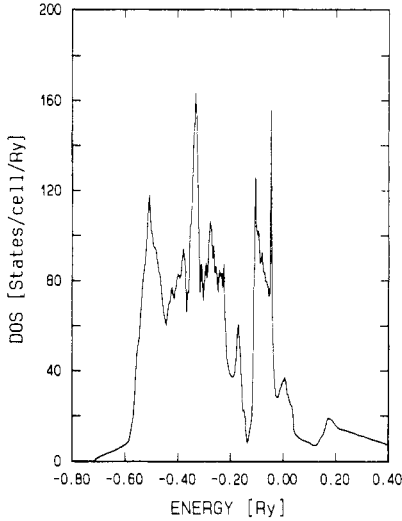


Figure 1. DOS for $s = 2.8732$ au for MoPd₃ versus energy.

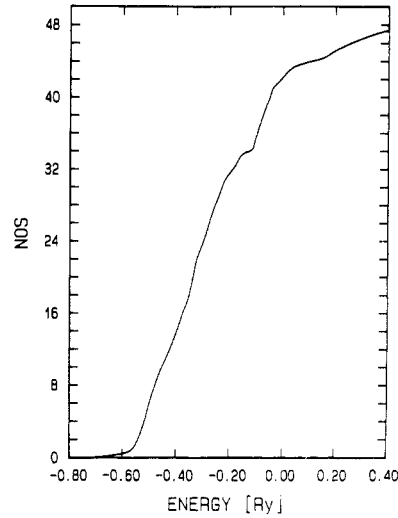


Figure 2. NOS for $s = 2.8732$ au for MoPd₃ versus energy.

of the curve $\overline{N(m, s)}$ with the horizontal line $1/I$ gives all stationary solutions for a given Wigner–Seitz radius s . The corresponding magnetic state is stable

$$(\partial^2(\Delta E_m)/\partial m^2 > 0)$$

if

$$\partial \overline{N(m, s)}/\partial m < 0. \quad (5)$$

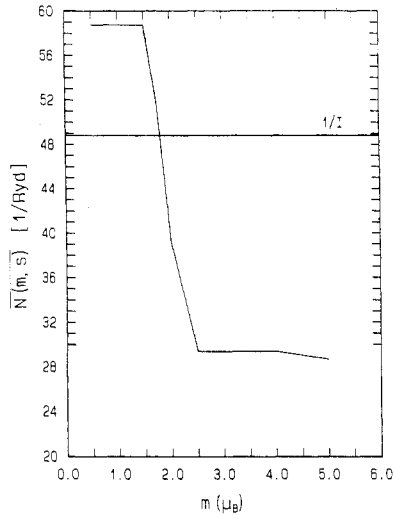
3. Results

MoPd₃ exists in the cubic Cu₃Au structure (like MnPd₃ (Cable *et al* 1962)). The experimental lattice parameter for this compound is $a = 3.89$ Å. We have used the LMTO method in the atomic-sphere approximation with no combined correction terms (Andersen 1975, Skriver 1984). The calculations are semi-relativistic in nature where mass-velocity and Darwin terms are included in the solution of the Dirac equation but with the spin-orbit terms excluded. The exchange-correlation potential used in calculations is the von Barth–Hedin potential. Our calculations have been done with Skriver's computer code. The spheres around the atoms were taken to be of equal size with their radii equal to $s = 2.8732$ au. We have used 220 k-points in the irreducible part of the Brillouin zone. The total DOS and number of states (NOS) of paramagnetic MoPd₃ are presented in figures 1 and 2 ($\epsilon_F = -0.0906$ Ryd).

When self-consistency was achieved, we extracted the d functions at the Fermi level and electron density at the Mo and Pd sites, for the calculation of the Stoner parameter according to equations (3) and (4). Values of the partial DOS $N_i(\epsilon_F)$ and the total DOS

Table 1. The partial DOS $N_i(\epsilon_F)$ and total DOS $N(\epsilon_F)$ at the Fermi level, the Stoner parameter I_i at Mo and Pd sites and the Stoner parameter I for MoPd₃.

	$N_i(\epsilon_F)$ (states Ryd ⁻¹ /cell)	$N(\epsilon_F)$ (states Ryd ⁻¹ /cell)	I_i (Ryd)	I (Ryd)
Mo	61.93	103.705	0.050 570 9	0.020 460 79
Pd	13.93		0.044 877 3	

**Figure 3.** Average DOS $\overline{N(m, s)}$ for MoPd₃ for $s = 2.8732$ au versus magnetic moment per cell. The ferromagnetic phase corresponds to intersection of $1/I$ with $\overline{N(m, s)}$.

$N(\epsilon_F)$ at the Fermi level are given in table 1. Table 1 also presents the Stoner parameters calculated at the Mo and Pd sites and the Stoner parameter I for MoPd₃. We obtained $IN(\epsilon_F) = 1.06 > 1$, implying magnetic ordering in MoPd₃ in agreement with the prediction of Nautiyal and Auluck (1989), but our value for the Stoner parameter is twice greater than that in the work of Nautiyal and Auluck (1989), because we used the expression for the Stoner parameter according to Gunnarsson (1976) (see note 27 in the work of Janak (1977)). In the Stoner criterion ($IN(\epsilon_F) > 1$) we used the DOS $N(\epsilon_F)$ at the Fermi level calculated for one spin (this value is twice smaller than that for $N(\epsilon_F)$ in table 1, because in table 1 we give the DOS at the Fermi level for both spins). Also, we calculate $\overline{N(m, s)}$ for one spin, while Nautiyal and Auluck (1989) calculated $N(\epsilon_F)$ for both spins. Because of that, Nautiyal and Auluck (1989) used the Stoner parameter according to Jaswal (1984), which is half that in our work since the product $IN(\epsilon_F)$ must be invariant according to the definition of density of states.

From figure 3, as described in section 2, we obtained for MoPd₃ a stable magnetic state with a magnetic moment m per unit cell of $1.8\mu_B$. This value of magnetic moment per cell is obtained on the assumption of ferromagnetic ordering, according to the Stoner theory. However, our preliminary results from spin-polarised LMTO calculations suggest opposite orientation of magnetic moments at Mo and Pd sites (ferrimagnetic ordering). In the case of ferrimagnetism, the Stoner model is still useful for calculating magnetic moments at different atomic sites. Using the partial DOS at Mo and Pd sites (figures 4

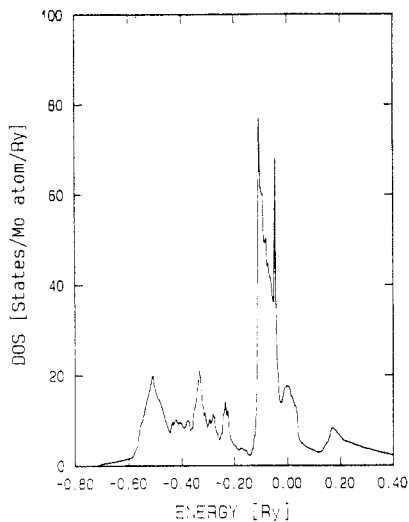


Figure 4. Partial DOS for Mo for $s = 2.8732$ au versus energy.

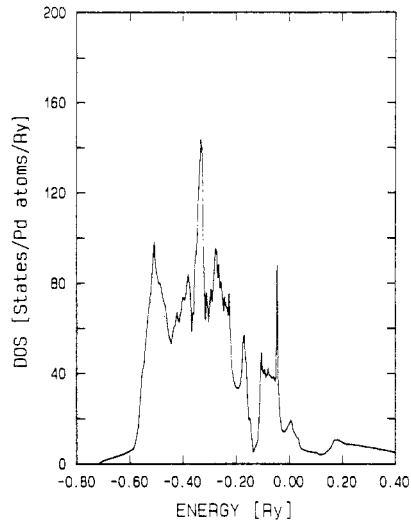


Figure 5. Partial DOS for Pd for $s = 2.8732$ au versus energy.

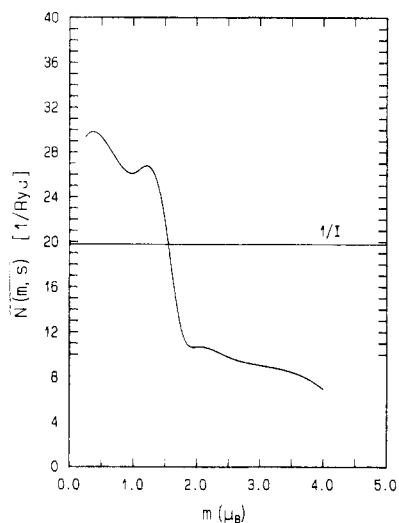


Figure 6. Average DOS $\overline{N(m, s)}$ for Mo atom for $s = 2.8732$ au versus magnetic moment m per atom. The magnetic phase corresponds to intersection of $1/I_{\text{Mo}}$ with $\overline{N(m, s)}$.

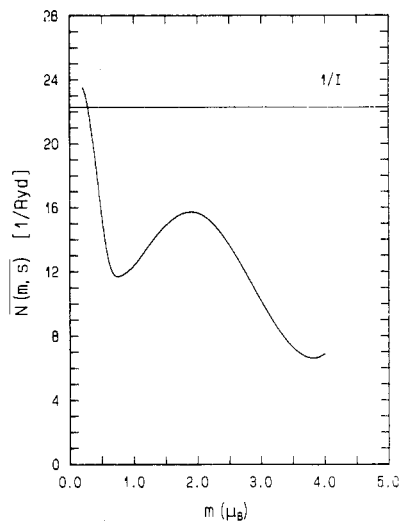


Figure 7. Average DOS $\overline{N(m, s)}$ for Pd atoms for $s = 2.8732$ au versus magnetic moment m per atom. The magnetic phase corresponds to intersection of $1/I_{\text{Pd}}$ with $\overline{N(m, s)}$.

and 5), we obtained, in the same way as described in section 2, magnetic moments $1.55\mu_{\text{B}}$ and $0.25\mu_{\text{B}}$ for Mo and Pd (for all three atoms in a cell), respectively (figures 6 and 7). So, in the case when MoPd_3 is a ferrimagnetic compound, we predict that the magnetic moment per cell is $1.3\mu_{\text{B}}$. Because of that, it would be very interesting to investigate experimentally the magnetic properties of MoPd_3 .

Our investigation of MoPd₃ can be understood as our contribution to the use of our understanding of transition-metal magnetism in the 'design' of new magnetic compounds composed of non-magnetic elements. The fundamental difficulty in this context is that the d states of the 3d transition elements are just barely localised enough to produce magnetism and the reduced localisation of the 4d and 5d transition elements reduces both the intra-atomic exchange interaction and the state density, the two essential ingredients of itinerant magnetism. Nonetheless, the intra-atomic exchange integrals of the 4d transition elements (Janak 1977) are only about 30% smaller than those of the 3d magnets; so we might hope to devise crystal structures that produce state densities that are large enough to compensate for the smaller exchange integrals. The compound MoPd₃ is a successful example of this idea, according to our investigations and the investigations of Williams *et al* (1982). Williams *et al* (1982) have performed self-consistent spin-polarised augmented-spherical-wave band calculations and they obtained that the density of d states in MoPd₃ indicates Stoner (approximately rigid-band) ferromagnetism. Their calculations predict that, if MoPd₃ is prepared in the Cu₃Au (FCC) structure, this compound will constitute a magnetic compound comprised of non-magnetic constituents. In their calculations they obtained that the magnetic moments for Mo and Pd sites are $1.7\mu_B$ and $0.1\mu_B$, respectively, which is in agreement with our calculations.

To sum up, we apply the Stoner model of ferromagnetism in combination with self-consistent LMTO calculations in order to investigate the magnetic properties of MoPd₃. Using the Stoner criterion for compounds we obtained that magnetic order in MoPd₃ exists and we calculated the magnetic moment per cell and the magnetic moments on Mo and Pd sites.

Acknowledgments

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