ОБЪЕДИНЕННЫЙ
ИНСТИТУТ
ЯДЕРНЫХ
ИССЛЕДОВАНИЙ

Дубна

E4 - 6506

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ЛАБОРАТОРИЯ ТЕОРЕТИЧЕСКОЙ ФИЗИКИ

MAGNETIC PHASE DIAGRAM IN NARROW-BAND MATERIALS

1972

E4 - 6506

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Submitted *to*

physica status solidi (b)

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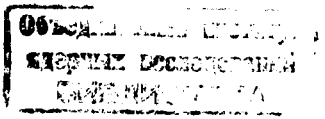

I. Introduction

The simplest model for the strong correlated d-electrons in transition metals, their compounds and alloys is the well-known Hubbard model[1]

$$H_{Hubb} = \sum_{ij\sigma} T_{ij} c^{+}_{i\sigma} c_{j\sigma} + U \sum_{i} n^{d}_{i\uparrow} n^{d}_{i\downarrow} \quad , \; n^{d}_{i\sigma} = c^{+}_{i\sigma} c_{i\sigma} \qquad ,(1)$$

where T_{ij} is the hopping integral and U describes the interaction between electrons at the same lattice site. $c^{+}_{i\sigma}$ ($c_{i\sigma}$) are creation (annihilation) operators for d-electrons with spin σ in a Wannier state at the lattice site i .

Although this model is very simple, it contains several difficulties. Especially, divergencies exist at $U \to \infty$ [2,3] and $n = 1$ [4]. Therefore, the magnetic phase diagram is not completely clarified at present. Furthermore, the neglect of s-bands in (1) can give wrong results, also for empty s-bands. The (virtual) scattering of d-electrons into the wide s-band changes the form of the d-band and the properties of the ground state[5].

Therefore a generalized model must be considered, which contains s- and d- electrons. It can be assumed, that in such a generalized model the divergencies of the operator (1), at least at n = 1, vanish, so that the same approximations can give better results as in the case of the pure Hubbard model.

As a starting point the operator

$$H = H_s^0 + H_d + V_{sd} - \zeta N \qquad (2)$$

is considered. H_s^0 describes the s-electrons by a simple Bloch band,

$$H_s^0 = \sum_{\vec{k}\sigma} \epsilon_{\vec{k}} n^s_{\vec{k}\sigma} \quad , \quad n^s_{\vec{k}\sigma} = a^+_{\vec{k}\sigma} a_{\vec{k}\sigma} \quad . \qquad (3)$$

Herein, $a^+_{\vec{k}\sigma}$ ($a_{\vec{k}\sigma}$) are creation (annihilation) operators of electrons with spin σ in an s-type Bloch state $\vec{k}$ with energy $\epsilon_{\vec{k}}$. The d-electrons are described by the Hubbard operator, $H_d = H_{Hubb}$, and V_{sd} is the interaction between s- and d-electrons in a usual form:

$$V_{sd} = \sum_{i\vec{k}} J_{\vec{k}} \, e^{i\vec{k}\cdot\vec{R}_i} \, \vec{\sigma}_{\vec{k}} \cdot \vec{S}_i \quad , \qquad (4)$$

where $J_{\vec{k}}$ is the s-d exchange integral, $\sigma_{\vec{k}}$ describes the spin density of the s-electrons and $\vec{S}_i$ the spins of d-electrons at the site i . ζ in (2) is the chemical potential, and N the number operator (s- and d-electrons).

Neglecting spin-flip processes and the $\vec{k}$-dependence of J , V_{sd} get the form

$$V_{sd} = - J \sum_i S_i^z \left(n_{i\uparrow}^s - n_{i\downarrow}^s \right) , \qquad (5)$$

where $S_i^z = S \left(n_{i\uparrow}^d - n_{i\downarrow}^d \right)$.

Using the identity

$$2 n_{i\uparrow} n_{i\downarrow} = - \left(n_{i\uparrow} - n_{i\downarrow} \right)^2 + n_{i\uparrow} + n_{i\downarrow} , \qquad (6)$$

H from (2) takes the following form:

$$H = \sum_{\vec{k}\sigma\nu} \left(\epsilon_{\vec{k}}^{\nu} - \zeta \right) n_{\vec{k}\sigma}^{\nu} - \frac{1}{2} \sum_{i\nu} J^{\nu} S_i^z \left(n_{i\uparrow}^{\nu} - n_{i\downarrow}^{\nu} \right) . \qquad (7)$$

Herein $\nu = 1$ or 2 corresponds to s- or d-bands. $\epsilon_{\vec{k}}^2$ is the Fourier transformation of T_{ij} , $J^1 = J$ and $J^2 = U / S$. In the form (7), H describes a two-band model, with a wide band and a narrow one. The limiting cases are

(a) the Hubbard model, where only d-electrons exist, and

(b) a s-d exchange model (in a simple form), where the d-electrons are described only by the spin operators S_i^z .

In the following these limiting cases are considered. Since the results in both cases (a) and (b) are very similar, as is shown in section 3 of this paper (fig. 1 and 2), it can be assumed, that also more realistic cases (with both s- and d-bands) have analogic properties.

2. Variational Ansatz

To solve (7), a variational method is used, basing on the thermodynamic Hartree-Fock approximation. On this purpose, the trial operator /6/

$$\tilde{H} = H_{el} + H_S \tag{8}$$

is considered, where

$$H_{el} = \sum_{\vec{k}\sigma} (\epsilon_{\vec{k}} - \zeta) n_{\vec{k}\sigma} - \frac{\tilde{J}}{2} \sum_i < S_i^z > (n_{i\uparrow} - n_{i\downarrow}) , \tag{9}$$

$$H_S = -\frac{\tilde{J}}{2} \sum_i S_i^z < n_{i\uparrow} - n_{i\downarrow} > ,$$

with $\tilde{J} = U/S$ in case (a) and $\tilde{J} = J$ in case (b). H_{el} and H_S contain average fields, which are used as variational parameters:

$$< S_i^z > = S M_i , \quad < n_{i\uparrow} - n_{i\downarrow} > = \mu_i . \tag{10}$$

Using Bogolubov's theorem

$$F = -\frac{1}{\beta} \ell n \, Tr e^{-\beta H} \leq \tilde{F} = -\frac{1}{\beta} \ell n \, Tr e^{-\beta \tilde{H}} + < H - \tilde{H} > , \tag{11}$$

the variational parameters M_i and μ_i are determined by minimalization of

$$\tilde{F} = F_{el} + F_S + \frac{\tilde{J}S}{2} \sum_i M_i \mu_i + \sum_i \gamma_i (M_i - \mu_i) . \qquad (12)$$

The last term in (12) is caused by the relation between S_i^z and $n_{i\sigma}$, valid in the Hubbard model. Hereby the relations between operators are replaced by the corresponding relations between average fields,

$$< S_i^z > = S < n_{i\uparrow} - n_{i\downarrow} > , \qquad (13)$$

which in (11) are expressed by Lagrange parameters γ_i x/. In the case (b) the condition (13) disappears ($\gamma_i = 0$). If only ferromagnetic (FM), antiferromagnetic (AF) and paramagnetic (PM) states are considered, the ansatz

$$M_i = M \delta_i , \quad \mu_i = \mu \delta_i , \quad \gamma_i = \gamma \delta_i$$

$$\delta_i = \begin{cases} +1 & \text{FM} \\ exp[i \vec{Q} \cdot \vec{R}_i] = \pm 1 \text{ in } \begin{matrix} A \\ B \end{matrix} & \begin{matrix} \text{AF} \\ \text{PM} \end{matrix} \\ \pm 1 \text{ , randomly} & \end{cases} \qquad (14)$$

x/ The replacing of operatos relations by conditions between average fields is in some respect analogous to the introduction of additional free parameters in several approximations, e.g. in Bogolubov's solution of superconductivity and in the random phase approximation.

is possible. The nonmagnetic case, which corresponds to a metallic one[7], follows for $M = \mu = \gamma = 0$. In (14) $\vec{Q}$ is half a vector in the reciprocal lattice, so that $(\vec{R}_i^A - \vec{R}_i^B) \cdot \vec{Q} = \pi$, where $\vec{R}_i^{A(B)}$ is a position vector in the sublattice A(B).

From (12) a coupled system of equations for M , μ and γ follows. In the special case of symmetric density of states ($D(\epsilon) = D(-\epsilon)$) and half-filled band ($n = 1$, $\zeta = 0$) a decoupling is possible, yielding a single equation for the parameter $\Delta = \frac{1}{2} \tilde{J} S M$ in the form ($B_S(x)$ = Brillonin's function)

$$\Delta = \begin{cases} -\frac{\tilde{J}S}{4} \frac{1}{N} \frac{\partial F_{el}}{\partial \Delta} + \frac{\tilde{J}S}{4} B_S(\beta\Delta) & \text{(a)} \\ \text{in case} & (15) \\ \frac{\tilde{J}S}{2} B_S\left(-\frac{\tilde{J}S}{2} \frac{\beta}{N} \frac{\partial F_{el}}{\partial \Delta}\right) & \text{(b)} \end{cases}$$

In (15) for the functions F_{el} the dependence on Δ must be known. These functions are different in the FM, AF and PM case. For simplification the following approximations are used:

a) For the unperturbed band $\epsilon_{\vec{k}}$ the density of states $D(\epsilon) = 2\sqrt{1 - \epsilon^2}/\pi$ is assumed, with the half band width $w = 1$.

b) For $\epsilon_{\vec{k}}$ is assumed $\epsilon_{\vec{k}+\vec{Q}} = -\epsilon_{\vec{k}}$, where $\vec{Q}$ is the same vector as in (14). This form is valid, for instance, in a tight-binding band for sc and bcc lattices.

c) The PM-case is solved using the coherent potential approximation[8], since the CPA seems to be the best approach to the disorder problem.

3. Results

Solving numerically both limiting cases (a) and (b), with $S = 1/2$ and $S = 5$, the free energy of all magnetic phases follows depending on T and $\tilde{J}S/2$. (The numerical calculations are carried out at the CDC-1604A in Dubna). At $T = 0$ (fig. 1) always

$$E^{AF} < E^{PM} < E^{FM} < E^{met} \qquad (16)$$

is valid. That means, that the ground state is always antiferromagnetic. A Mott transition by variation of $\tilde{J}S/2$ does not occur.

Furthermore possible phase transitions of second order ($\Delta \to 0$) are considered, and the corresponding critical temperatures $T_C\,(FM \to met)$, $T_N\,(AF \to met)$ and $T_M\,(PM \to met)$ are numerically calculated depending on $\tilde{J}S/2$. The result is, that always T_N is the highest

critical temperature (fig. 2). That means, that the AF phase is the most stable one.

Therefore in this model also by variation of T a Mott transition does not exist. This result follows in both limiting cases, in the Hubbard model as well as in the s-d model. So it should be concluded that also more realistic cases, in which both s- and d-electrons are considered, do not contain such a transition. To obtain a first order transition from a paramagnetic (isolating) state to a metallic (nonmagnetic) one, more complicated models must be considered.

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Received by Publishing Department
on June 7, 1972.

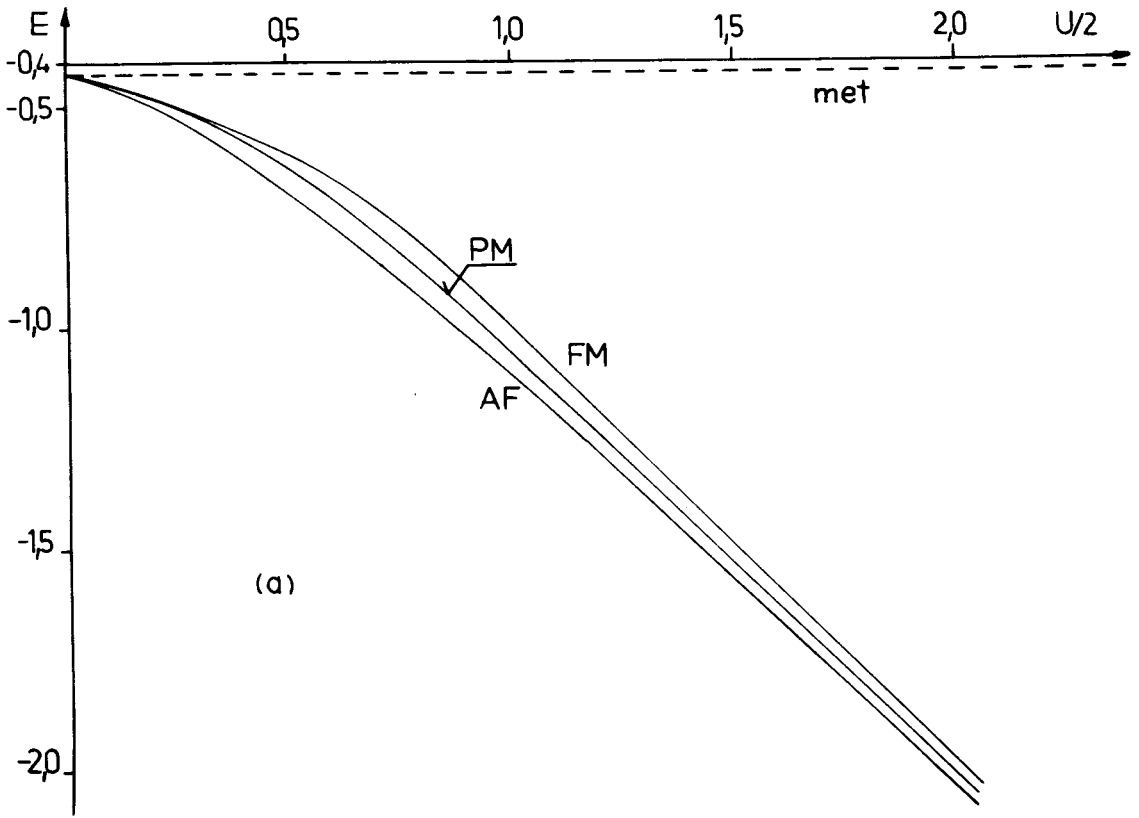

Fig. 1. Energy of the magnetic states at $T = 0$. a) Hubbard model.

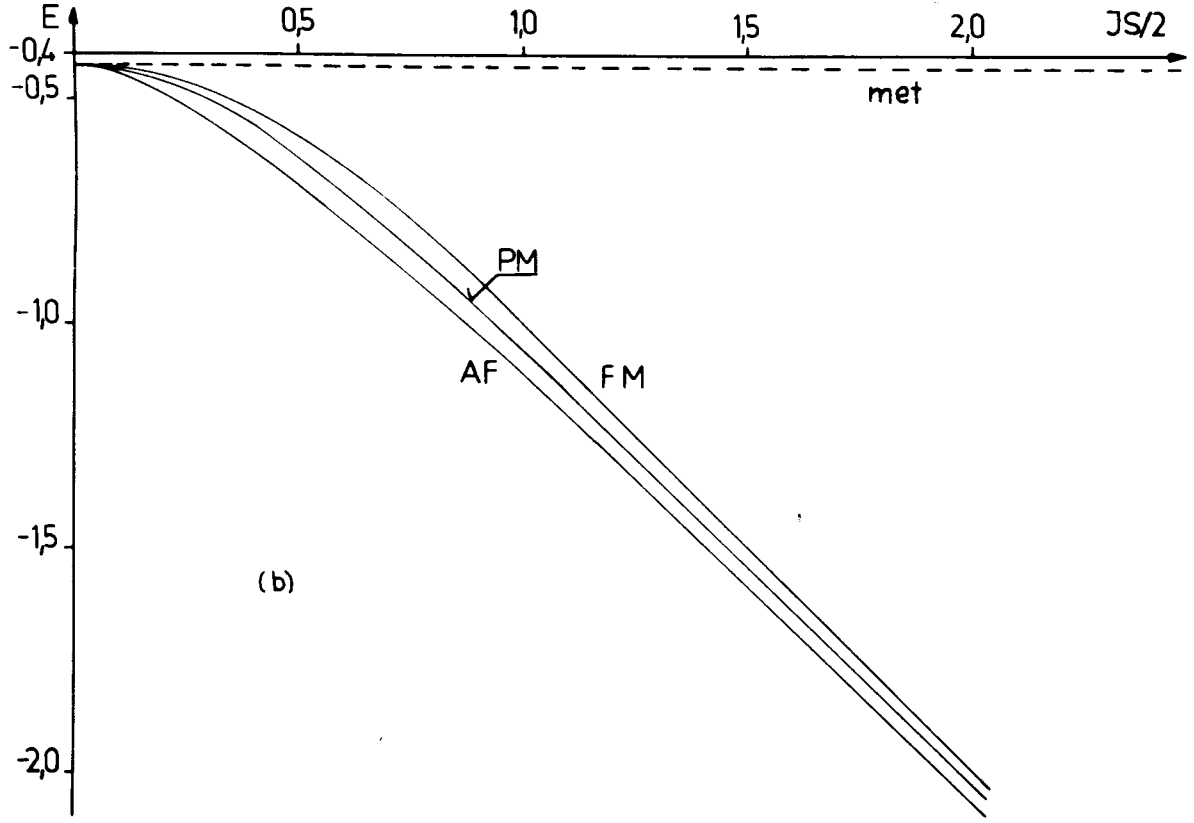

Fig. 1. Energy of the magnetic states at $T = 0$. b) s-d model.

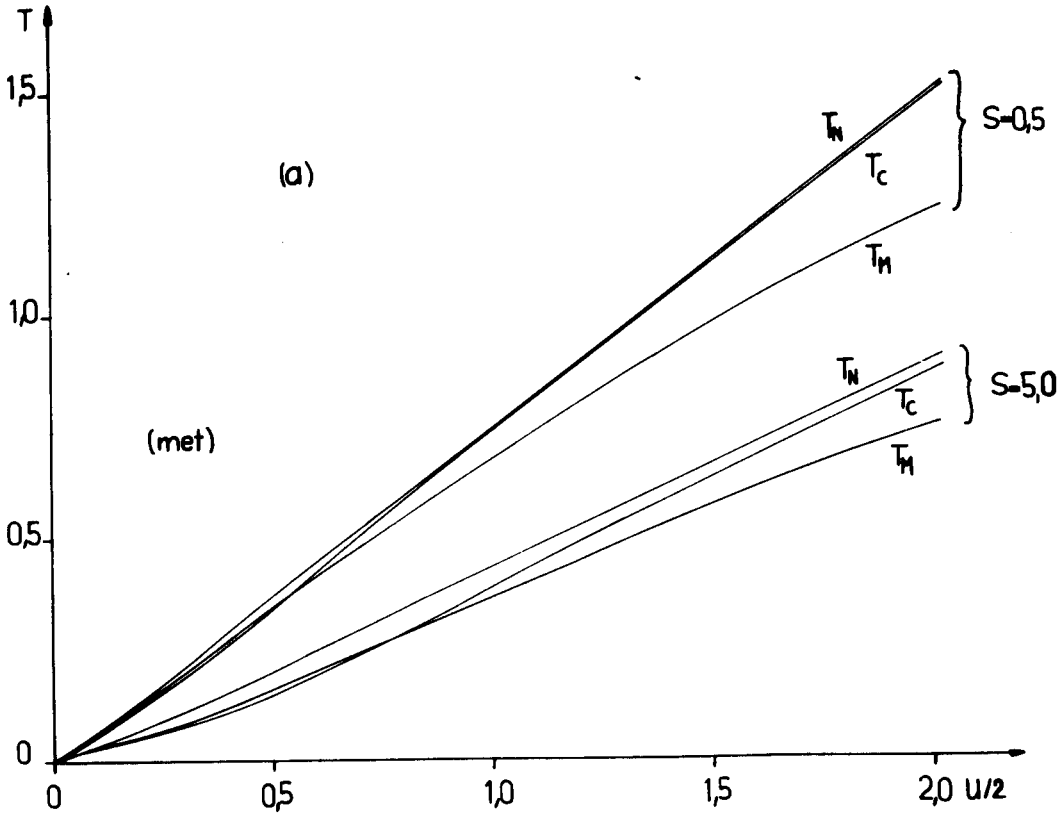

Fig. 2. Phase transitions of second order. a) Hubbard model.

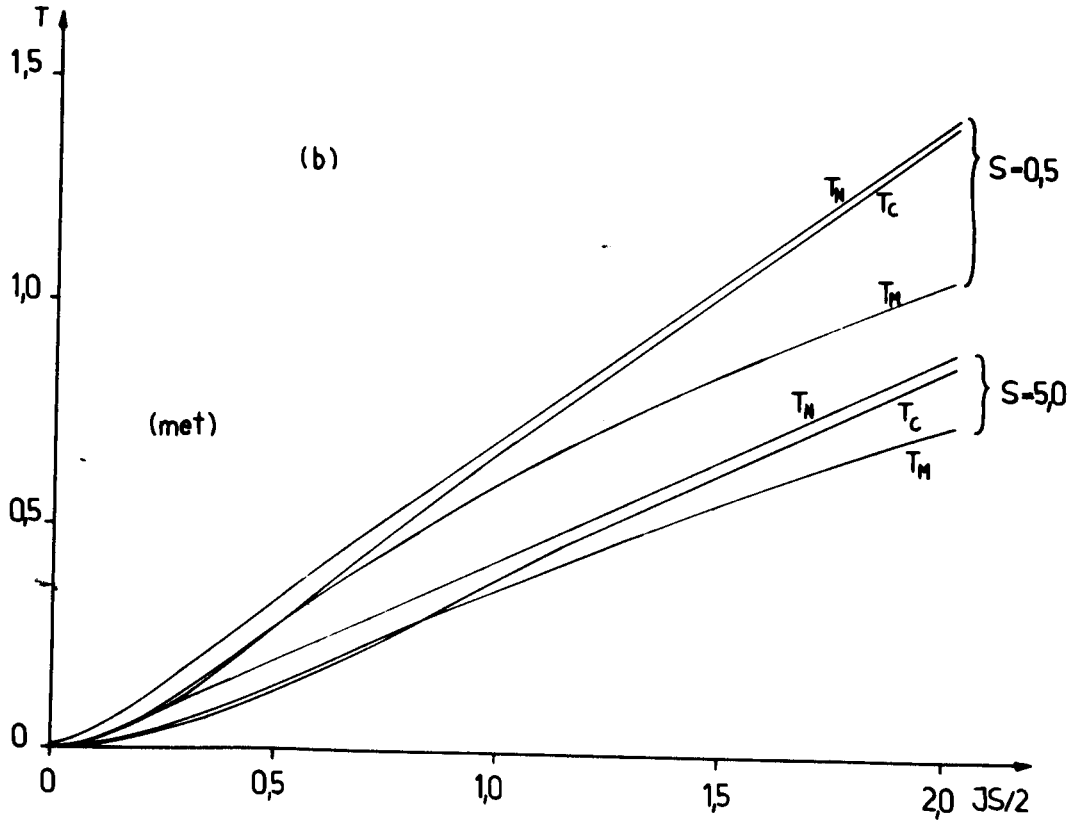

Fig. 2. Phase transitions of second order. b) s-d model.