MAGNETISM IN ACTINIDE TRANSITION METAL INTERMETALLICS

O. Eriksson (1), M. S. S. Brooks (2) and B. Johansson (1)

(1) Condensed Matter Theory Group, Department of Physics, University of Uppsala, Box 530, S-751 21 Uppsala, Sweden
(2) Commission of the European Communities, Joint Research Centre, European Institute for Transuranium Elements, Postfach 2340, D-7500 Karlsruhe, F.R.G.

Abstract. – Energy band calculations of the ground state properties of compounds in the range UM2 – PuM2 (where M = Mn, Co, Fe, Ni) have been made. The Stoner criterion for the ferromagnetic instability is in good agreement with experiment for the uranium compounds, but spin-orbit coupling induces a large orbital moment in UFe2. In PuFe2 and PuCo2 the effects of spin-orbit coupling are even larger, leading to suppression of magnetism in PuCo2.

UFe2 is magnetic with a moment of 1.09 \(\mu_B/\text{f.u.}\) [1] situated mainly on the Fe site, whereas UCo2 is an enhanced paramagnet [2] and UNi2 has a small moment on the U site [1]. The simplest model [3] of the magnetism in this series assumes charge transfer to the transition metal 3d bands from uranium and a reduction of the 3d moment due to the filling of the 3d bands between Fe and Ni. Recent self-consistent band structure studies [4] have shown that the situation is more complex: there is strong hybridization between transition metal 3d and uranium 5f-6d bands, and it would be more accurate to say that it is the bonding bands that fill. In this way the absence of a moment in UCo2 and its presence in UFe2 can be explained [5] by multi-band Stoner theory, as can the anti-parallel alignment of conduction electron contributions to the moment at the Fe and U sites [4].

There is, however, a more unexpected aspect of the magnetism that is revealed experimentally by the observation of anisotropic spin fluctuations [2] in UCo2, and magnetic anisotropy and magnetostriction [6] in UFe2. These properties arise from the large spin-orbit coupling at the uranium sites. Self-consistent band calculations that include spin-orbit coupling yield spin and induced orbital components of the total moment that are anti-parallel and almost equal in UFe2, hence the very small uranium moment [4]. The resulting anomalous uranium form factors in UFe2 and UNi2 have both been observed [7, 8]. The magnetic anisotropy of the uranium should therefore be large and, through 3d-5f exchange interactions, the Fe sites should also have relatively large magnetic anisotropy. In cubic UFe2 this large intrinsic anisotropy is cancelled by the anisotropy due to magnetostriction [6], but we are led by these considerations to consider the effects of spin-orbit coupling in other actinide intermetallics.

NpFe2, NpNi2 and PuFe2 are ferromagnets whereas PuCo2 and PuNi2 are paramagnets and NpCo2 a very weak anti-ferromagnet [1]. The results of earlier relativistic energy band calculations for actinide NaCl-type compounds showed that spin-orbit coupling produces a dip in the density of states when the number of 5f-electrons approaches 6 per atom, corresponding approximaely to the filling of 5f\(_{\frac{1}{2}}\)-bands, which may even produce a semi-conductor [9]. Such a dip could inhibit magnetism, therefore we have studied the magnetic instability in the relativistic case.

We first made self-consistent Linear Muffin Tin Orbital (LMTO) calculations [10] for the compounds PuFe2 and PuCo2 followed by self-consistent fully relativistic (RLMTO) calculations [11] for the same compounds using throughout the functional form of the exchange-correlation energy derived by von Barth and Hedin [12]. The results are summarized in table I. It is immediately noticeable that the state densities at the Fermi level are reduced in the fully relativistic calculations. We concentrate henceforth on this aspect of the problem and note that, since the \(z\)-component of the spin is not a good quantum number in such calculations, the normal Stoner criterion cannot be used to describe the ferromagnetic instability. This is easily seen if the uniform susceptibility is written in terms

| Table I. – Experimental and theoretical volumes, magnetic moments and Stoner products (fully relativistic and scalar relativistic) for PuFe2, PuCo2, YRh3, Pu(fcc) and Np(fcc). Normal multiband Stoner products are given in parenthesis. |
|---|---|---|---|---|---|
| V (exp) \((\AA^3)\) | 92.9 | 88.8 | 19.2 | 20.0 | 68.0 |
| V (theo) \((\AA^3)\) | 91.0 | 84.5 |   |   |   |
| V (theo) \((\AA^3)\) (Scalar) | 91.4 | 81.5 |   |   |   |
| Mag. Mom. \((\text{pg})\) | \(\mu (\text{Pu}) = 0.1\) | para | para | para | para |
| Mag. Mom. \((\text{pg})\) (theo) | \(\mu (\text{Pu}) = 1.0\) | para | para | para | para |
| Stonerprod. (normal) | 1.7 | 1.3 | 1.2 (1.2) | 0.9 (0.9) | 0.4 (0.4) |
| Stonerprod. (rel) | 1.4 | 0.9 | 0.3 | 0.3 | 0.4 |

Article published online by EDP Sciences and available at http://dx.doi.org/10.1051/jphyscol:19883314
of the unperturbed Green function, $G^0$, and the Pauli matrix, $\sigma^z$, as

$$\chi = -(1/\pi) \text{Im} \text{Tr} \sigma^z G^0 \sigma^z G^0$$

which follows directly from linear response theory. When $\sigma^z$ is diagonal in the unperturbed representation the right hand side of (1) becomes the density of states at the Fermi level, $N(E_F)$. But our problem is less simple because $\sigma_z$ has non-zero interband matrix elements, producing a van Vleck contribution to the uniform susceptibility which is absent in the non-relativistic case.

We investigate by applying a small uniform fictitious field to the paramagnetic system of the converged relativistic problem. The coupling is through the perturbation $H' = -H\sigma^z$, and the partial response of the $\ell$'th orbital at the $Q$'th site in the primitive cell ($i = Q, \ell$) is the moment

$$m_i = \chi_i^0 H.$$  

The uniform susceptibility $\chi^0 = \sum \chi_i^0$. The field is enhanced by exchange interactions [13], the effective field, $K$, being given by

$$K_j = J_j m_j.$$  

Then if it is assumed, following Janak [13], that the atomic moments are related to the total moment, $m$, by

$$m_j = m \chi_j^0 / \chi^0$$  

it is easy to show that the enhanced susceptibility is

$$\chi = \chi^0 / \left(1 - \chi^0 I \right)$$

where the Stoner parameter is given by

$$I = \sum J_i \left(\chi_i^0 / \chi^0 \right)^2.$$  

In practice, we calculate the exchange integrals, $J_i$, from the local spin density approximation in the usual way (4, 5) and the applied field is a spin splitting (transformed into the $jj$-coupled relativistic representation) for which we are able to calculate the response – and hence $\chi^0$ and $\chi_j^0$ – from a single iteration of a fully relativistic energy band calculation. Before calculating the Stoner enhancement of PuFe$_2$ and PuCo$_2$ in the relativistic limit we have tested the theory for a few cases cutting out the spin-orbit coupling artificially. Then the susceptibilities calculated from (1) should equal the state density at the Fermi level and equation (5) should yield the same answers as normal multiband Stoner theory. From table I it becomes obvious that this indeed is so both for Pu(fcc) metal and Np(fcc) metal and the compound YRh$_3$. The results for the fully relativistic calculation of Np(fcc), Pu(fcc), PuFe$_2$ and PuCo$_2$ are shown in table I, from which it is clear that we obtain the magnetic instability correctly.

Acknowledgments

This work was partially supported by the Swedish Natural Science Research Council and The Bank of Sweden Tercentenary Foundation.