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THE MAGNETIC SUSCEPTIBILITY OF INTERMEDIATE VALENCE COMPOUNDS IN THE SIMPLIFIED PERIODIC ANDERSON MODEL WITH *f* LEVELS SPLIT BY CRYSTAL FIELD

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The investigation of the exactly solvable limit of the simplified periodic Anderson model with *f* levels split by crystal field is presented. The standard method of Green's functions is applied. The magnetic susceptibility and the occupation number of the *f* electrons versus temperature is obtained. Different types of temperature dependence for magnetic susceptibility can be derived by changing the values of the model parameters. Comments on the relation to experimental data are also given.

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1. *Introduction*

There are several approaches to the problem of electronic properties of intermediate valence compounds (see, e.g. [1, 2] for a review). Some of them are based on the periodic Anderson model, e.g. [3, 4]. In the present paper the $U = 0$ limit of this model, taking into account the crystalline field splitting of *f* levels, is considered. Hund's rule type correlations are also neglected i.e. $J = 0$. Such a simplified approach seems to be reasonable as a first approximation. This is due to the fact that crystalline field effects can be of importance for rare earth ions in intermediate valence compounds [1, 2]. Besides that, because of the absence of magnetic ordering in most of the intermediate valence compounds, the case $U, J = 0$ can serve to estimate the behaviour of magnetic susceptibility for the paramagnetic case.

The main aim of the present paper is to calculate the temperature dependence of the magnetic susceptibility in order to see how the splitting of *f* levels influences the behaviour of this quantity.

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We shall start from the simplified Anderson Hamiltonian in the presence of a magnetic field h

$$H = \sum_{k,\sigma} \varepsilon_{k\sigma} d^+_{k\sigma} d_{k\sigma} + \sum_{\substack{l,\\ i,\sigma}} E_{l\sigma} f^{l+}_{i\sigma} f^{l}_{i\sigma} + \sum_{\substack{l,k\\ i,\sigma}} \frac{V_l}{\sqrt{N}} \left(d^+_{k\sigma} f^{l}_{i\sigma} e^{ikR_i} + f^{l+}_{i\sigma} d_{k\sigma} e^{-ikR_i}\right), \tag{1}$$

where

$$E_{l\sigma} = E_l - \sigma h, \quad l = 1, 2 \tag{2}$$

$$\varepsilon_{k\sigma} = \varepsilon_k - \sigma h, \quad \sigma = \pm\tfrac{1}{2}\,. \tag{3}$$

$d^+_{k\sigma}$ and $d_{k\sigma}$ are creation and annihilation operators for band electrons. For the f levels creation and annihilation operators are denoted by $f^{l+}_{i\sigma}$ and $f^{l}_{i\sigma}$ respectively. The superscript l is introduced to distinguish two different f levels split by the crystalline field. $E_{l\sigma}$ and V_l $l = 1, 2$ describe the positions of the f level and the hybridization between the band and f electrons, respectively. Further, $V_1 = V_2 = V$ will be assumed for simplicity.

2. Calculations

In order to calculate the magnetic susceptibility and the average occupation of f levels we need to calculate the Green's functions [5]. For the Hamiltonian (1) the sequence of equations of motion terminates so it is unnecessary to use any decoupling procedure. For the Lorenzian density of states

$$\varrho_{\mathrm{L}}(\omega) = \frac{1}{\pi} \frac{D}{\omega^2 + D^2} \tag{4}$$

we can find, after simple calculations, the following formula for the total density of states in units of the band half-width D

$$\varrho(\omega) = \frac{1}{D} \times \frac{(\omega - E_{1\sigma})^2 (\omega - E_{2\sigma})^2 + (\omega - E_{2\sigma})^2 V^2 + (\omega - E_{1\sigma})^2 V^2}{[(\omega + \sigma h)(\omega - E_{1\sigma})(\omega - E_{2\sigma}) - (\omega - E_{1\sigma}) V^2 - (\omega - E_{2\sigma}) V^2]^2 + (\omega - E_{1\sigma})^2 (\omega - E_{2\sigma})^2}\,. \tag{5}$$

The three terms in the numerator refer to the band, first and second f levels, respectively. It is useful to approximate the density of states (5) by

$$\bar{\varrho}(\omega) = \frac{1}{D} \times \frac{(\omega - E_{1\sigma})^2 (\omega - E_{2\sigma})^2 + (\omega - E_{2\sigma})^2 V^2 + (\omega - E_{1\sigma})^2 V^2}{(\omega + i)(\omega - i)\left[(\omega - E_{1\sigma})(\omega - E_{2\sigma}) + \frac{V^2}{i}(2\omega - E_{1\sigma} - E_{2\sigma})\right] \times \left[(\omega - E_{1\sigma})(\omega - E_{2\sigma}) - \frac{V^2}{i}(2\omega - E_{1\sigma} - E_{2\sigma})\right]} \tag{6}$$

in order to perform analytical calculations. The above approximation was verified by numerical calculations, the maximum difference between ϱ and $\bar{\varrho}$ is about 1%. The correlation functions of interest

$$\langle n^d_\sigma \rangle = \langle d^+_{i\sigma} d_{i\sigma} \rangle, \qquad \langle n^{f1}_\sigma \rangle = \langle f^{1+}_{i\sigma} f^1_{i\sigma} \rangle, \qquad \langle n^{f2}_\sigma \rangle = \langle f^{2+}_{i\sigma} f^2_{i\sigma} \rangle \tag{7}$$

can be derived from the spectral theorem with the use of (6)

$$\langle n_\sigma \rangle = \langle n^d_\sigma \rangle + \langle n^{f1}_\sigma \rangle + \langle n^{f2}_\sigma \rangle = \frac{1}{\pi} \int\limits_{-\infty}^{+\infty} \bar{\varrho}(\omega) f(\omega) d\omega, \tag{8}$$

where $f(\omega)$ is the Fermi function. The integral (8) can be evaluated by contour integration and our final result reads

$$\langle n_\sigma \rangle = \sum_{j=1}^{3} \left\{ C_j + \frac{2}{\pi} \operatorname{Im} \left[\sum_{r=1}^{3} \psi \left[\tfrac{1}{2} - \frac{i\beta}{2\pi} (\omega_r - \mu) \right] P_{jr} \right] \right\}. \tag{9}$$

The individual contributions to $\langle n_\sigma \rangle$ from band electrons, f^1 and f^2 electrons are given by terms with $j = 1, 2$ and 3, respectively. In (9)

$$C_1 = \sum_{r=1}^{3} \frac{(\omega_r - E_{1\sigma})^2 (\omega_r - E_{2\sigma})^2}{W(\omega_r)} = \sum_{r=1}^{3} P_{1r}, \tag{10}$$

$$C_2 = \sum_{r=1}^{3} \frac{(\omega_r - E_{2\sigma})^2 V^2}{W(\omega_r)} = \sum_{r=1}^{3} P_{2r}, \tag{11}$$

$$C_3 = \sum_{r=1}^{3} \frac{(\omega_r - E_{1\sigma})^2 V^2}{W(\omega_r)} = \sum_{r=1}^{3} P_{3r}, \tag{12}$$

$$\begin{aligned} \omega_1 &= i, \\ \omega_2 &= \omega_N + i\omega_Y, \\ \omega_3 &= \omega_P + i\omega_Y, \\ \omega_Y &= V^2, \end{aligned} \tag{13}$$

$$\begin{aligned} \omega_N &= E_{1\sigma} + E_{2\sigma} + [(E_{1\sigma} - E_{2\sigma})^2 - 4V^4]^{1/2}, \\ \omega_P &= E_{1\sigma} + E_{2\sigma} - [(E_{1\sigma} - E_{2\sigma})^2 - 4V^4]^{1/2} \end{aligned} \tag{14}$$

and

$$\begin{aligned} W(\omega_r) = (\omega_r + i)(\omega_r - i) &\left[(\omega_r - E_{1\sigma})(\omega_r - E_{2\sigma}) + \frac{V^2}{i} (2\omega_r - E_{1\sigma} - E_{2\sigma}) \right] \\ \times &\left[(\omega_r - E_{1\sigma})(\omega_r - E_{2\sigma}) - \frac{V^2}{i} (2\omega_r - E_{1\sigma} - E_{2\sigma}) \right]. \end{aligned} \tag{15}$$

ψ stands for the digamma function ($\beta = (kT)^{-1}$). From (9) we can obtain the magnetic susceptibility which is defined by

$$\chi = \frac{\partial m}{\partial h}\bigg|_{h=0}, \tag{16}$$

where

$$m = 1/2[\langle n_\uparrow \rangle - \langle n_\downarrow \rangle]. \tag{17}$$

We have chosen the units in which the gyromagnetic ratio equals unity. Finally we have

$$\chi = \frac{\beta}{2\pi^2} \sum_{j,r=1}^{3} \left\{ \psi'_{\mathrm{R}} \left[1/2 - \frac{i\beta}{2\pi} (\omega_r - \mu) \right] \mathrm{Re}\, P_{jr} - \psi'_{\mathrm{I}} \left[1/2 - \frac{i\beta}{2\pi} (\omega_r - \mu) \right] \mathrm{Im}\, P_{jr} \right\} \tag{18}$$

μ is determined self-consistently if one fixes the total number of electrons per site, $\langle n_\sigma \rangle = N$ and $\psi'_{\mathrm{R}}(\psi'_{\mathrm{I}})$ stands for the real (imaginary) part of the ψ function derivative (see e.g. [6] for series expansion).

3. *Results*

The occupation number of f levels as a function of temperature is shown in Fig. 1, the temperature is given in units of kD^{-1}. It increases quasi-linearly with temperature. This is typical behaviour for different values of the total occupation number and other parameters of the model E_{16}, E_{26} and V.

In Fig. 2, 3 and 4 we see different types of temperature dependence of the susceptibility. We can make a qualitative comparison of our results to the bahaviour of magnetic sus-

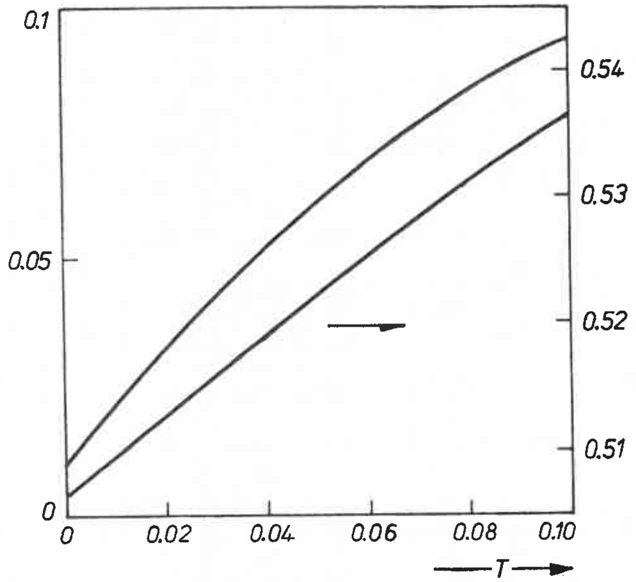

Fig. 1. The occupation number of f levels as a function of temperature for $E_{1\sigma} = -0.02$, $E_{2\sigma} = -0.021$, $V = 0.01$ and for $\langle n_\sigma \rangle = 0.5$ (left scale) and for $\langle n_\sigma \rangle = 1$ (right scale)

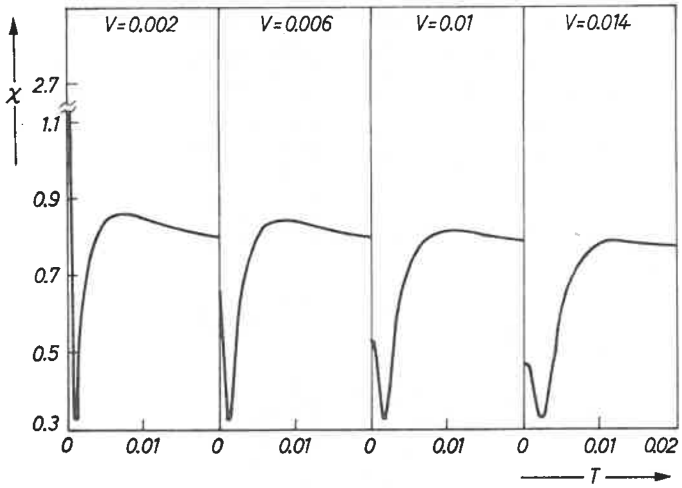

Fig. 2. Temperature dependence of the susceptibility for $\langle n_\sigma \rangle = 0.5$ and $E_{1\sigma} = -0.01$, $E_{2\sigma} = -0.0105$

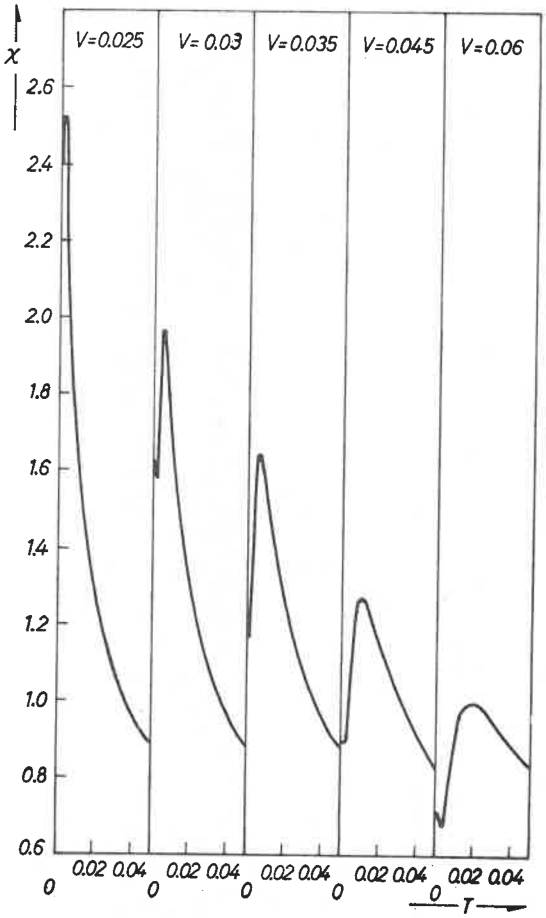

Fig. 3. Temperature dependence of the susceptibility for $\langle n_\sigma \rangle = 0.5$ and $E_{1\sigma} = -0.1$, $E_{2\sigma} = -0.11$

ceptibility of some intermediate valence compounds observed experimentally. For instance, the susceptibility of SmB_6 [7] exhibits a temperature dependence similar to that presented in Fig. 2 for $V = 0.006$. Reasonable similarity occurs between the susceptibility of $YbAl_3$ [8], Sm_2In_3 [9] and the curve corresponding to $V = 0.025$ in Fig. 3. The behaviour which is shown in Fig. 4 is similar to e.g. $SmPd_3$ [10]. It is noteworthy that a simple model gives reasonable, qualitative agreement with a wide class of intermediate valence compounds without magnetic ordering. Thus, we think that it would be interesting to take the crystal field splitting into account within the more realistic model in which the intra-atomic correlations should be present.

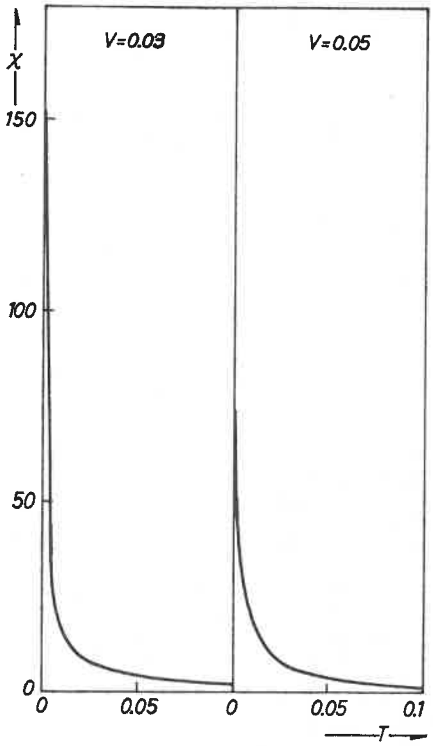

Fig. 4. Temperature dependence of the susceptibility for $\langle n_\sigma \rangle = 1$ and $E_{1\sigma} = -0.1$, $E_{2\sigma} = -0.11$

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