

ОБЪЕДИНЕННЫЙ
ИНСТИТУТ
ЯДЕРНЫХ
ИССЛЕДОВАНИЙ
ДУБНА



19/61-78

E17-11340

F-87
2597/2-78

Th.Frauenheim, G.Röpke

MAGNETIC BEHAVIOUR OF THE s-f TWO-BAND
HUBBARD MODEL

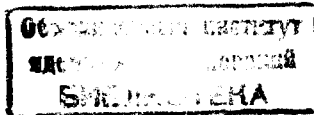
1978

E17 - 11340

Th. Frauenheim, G. Röpke

**MAGNETIC BEHAVIOUR OF THE s-f TWO-BAND
HUBBARD MODEL**

Submitted to "physica status solidi"



Фрауенхайм Т., Репке Г.

E17-11340

Магнитные свойства двухзонной s-f модели Хаббарда

С помощью континуального интегрирования конструируется магнитная фазовая диаграмма двухзонной s-f модели Хаббарда, которую предлагали Жульен и Кодблин для актинидов. Обсуждается прямое обменное взаимодействие между локализованными электронными состояниями, а также не прямое взаимодействие этих состояний через s-электроны проводимости.

Работа выполнена в Лаборатории нейтронной физики ОИЯИ.

Препринт Объединенного института ядерных исследований. Дубна 1978

Frauenheim Th., Röpke G.

E17-11340

Magnetic Behaviour of the s-f Two-Band Hubbard Model

The magnetic phase diagram of a two-band Hubbard Hamiltonian as proposed by Jullien and Coqblin in Actinide Metals is constructed using the functional integral technique. The direct exchange interaction between the localized electron states and the indirect one via the conduction electrons is considered.

The investigation has been performed at the Neutron Physics Laboratory, JINR.

Preprint of the Joint Institute for Nuclear Research.

Dubna 1978

1. Introduction

The conventional Hubbard model^{/1/} describes the physical behaviour of transition metal compounds with strongly isolated small energy bands qualitatively well^{/2/}. Electron correlation effects in small energy bands also are of importance for Rare Earth and Actinide Metals, where near the Fermi energy we have strongly correlated f-electrons as well as delocalized s-d-conduction electrons. In order to explain the magnetic properties of the Actinide Metals Jullien and Coqblin^{/3/} proposed a two-band Hubbard Hamiltonian without hybridization, where intraatomic correlation effects within the different bands as well as between the bands are taken into account.

We will start from a similar model Hamiltonian,

$$H = \sum_{i,j,\nu,\sigma} T_{ij}^{\nu} c_{i\nu\sigma}^{\dagger} c_{j\nu\sigma} + \sum_{i,\nu,\sigma} \frac{U_{\nu}}{2} n_{i\nu\sigma} n_{i\nu-\sigma} + \sum_{i,\sigma,\nu \neq \nu'} \left\{ \frac{U_{\nu\nu'}^{(1)}}{2} n_{i\nu\sigma} n_{i\nu'\sigma} + \frac{U_{\nu\nu'}^{(2)}}{2} n_{i\nu\sigma} n_{i\nu'-\sigma} \right\}, \quad (1)$$

where $c_{i\nu\sigma}^{\dagger}$, $c_{i\nu\sigma}$ and $n_{i\nu\sigma}$ are creation, annihilation or occupation number operators, respectively, for electrons with spin directions σ at lattice point i in

the band labeled by $\nu = s, f$. $\Delta_{\nu} = \left(\frac{1}{N} \sum_{i \neq j} |T_{ij}^{\nu}|^2 \right)^{1/2}$ is the

bandwidth, U_{ν} the intraatomic Coulomb interaction in the corresponding band, $U^{(1)}$ and $U^{(2)}$ describe the spin dependent intraatomic coupling of electrons in two different bands.

In order to discuss the magnetic phase diagram for this model system, the functional integral technique will be used^{/4/}. We are mainly interested in the case $\Delta_s/U_s \gg 1$, $\Delta_f/U_f \ll 1$, where local magnetic moments may arise in the f -band and are coupled directly and via the s -electrons to give a long range ordered state at low temperatures.

Compared with real systems like Rare Earth and Actinide Metals the model Hamiltonian (1) contains the following simplifications:

- (i) No hybridization between the two bands, which only becomes important a secondary fashion, namely between delocalized f -states and d - s -states^{/5/}. In the following, the densities of state of the two bands are characterized only by the position of the band centre $E_\nu = T_{ii}^\nu$ and the bandwidth Δ_ν .
- (ii) No degeneration of the bands. Especially the 7-fold degeneration of the f -states will scale the magnitude of the atomic moments and the magnitude of the coupling between the moments, but we assume that the qualitative picture of the phase diagram described here is not changed if degeneration is taken into account. However, instead of the real electron density we must work with an suitable effective electron density $0 \leq n \leq 2$ for each band, if comparison with real substances is performed. Spin-orbit coupling and crystal field effects are not taken into account.
- (iii) Instead of a rotational invariant exchange term $J S_{s,i} S_{f,i}$ an Ising-type exchange term between the two bands, $U^{(1)} - U^{(2)} = J$, is used. A consideration of the rotational invariant exchange term also is possible within the functional integral technique. However, as is shown in ref.^{/6/}, the qualitative properties considered here are only little affected. To discuss the magnetic properties we will use the one-field scheme to linearize the Coulomb interaction terms.

2. The Functional of Free Energy

We start with the partition function in an external magnetic field

$$Z[B] = \text{Tr} \exp \left\{ -\beta H + \sum_{i,\nu,\sigma} \beta \sigma B_i n_{i\nu\sigma} \right\}, \quad (2)$$

which can be considered as generating functional for the non-local susceptibilities and other physical quantities, see ref.^{/4/}. For Z , an exact expression in form of a functional integral can be found:

$$Z[B] = \int Z_0 \prod_{i,n} dx_{1,i,n} \dots dx_{6,i,n} \exp \left\{ -\pi \sum_{i,n,\lambda=1}^6 x_{\lambda,i,n}^2 \right\} * \\ * \exp \left\{ \text{Tr} \ln(1 - v_1 G^{\text{of}}) + \text{Tr} \ln(1 - v_2 G^{\text{of}}) \right\} + \\ + \text{Tr} \ln(1 - v_3 G^{\text{os}}) + \text{Tr} \ln(1 - v_4 G^{\text{os}}) \quad (3)$$

with

$$(G^{\text{os}})_{ij,nn'}^{\nu\nu',\sigma\sigma'} = \frac{1}{N} \sum_{\vec{k}} \frac{e^{i\vec{k}(\vec{R}_i - \vec{R}_j)}}{i\omega_n - \beta(E_{\vec{k},\nu} - \mu)} \delta_{n,n'} \delta_{\nu,\nu'} \delta_{\sigma,\sigma'}$$

and

$$v_{ij,nn'}^{\nu\nu',\sigma\sigma'} = \begin{cases} (-c_f x_{1,i,n-n'} - c_1 x_{3,i,n-n'} - c_2 x_{5,i,n-n'} - \beta B_i) \delta_{ij} \delta_{\nu\nu'} \delta_{\sigma\sigma'} \\ (\nu=f, \sigma=+) \\ (c_f x_{1,i,n-n'} - c_1 x_{4,i,n-n'} - c_2 x_{6,i,n-n'} + \beta B_i) \delta_{ij} \delta_{\nu\nu'} \delta_{\sigma\sigma'} \\ (\nu=f, \sigma=-) \\ (-c_s x_{2,i,n-n'} + c_1 x_{3,i,n-n'} + c_2 x_{6,i,n-n'} - \beta B_i) \delta_{ij} \delta_{\nu\nu'} \delta_{\sigma\sigma'} \\ (\nu=s, \sigma=+) \\ (c_s x_{2,i,n-n'} + c_1 x_{4,i,n-n'} + c_2 x_{5,i,n-n'} + \beta B_i) \delta_{ij} \delta_{\nu\nu'} \delta_{\sigma\sigma'} \\ (\nu=s, \sigma=-) \end{cases}$$

$$c_\nu = (2\pi\beta U_\nu)^{1/2}, \quad c_\lambda = (2\pi\beta U^{(\lambda)})^{1/2} \quad (4)$$

Z_0 is the partition function for the system without Coulomb interactions. $x_{k,i,n}$ are the fourier components of the time-dependent fictitious fields, see ref.^{/4/}. Because of the six Coulomb terms at each lattice site, we must for linearizing introduce six fictitious fields x_1, \dots, x_6 at each lattice site. However, as follows from (3) and (4) the number of fictitious fields can be reduced by a suit-

able transformation within the six dimensional space of fictitious fields x_{1i}, \dots, x_{6i} so that the v_{1i}, v_{2i}, v_{3i} appear as a part of a new (nonorthogonal) vector base. The other fields can be chosen to be orthogonal and thus are eliminated performing the corresponding integrations. The final result of this transformation will be given here only for the case $(U^{(1)} - U^{(2)})^2 \ll U_s \cdot U_f$.

$$Z[B] = Z_0 \int \prod_{i,n} dv_{f,i,n} dv_{s,i,n} \exp\{-\beta F[v_f, v_s, B]\},$$

$$\beta F[v_f, v_s, B] = 2\pi \left\{ \frac{v_f^2}{c_f^2} + \frac{v_s^2}{c_s^2} + \frac{4(c_1^2 - c_2^2)}{c_f^2 c_s^2} v_f v_s \right\} -$$

$$-\sum_{\sigma} \text{Tr} \ln [1 - \sigma (v_f^{\sigma} - \beta B) G^{\text{of}\sigma}] -$$

$$-\sum_{\sigma} \text{Tr} \ln [1 - \sigma (v_s^{\sigma} - \beta B) G^{\text{os}\sigma}]. \quad (5)$$

In order to evaluate the functional integral we perform the following approximations:

- (i) In the static approximation in the trace all non diagonal terms in n are neglected.
- (ii) A cluster expansion of F is performed and we restrict only to single center and two center part, see ref. 7.
- (iii) A Lorentzian one, respectively, a squared Lorentzian density of state has been used to calculate F_1 and F_2 .

We arrive at the following expressions:

$$\beta F[v_f, v_s, B] = \sum_i \beta F_1^f(v_{fi}, B_i) + \sum_i \beta F_1^s(v_{si}, B_i) +$$

$$+ \sum_i \beta F_1^{sf}(v_{si}, v_{fi}, B_i) + \sum_{\langle ij \rangle} \beta F_2^f(v_{fi}, v_{fj}) +$$

$$+ \sum_{\langle ij \rangle} \beta F_2^s(v_{si}, v_{sj}) \quad (6)$$

with

$$\beta F_1^f[v_f, B, \mu - E_f] = \frac{2\pi}{c_f^2} (v_f + \beta B)^2 +$$

$$+ \frac{\beta \Delta_f}{2\pi} \ln \frac{(\mu - E_f - v_f/\beta)^2 + \Delta_f^2}{(\mu - E_f)^2 + \Delta_f^2} + \frac{v_f}{\pi} \arctan \frac{\mu - E_f - v_f/\beta}{\Delta_f} +$$

$$+ \frac{\beta \Delta_f}{2\pi} \ln \frac{(\mu - E_f + v_f/\beta)^2 + \Delta_f^2}{(\mu - E_f)^2 + \Delta_f^2} - \frac{v_f}{\pi} \arctan \frac{\mu - E_f + v_f/\beta}{\Delta_f},$$

$$\beta F_1^{sf}[v_f, v_s, B] = \frac{8\pi}{c_s^2 c_f^2} (c_1^2 - c_2^2) (v_f + \beta B)(v_s + \beta B),$$

$$\beta F_2^f(v_{fi}, v_{fj}) = \frac{1}{2\pi} \frac{\Delta_f^2}{\beta U_f^3} v_{fi} v_{fj},$$

$$\beta F_2^s(v_{si}, v_{sj}) = \frac{\Lambda_s^3}{2\pi \beta} \frac{(\Delta_s^2/3 - (\mu - E_s)^2)}{(\Delta_s^2 + (\mu - E_s)^2)^3} v_{si} v_{sj}.$$

The discussion of the one center part F_1^f of the free energy functional yields the condition for the occurrence of local magnetic moments. For a single band Hubbard model this problem has been discussed in ref. 8.

3. Construction of the Magnetic Phase Diagram

The expression (6) for the free energy functional can be given in a very transparent form in the case $\frac{U_s}{\Delta_s} \ll 1, \frac{U_f}{\Delta_f} \gg 1$ which is of special interest. Then we have for F_1

$$\beta F_1[v_f, v_s, B] = \frac{2\pi}{c_f^2} v_f^2 - |v_f| + \frac{2\pi}{c_s^2} v_s^2 + \frac{8\pi(c_1^2 - c_2^2)}{c_s^2 c_f^2} v_s v_f.$$

At low temperatures the integrals over v_f , v_s can be evaluated around the minima of the free energy functional F_1 . These minima are situated at

$$v_f^o = \frac{c_f^2}{4\pi}, \quad v_s^o = -\frac{c_1^2 - c_2^2}{2\pi}$$

and

$$v_f^o = -\frac{c_f^2}{4\pi}, \quad v_s^o = \frac{c_1^2 - c_2^2}{2\pi}.$$

The minima are connected with the local magnetic moment in the local approximation for F_1 . Due to (7), the moments are not purely of f -character, but due to the s - f -Coulomb interaction an s -component is mixed in.

If we consider the case that the lower f -subband lies below, the other above the Fermi level, we have the case $n_f = 1$. The number n_s of electrons in the s -band is determined by the chemical potential to be

$$n_s = 1 + \frac{2}{\pi} \arctan \frac{\mu - E_s}{\Delta_s},$$

see ref. /8/.

In order to obtain the long range order at least we must discuss the two center part F_2 of the free energy functional. Singularities in the q -dependent static susceptibility $\chi(0, q)$ indicate phase transition to long range ordered phases. The nonlocal static susceptibility $\chi_{\ell m}(0)$ can be derived from the generating functional (2),

$$\chi_{\ell m}(0) = \frac{\mu_B^2}{\beta} \frac{\delta^2 \ln Z[B]}{\delta B_\ell \delta B_m} \Big|_{B=0}$$

$$= \frac{\mu_B^2}{\beta} (4\pi\beta)^2 \left\{ \frac{1}{c_f^4} \langle v_{f\ell} v_{fm} \rangle + \frac{1}{c_s^4} \langle v_{s\ell} v_{sm} \rangle + \frac{2}{c_s^2 c_f^2} \langle v_{f\ell} v_{sm} \rangle \right. \\ \left. + \frac{4(c_1^2 - c_2^2)}{c_s^4 c_f^4} \langle v_{f\ell} v_{fm} + v_{s\ell} v_{sm} + 2v_{f\ell} v_{sm} \rangle \right\}$$

$$- \delta_{\ell m} \left\{ \frac{4\pi\beta^2}{c_f^2} (1 + \frac{2}{c_s^2} (c_1^2 - c_2^2)) + \frac{4\pi\beta^2}{c_s^2} (1 + \frac{2}{c_f^2} (c_1^2 - c_2^2)) \right\}. \quad (8)$$

The calculation of these correlation functions is not directly possible because the integrals at different lattice points are coupled due to F_2 . In ref. /9/ a diagram technique has been given to evaluate these coupled integrals. In the model considered here the result also can be represented by diagrams if expanding with respect to F_2 . In general, partial summation of geometrical series are not possible, but approximately we get for the susceptibility an expression with a numerator (z - nearest neighbours)

$$1 + z\lambda_s M_{0,2} + z\lambda_f M_{2,0} \quad (9)$$

for the ferromagnetic (FM) susceptibility ($q=0$) and

$$1 - z\lambda_s M_{0,2} - z\lambda_f M_{2,0} \quad (10)$$

for the antiferromagnetic (AFM) susceptibility, where we introduce the moments

$$M_{n,m} = \frac{\int dv_s dv_f v_f^n v_s^m e^{-\beta F_1^f[v_f] - \beta F_1^s[v_s] - \beta F_1^{sf}[v_s, v_f]}}{\int dv_s dv_f e^{-\beta F_1^f[v_f] - \beta F_1^s[v_s] - \beta F_1^{sf}[v_s, v_f]}} \quad (11)$$

Expressions (9) and (10) explicitly show that two different mechanisms act for the onset of the long range order:

- (i) The direct exchange between the f -electrons, which is determined by λ_f .
 - (ii) The indirect exchange between the localized f -states via the s -electrons determined by λ_s .
- Using the expressions for λ_s, λ_f we get the following result:

The half-filled f -band favours the antiferromagnetic order of the moments.

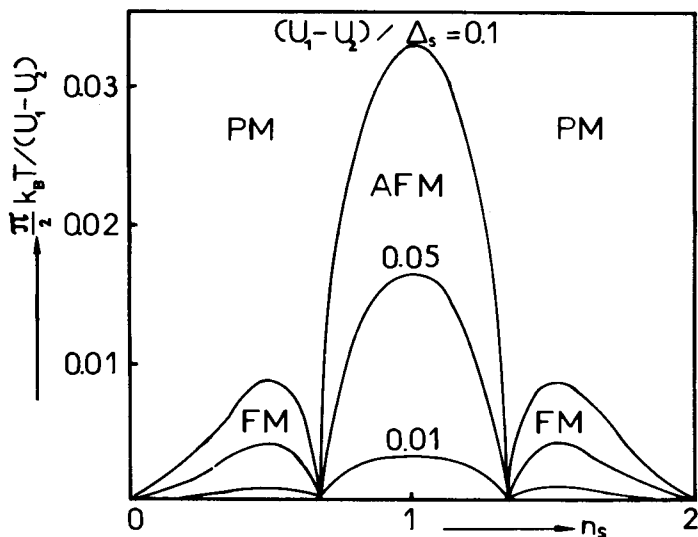
The s -band tends, in dependence on the chemical potential, to an antiferromagnetic order if $|\mu - E_s| < \Delta_s / \sqrt{3}$ or to a ferromagnetic order if $|\mu - E_s| > \Delta_s / \sqrt{3}$. In the latter case there is a concurrence between the direct and the indirect coupling mechanism.

4. Discussion

A phase diagram which is obtained by determining the zeros of (9) and (10) is presented in the figure for $(U^{(1)} - U^{(2)}) / \Delta_s = 0.01, 0.05, 0.10$.

We make the following conclusions:

If $|n_s - 1| > 1/\sqrt{3}$, the indirect exchange interaction via the s -electrons gives a ferromagnetic state. The occurrence of the ferromagnetic state is influenced by the magnitude



Magnetic phase diagram for the indirect coupling of localized f -electrons via the conduction electrons for various parameters $(U^{(1)} - U^{(2)}) / \Delta_s$.

of the direct interaction which tends to an antiferromagnetic state.

We can compare the qualitative features of this model with the properties of Rare Earth and Actinide Metals. It is well known from the properties of Rare Earth Metals, that if we go from the heavier to the lighter Rare Earth Metals the tendency from the ferromagnetic state to the antiferromagnetic ordering is obtained. This effect qualitatively can be described in our model because the bandwidth Δ_f of the $4f$ -electrons is increasing in this direction, but remains small compared with U_f , so that local $4f$ -moments can occur for low temperatures^{/5/}. For the light Rare Earth Metals so the direct exchange between the localized moments becomes more important. The s - d -band for Rare Earth Metals can be filled up to 12 electrons per atom, but in our case we only have three electrons per atom. That means, the s - d -band is $1/4$ -filled and for this electron concentration of the conduction band, see the figure, for heavy Rare Earth Metals a ferromagnetic coupling can occur at low temperatures.

For the light Actinide Metals the $5f$ -electrons are delocalized^{/11/} and the relation of $5f$ -bandwidth to the correlation strength U_f in the $5f$ -band is so big that no long range order can occur^{/8/} and paramagnetic behaviour (PM) is obtained. Following from refs.^{/10,11/}, the heavy Actinides, starting with Plutonium, can be considered as a second delayed Rare Earth series with strongly localized character of the $5f$ -electrons. For decreasing atomic number in these heavy Actinide Metals one also can describe the same tendency from ferromagnetic to antiferromagnetic ordering, as it is the case for Rare Earth Metals.

The direct antiferromagnetic coupling for half-filled f -bands in this direction dominates the indirect ferromagnetic coupling for the $1/4$ -filled conduction bands. A more detailed description of conduction electrons in this model also will give the Ruderman-Kittel coupling between localized spins, see ref.^{/12/}.

However, we only would give here a qualitative picture of the two-band Hubbard model in order to show, how is

within this model the role of Coulomb interactions between the different bands as it occurs in Rare Earth and Actinide Metals.

References

1. Hubbard J. *Proc.Roy.Soc. (London)*, 1963, A276, p.238.
2. Mc Whan D.B. et al. *Phys.Rev.*, 1973, B7, p. 1920.
Röpke G., Albani B., Schiller W. *phys.stat.sol.*, 1975, (b)69, p. 407.
3. Coqblin B. et al. *Proc. 2nd Int.Conf.Electr.Str.Actin.*, 1976, Wroclaw, Poland.
Iglesias-Sicardi I.R. et al. *Journ. of Low Temp.Phys.*, 1976, 25, p. 871.
4. Schrieffer J.R., Evanson W.E., Wang S.Q. *C.A.P. Summer School, Banff (1969)*.
5. Johansson B. *Phys.Rev.*, 1975, B11, p. 2740.
6. Tyagi J.S., Kishore R., Joshi S.K. *Phys.Rev.*, 1975, B12, p. 3809.
7. Cyrot M. *Phys.Rev.Lett.*, 1970, 25, p. 871.
8. Frauenheim T. *Dissertation, Technical University Dresden (1976)*.
9. Röpke G., Albani B., Schiller W. *phys.stat.sol.*, 1975, (b)69, p. 45.
10. Johansson B., Rosengren A. *Phys.Rev.*, 1975, B11, p. 2836.
11. Freeman A.J., Koelling D.D. *The Actinides: Electronic Structure and Related Properties*, Academic Press, New York (1974).
12. Evanson W.E., Wang S.Q., Schrieffer J.R. *Phys.Rev.*, 1970, B2, p. 2604.

Received by Publishing Department
on February 22, 1978.