

## Coulomb correlation effects in the quasiparticle band structure of ferromagnetic rare-earth insulators

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We present a method for calculating the temperature dependence of the electronic quasiparticle density of states (QDOS) of a ferromagnetic rare-earth insulator like EuO. Special attention is devoted to how the "localized" ferromagnetism manifests itself in x-ray photoemission and bremsstrahlung isochromat spectra. Our study includes the first six conduction bands of EuO (the first five are Eu 5*d* like, the sixth is mainly of Eu 6*s* character) as well as the rather flat 4*f* levels. The starting point is an extended *d-f* exchange model, the main parts of which are an exchange interaction between 4*f* moments and conduction electrons, a Coulomb repulsion between highly correlated 4*f* electrons, and a hybridization of 4*f* with conduction-band states. We use an exact  $T=0$  relationship between spin-up quasiparticle energies and one-electron Bloch energies  $\epsilon_m(\mathbf{k})$  for an optimal determination of the latter by performing a self-consistent, spin-polarized band-structure calculation based on density-functional theory. For finite temperatures the model is approximately solved by a many-body procedure. The QDOS exhibits a striking temperature dependence mainly due to the *d-f* exchange. Two 4*f*-like peaks appear in the spin-polarized QDOS, the low-energy one being occupied, the high-energy one being empty. The temperature dependence of the localized ferromagnetism appears in the QDOS as a temperature-dependent shift of spectral weight between the low- and the high-energy peak.

### I. INTRODUCTION

In two preceding papers<sup>1,2</sup> we have proposed a new method, which combines a self-consistent one-electron band-structure calculation with a reliable many-body procedure, in order to get temperature-dependent quasiparticle densities of states (QDOS) for certain ferromagnetic materials. The QDOS has a direct relationship to experiment: the part below the Fermi edge to x-ray photoelectron spectroscopy (XPS) and ultraviolet photoelectron spectroscopy (UPS); the part above the Fermi edge to bremsstrahlung isochromat spectroscopy (BIS).<sup>3</sup>

Our "renormalized" many-body procedure may roughly be divided into three steps. In the first step we pick out all the interactions and correlations which seem to be important with respect to the physical problem under study, i.e., we construct a physical model:

$$H = H_0 + H_1 \quad (1.1)$$

The part  $H_1$  of the model Hamiltonian consists of the mentioned important interaction terms. The one-particle term  $H_0$ ,

$$H_0 = \sum_{m, \mathbf{k}, \sigma} \epsilon_m(\mathbf{k}) c_{\mathbf{k}m\sigma}^\dagger c_{\mathbf{k}m\sigma} \quad (1.2)$$

contains the Bloch energies  $\epsilon_m(\mathbf{k})$  ( $m$  is the band index,  $\mathbf{k}$  is the wave vector from the first Brillouin zone), which are determined in the second step by use of a self-

consistent band-structure calculation based on density-functional theory (DFT). In doing this we take into account, in a highly realistic manner, all the interactions in the system, which are not explicitly covered by  $H_1$ . In the third step we apply a many-body treatment, in order to find out how the interaction  $H_1$  modifies the renormalized one-electron energies  $\epsilon_m(\mathbf{k})$  into temperature- and concentration-dependent quasiparticle energies. The main problem of this procedure lies in the fact that the DFT, in principle, considers all interactions, i.e., the terms of  $H_1$  as well. Therefore, we must carefully pay attention not to count any of them twice.

In Refs. 1 and 2 we have applied this method to the classical Heisenberg-ferromagnets EuO and EuS. Strictly speaking, we have calculated the temperature dependence of the empty conduction band, which is predominantly built up by Eu 5*d* states. There is no doubt that the temperature effects are rather exclusively due to an exchange interaction between localized 4*f* electrons and itinerant conduction electrons. The choice of a proper model is therefore rather uniquely predetermined.<sup>1,2,4</sup> We have shown in Refs. 1 and 2 how to avoid in this case the above-mentioned double counting of certain interactions. The strictly localized 4*f* states entered our model calculations only in that sense that they give rise to localized magnetic moments, which order ferromagnetically below  $T_c$ . Their concrete energetic position was therefore unimportant. The main intention of the present study, however, is to extend the theory to the 4*f* states as well.

The DFT predicts for EuO at  $T=0$  the occupied  $4f \uparrow$  states with rather flat dispersion only 0.07 eV below the  $5d$  conduction-band minimum, and the  $4f \downarrow$  states some 6 eV higher (see Figs. 2 and 3). It is a well-known fact that local DFT theory fails to predict correct gaps in semiconductors and insulators, although some recent attempts have been made to overcome this difficulty.<sup>5,6</sup> The experimental value for the  $4f$ - $5d$  gap is indeed, with 1.12 eV,<sup>7</sup> very much larger. Nevertheless, DFT describes EuO at  $T=0$  correctly as a ferromagnetically saturated semiconductor. However, this is only a cursory view. Magnetism is produced in the DFT scheme by use of the “Stoner mechanism,” i.e., the magnetization is proportional to the difference of self-consistently calculated  $\uparrow$ - and  $\downarrow$ -particle numbers. Since the  $4f$  “band” is with seven electrons just half-filled, the splitting into a  $\uparrow$  subband below and a  $\downarrow$  subband above the Fermi-level yields, simultaneously, an insulator and a saturated ferromagnet. However, for finite temperatures the magnetization deviates from saturation. In the “Stoner mechanism” this is possible only by a partial overlapping of the  $4f$  spin bands. Then EuO would become a metal, in contradiction to the experiment.

In conclusion, we have to state two severe shortcomings of the DFT in connection with the  $4f$  states. It predicts a wrong position of the occupied  $4f$  states relative to the lower conduction-band edge, and secondly, it cannot describe “localized” magnetism as realized in substances like EuO. The main intention of our present study is therefore to implement as realistically as possible “localized”  $4f$  ferromagnetism into our renormalized many-body procedure. We want to find out how the magnetic state of the  $4f$  moments manifests itself in an XPS and a BIS experiment, respectively. Our study is not focused on the origin of ferromagnetism in EuO or EuS. We rather postulate its existence as a consequence of a certain kind of superexchange,<sup>8</sup> concentrating ourselves on its influence on the electronic band structure. This influence is twofold, where in a certain sense the one part is to ascribe to the occupied, the other to the empty  $4f$  states. The whole electronic quasiparticle spectrum will depend on the  $4f$ -moment ordering because of the already mentioned exchange interaction between localized  $4f$  electrons and itinerant conduction electrons. This aspect has been extensively discussed in our previous papers.<sup>1,2</sup> The other contribution stems from the unoccupied  $4f$  states being observable in an inverse photoemission experiment. The latter is based on the elementary process

$$(4f)^7 + e^- \rightarrow (4f)^8. \quad (1.3)$$

The final  $(4f)^8$  state, too, is highly localized as a consequence of strong intra-atomic Coulomb correlations. The attempt to determine the respective Coulomb matrix element  $U_f$  has quite a long history. Now it seems to be well-established<sup>9</sup> that the energy-distance between occupied and empty  $4f$  levels varies in the 5–7 eV range for all rare-earth metals, except for Eu and Gd, for which  $U_f$  is some 5 eV larger. It is a further interesting fact that the  $U_f$  values for the metals do agree surprisingly well with those for the insulators.<sup>9,10</sup> Although not measured

directly up to now, we believe that the  $U_f$  value for EuO is nearly the same as for the Eu metal. In our opinion the most reliable value has been given by Lang *et al.*<sup>11</sup> from a combination of XPS and BIS measurements:

$$U_f(\text{Eu metal}) = 10.14 \pm 0.3 \text{ eV}. \quad (1.4)$$

Let us assume that this value is correct for EuO as well. The occupied  $4f$  states, observed in XPS, are energetically separated from the conduction-band states, while the empty  $4f$  states, seen in BIS fall into the band region. If we take from the experiment,<sup>7</sup> the position of the occupied  $4f^7$  level 1.12 eV below the conduction-band edge, then we have to expect the empty  $4f^8$  states in the uppermost part of the mainly  $5d$ -like band region, which we have discussed in our previous papers.<sup>1,2</sup> There we disregarded, however, the  $4f^8$  peak. This is of course questionable for the high-energy part of the  $5d$ -like spectrum, and also for the following  $6s$ -like spectrum because besides the bare existence of the  $4f$  peak, there will also occur a strong hybridization with the conduction-band states. It is easy to see that the effect of this hybridization will be temperature dependent. At  $T=0$ , e.g., all  $4f$  moments are aligned parallel. The seven  $4f$  electrons are spin-up electrons. According to Hund’s rule, the  $4f^8$  state is then reachable in a BIS experiment only by a spin-down electron. The empty  $4f^8$  peak therefore appears at  $T=0$  exclusively in the spin-down spectrum. For finite temperatures the seven  $4f$  electrons are still completely aligned by Hund’s-rule coupling, forming a total spin  $S = \frac{7}{2}$ , but this holds, of course, only in a local atomic frame. From site to site the localized spins will deviate from the  $z$  direction, so that an externally prepared spin-up electron will enter the local frame with finite probability as will the spin-down electron. Therefore we have to expect the  $4f^8$  peak at  $T > 0$  in the spin-up BIS spectrum, too.

The intention of this paper is to present an extension of the theory in Refs. 1 and 2 by taking into account explicitly the  $4f$  states and the  $6s$  conduction-band states. We want to find out how the “localized” ferromagnetism manifests itself in the QDOS and therewith in XPS and BIS spectra. In order to be concrete, we concentrate ourselves again on EuO, the prototype of a rare-earth insulator. The concept is, however, more general.

## II. “RENORMALIZED” MANY-BODY PROCEDURE

### A. The model

We want a theoretical model for a ferromagnetic  $4f$  insulator like EuO. The localized  $4f$  electrons of the rare-earth ion form magnetic moments with long-range order below the Curie temperature  $T_C$  [ $T_C = 69.33$  K in EuO (Ref. 7)]. The conduction-band states are highly affected by the magnetic ordering of the localized moment system. The corresponding correlations have to be taken into account by our model with special care.

The EuO conduction band consists of hybridized  $5d$  and  $6s$  subbands, all being built up by original Eu states. We number them by a band index  $m$ . Conduction electrons are then described by

$$H_c = \sum_{\mathbf{k}, m, \sigma} \varepsilon_m(\mathbf{k}) c_{\mathbf{k}m\sigma}^\dagger c_{\mathbf{k}m\sigma}, \quad (2.1)$$

where  $c_{\mathbf{k}m\sigma}^\dagger$  ( $c_{\mathbf{k}m\sigma}$ ) is the creation (annihilation) operator for an electron with wave vector  $\mathbf{k}$  and spin  $\sigma$ . For the strictly localized  $4f$  electrons we use an atomic-limit Hubbard model

$$H_f = \sum_{i, \sigma, \alpha} (E_f f_{i\alpha\sigma}^\dagger f_{i\alpha\sigma} + \frac{1}{2} U_f n_{f i\alpha\sigma} n_{f i\alpha-\sigma}). \quad (2.2)$$

$E_f$  is the energy of the  $f$  level, which has a sevenfold orbital degeneracy ( $\alpha$ ) in addition to the spin degeneracy ( $\sigma$ ).  $U_f$  is the intra-atomic Coulomb interaction, which takes care for the fact that there are seven occupied low-energy states and seven empty high-energy states. Multiplet effects are neglected by choosing  $U_f$  and  $E_f$   $\alpha$  independent. We assume, however, that the seven  $4f$  electrons are Hund's-rule coupled to a total spin  $S = \frac{7}{2}$ . At a later stage we shall explain in detail the connection between the local spin operator  $\mathbf{S}_i = (S_i^x, S_i^y, S_i^z)$  and the creation and annihilation operators  $f_{i\alpha\sigma}^\dagger, f_{i\alpha\sigma}$  for the  $f$  electrons ( $n_{f i\alpha\sigma} = f_{i\alpha\sigma}^\dagger f_{i\alpha\sigma}$ ).

The  $4f$  moments are coupled by a certain kind of superexchange<sup>7,8</sup> enabling ferromagnetic order below  $T_C$ . Since we are interested in the electronic excitation energies, only, and magnon energies are smaller by some orders of magnitude, we are allowed to neglect for simplicity in our model a direct exchange between the moments. Nevertheless, certain  $f$ -spin correlations like  $\langle S_i^z \rangle, \langle S_i^+ S_j^- \rangle, \dots$ , will enter into the final results which are determined at a later stage separately by use of the pure Heisenberg model. This is justified because a possible influence of itinerant conduction electrons on the magnon spectrum via  $d$ - $f$  exchange is excluded for the insulator EuO.

The empty  $4f$  level will appear in the conduction-band region, deforming it by hybridization. This is mediated by

$$H_v = \frac{1}{\sqrt{N}} \sum_{\mathbf{k}, m, i, \sigma, \alpha} [V_{m\alpha}(\mathbf{k}) e^{-i\mathbf{k}\cdot\mathbf{R}_i} c_{\mathbf{k}m\sigma}^\dagger f_{i\alpha\sigma} + \text{H.c.}]. \quad (2.3)$$

The main influence on the temperature dependence of the electronic quasiparticle spectra is to expect from an exchange interaction between localized  $4f$  moments and itinerant conduction electrons:<sup>1,2,4</sup>

$$H_{\text{ex}} = \frac{1}{2N} \sum_{i, \sigma} \sum_{\mathbf{k}, \mathbf{q}} \sum_m g_m e^{-i\mathbf{q}\cdot\mathbf{R}_i} (z_\sigma S_i^z c_{\mathbf{k}m\sigma}^\dagger c_{\mathbf{k}+\mathbf{q}m\sigma} + S_i^\sigma c_{\mathbf{k}m-\sigma}^\dagger c_{\mathbf{k}+\mathbf{q}m-\sigma}). \quad (2.4)$$

Here we have written for abbreviation:

$$S_j^\sigma = S_j^x + iz_\sigma S_j^y, \quad z_\uparrow = +1, \quad z_\downarrow = -1. \quad (2.5)$$

We neglect from the very beginning a possible  $\mathbf{k}$  dependence of the  $d$ - $f$  exchange constant  $g_m$ , and also intersubband scattering. Furthermore, we treat the hybridization matrix elements  $V_{m\alpha}(\mathbf{k})$  as adjustable parameters, and as

usual but somewhat arbitrarily as  $\mathbf{k}$ ,  $m$ , and  $\alpha$  independent, i.e.,  $V_{m\alpha}(\mathbf{k}) = V$ . We are aware of the fact that this may be considered as a serious approximation, because there are symmetry requirements especially at certain symmetry points. However, we believe that this affects mainly the dispersion relations, being, however, more or less washed out for the quasiparticle densities of states which represent, in a certain sense,  $\mathbf{k}$ -averaged quantities.

Finally we describe the ferromagnetic  $4f$  insulator EuO by the following model Hamiltonian:

$$H = H_c + H_f + H_{\text{ex}} + H_v, \quad (2.6)$$

which provokes a very complicated many-body problem.

### B. The effective-medium approach

Complications arise from the exchange interaction  $H_{\text{ex}}$  (2.4), and from the Coulomb correlations (2.2) between the  $f$  electrons. These terms lead to rather involved, temperature-dependent self-energies  $M_{\mathbf{k}\sigma}^{(m)}(E)$  for conduction electrons and  $M_{f\sigma}^{(\alpha)}(E)$  for  $f$  electrons. Let us assume for the moment that we have found a physically reasonable way to determine them. Then we can replace the Hamiltonian  $H$  in Eq. (2.6) by an effective one with the same thermodynamic properties:

$$H_{\text{eff}} = \sum_{\mathbf{k}, \sigma, m} E_{\mathbf{k}\sigma}^{(m)}(E) c_{\mathbf{k}m\sigma}^\dagger c_{\mathbf{k}m\sigma} + \sum_{i, \sigma, \alpha} E_{f\sigma}^{(\alpha)}(E) f_{i\alpha\sigma}^\dagger f_{i\alpha\sigma} + V \sum_{i, \sigma, m, \alpha} (c_{i\sigma m}^\dagger f_{i\alpha\sigma} + f_{i\alpha\sigma}^\dagger c_{i\sigma m}). \quad (2.7)$$

Here we have defined

$$E_{\mathbf{k}\sigma}^{(m)}(E) = \varepsilon_m(\mathbf{k}) + M_{\mathbf{k}\sigma}^{(m)}(E), \quad (2.8)$$

$$E_{f\sigma}^{(\alpha)}(E) = E_f + M_{f\sigma}^{(\alpha)}(E). \quad (2.9)$$

The problem connected with (2.7) is exactly solvable. We derive it in terms of the Green function

$$\underline{G}_{\mathbf{k}\sigma}(E) = \langle \mathbf{k}\sigma | (E - H_{\text{eff}})^{-1} | \mathbf{k}\sigma \rangle \quad (2.10)$$

which is a  $13 \times 13$  matrix of the following form:

$$\underline{G}_{\mathbf{k}\sigma}(E) = \begin{bmatrix} \underline{D}_{\mathbf{k}\sigma}(E) & \underline{V}^T \\ \underline{V} & \underline{E}_\sigma(E) \end{bmatrix}^{-1}, \quad (2.11)$$

where  $\underline{D}_{\mathbf{k}\sigma}(E)$  is a diagonal  $6 \times 6$  matrix

$$\underline{D}_{\mathbf{k}\sigma}(E) = \begin{bmatrix} E - E_{\mathbf{k}\sigma}^{(1)}(E) & & 0 \\ & \ddots & \\ 0 & & E - E_{\mathbf{k}\sigma}^{(6)}(E) \end{bmatrix}, \quad (2.12)$$

$\underline{E}_\sigma(E)$  a diagonal  $7 \times 7$  matrix

$$\underline{E}_\sigma(E) = \begin{bmatrix} E - E_{f\sigma}^{(1)}(E) & & 0 \\ & \ddots & \\ 0 & & E - E_{f\sigma}^{(7)}(E) \end{bmatrix}, \quad (2.13)$$

and  $\underline{v}$  a  $7 \times 6$  matrix, the elements of which are all equal  $-V$ . It is evident from the ansatz (2.7) that all renormalized  $f$  energies  $E_{f\sigma}^{(\alpha)}(E)$  are degenerate, since all the bare  $f$  levels  $E_f$  are so, and  $U_f$ , as well as  $V$ , are assumed  $\alpha$  independent.

We invert the matrix (2.11) by use of the Gauss formula

$$G_{\mathbf{k}\sigma}^{(m)}(E) = \frac{E - E_{f\sigma}(E) - 7V^2 P_{m\sigma}(\mathbf{k}, E)}{[E - E_{f\sigma}(E) - 7V^2 P_{\sigma}(\mathbf{k}, E)][E - E_{\mathbf{k}\sigma}^{(m)}(E)]} \quad m = 1, 2, \dots, 6 \quad (2.15)$$

$$F_{\mathbf{k}\sigma}^{(\alpha)}(E) \equiv F_{\mathbf{k}\sigma}(E) = \frac{E - E_{f\sigma}(E) - 6V^2 P_{\sigma}(\mathbf{k}, E)}{[E - E_{f\sigma}(E) - 7V^2 P_{\sigma}(\mathbf{k}, E)][E - E_{f\sigma}(E)]} \quad \alpha = 1, 2, \dots, 7. \quad (2.16)$$

Here we have defined

$$P_{m\sigma}(\mathbf{k}, E) = P_{\sigma}(\mathbf{k}, E) - [E - E_{\mathbf{k}\sigma}^{(m)}(E)]^{-1}, \quad (2.17)$$

$$P_{\sigma}(\mathbf{k}, E) = \sum_{m=1}^6 [E - E_{\mathbf{k}\sigma}^{(m)}(E)]^{-1}. \quad (2.18)$$

From the so-determined Green function we derive physical quantities of special importance, namely the one-electron spectral density

$$A_{\mathbf{k}\sigma}(E) = -\frac{1}{\pi} \text{Im} \left[ \sum_{m=1}^6 G_{\mathbf{k}\sigma}^{(m)}(E + i0^+) + 7F_{\mathbf{k}\sigma}(E + i0^+) \right] \quad (2.19)$$

and the quasiparticle density of states QDOS:

$$\rho_{\sigma}(E) = \frac{1}{N} \sum_{\mathbf{k}} A_{\mathbf{k}\sigma}(E). \quad (2.20)$$

It is an interesting detail that the  $f$ -like Green functions (2.16) have a pole at  $E \equiv E_{f\sigma}^{(\alpha)}(E)$  with a residue ("spectral weight") of  $\frac{6}{7}$  for each  $\alpha$ . This means that effectively only one of the seven degenerate  $4f$  levels will hybridize with a conduction  $\mathbf{k}$  state, the rest will be rather unaffected. Anderson,<sup>12</sup> has qualitatively discussed this aspect in another context.<sup>13</sup> We shall come back to this point when discussing our results.

For a concrete evaluation of the above equations we have to fix the hybridization matrix element  $V$ . We consider

$$V = 0.1 \text{ eV} \quad (2.21)$$

to be a reasonable value.

### C. $f$ -electron self-energy

It is impossible to calculate the  $f$ -electron self-energy  $M_{f\sigma}^{(\alpha)}(E)$  self-consistently within the framework of the total model (2.6). Among other things this term has to express the local-moment ferromagnetism. For the description of the  $4f$  electrons we have to distinguish two reference systems  $\Sigma$  and  $\Sigma'$ .  $\Sigma$  is an external frame of coordinates, the  $z$  axis of which is defined by the measuring apparatus.  $\Sigma'$  is an atomic frame, for which all the seven  $4f$  electrons are spin-up electrons, and that for all tempera-

$$\det \underline{G}_{\mathbf{k}\sigma}^{-1}(E) = \det \underline{F}_{\sigma}(E) \det [\underline{D}_{\mathbf{k}\sigma}(E) - \underline{V}^T \underline{F}_{\sigma}^{-1}(E) \underline{V}]. \quad (2.14)$$

After some straightforward manipulations we find the following expressions for the diagonal elements of the Green-function matrix, which we need for calculating the QDOS:

tures. In  $\Sigma$ , however, they appear as spin-up electrons only for  $T=0$ . For finite temperatures the total spin  $S = \frac{7}{2}$  will deviate from the  $z$  direction (Fig. 1). The actual deviation varies statistically from site to site, where the averaged deviation is determined by the macroscopic magnetization  $\langle S^z \rangle$ . In a semiclassical picture  $\langle S^z \rangle$  corresponds to the averaged projection of the  $4f$  spin  $\mathbf{S}_i$  on the "external"  $z$  direction:

$$\langle \cos \vartheta_i \rangle \approx \langle S^z \rangle / S. \quad (2.22)$$

According to elementary quantum mechanics the probability for an electron, which has been prepared as a  $\sigma$  electron in  $\Sigma$ , to appear as  $\sigma'$  electron in  $\Sigma'$  is given by

$$\beta_{\sigma\sigma'}^{(i)} = \frac{1}{2} (1 + z_{\sigma} z_{\sigma'} \cos \vartheta_i). \quad (2.23)$$

In the case of EuO, all  $f$  electrons have  $\sigma' = \uparrow$  in  $\Sigma'$ , i.e.,  $z_{\sigma'} = 1$ . On the other hand, when we extract from the  $f$  levels an electron by photoemission, it will appear in the external frame  $\Sigma$  as a  $\sigma$  electron with the probability  $\langle n_{f\sigma} \rangle$ , which is the average occupation number of the respective  $f$  level. With (2.22) and (2.23) it should therefore hold in reasonable approximation

$$\langle n_{f\sigma} \rangle = \langle \beta_{\sigma\uparrow}^{(i)} \rangle = \frac{1}{2} (1 + z_{\sigma} \langle S^z \rangle / S). \quad (2.24)$$

This expression provides the connection between the  $f$ -electron construction operators  $f, f^{\dagger}$  and the localized spin operator  $\mathbf{S}_i$ .

The one-electron Green function for the subsystem

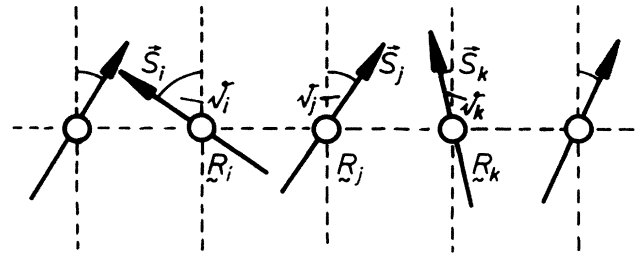


FIG. 1. Semiclassical model for the localized  $4f$  spin system (see text).

(2.2) can be found rigorously:

$$F_{\alpha\sigma}^{(0)}(E) = \frac{1 - \langle n_{f\alpha-\sigma} \rangle}{E - E_f} + \frac{\langle n_{f\alpha-\sigma} \rangle}{E - E_f - U_f}. \quad (2.25)$$

It allows an approximate determination of the  $f$  self-energy by the ansatz:

$$F_{\alpha\sigma}^{(0)}(E) = [E - E_f - M_{f\sigma}^{(\alpha)}(E)]^{-1}. \quad (2.26)$$

The comparison yields, if we use (2.24),

$$\begin{aligned} M_{f\sigma}^{(\alpha)}(E) &\equiv M_{f\sigma}(E) \\ &= \frac{U_f}{2} \left[ 1 - z_\sigma \frac{\langle S^z \rangle}{S} \right] \\ &\quad \times \frac{E - E_f}{E - E_f - (U_f/2)(1 + z_\sigma \langle S^z \rangle / S)}. \end{aligned} \quad (2.27)$$

This self-energy contains two important parameters  $E_f$  and  $U_f$ , which must be taken from the experiment.  $E_f$  is determined by the  $4f$ - $5d$  gap [1.12 eV in EuO (Ref. 7)]. As discussed in the Introduction, the choice (1.4) for the so-called ‘‘Hubbard  $U_f$ ’’ should be realistic.

#### D. Conduction-electron self-energy

We derive the conduction-electron self-energy  $M_{\mathbf{k}\sigma}^{(m)}(E)$  approximately from the partial Hamiltonian

$$\bar{H}_c = H_c + H_{\text{ex}}, \quad (2.28)$$

being nothing else than the ‘‘normal’’  $s$ - $f$  model<sup>4</sup> (Kondo lattice).  $\bar{H}_c$  provokes a nontrivial many-body problem, which is, however, exactly solvable for  $T=0$ . For details of the rigorous  $T=0$  treatment, the reader is referred to our previous paper.<sup>1</sup> The spin-up quasiparticle spectrum is found to be particularly simple:

$$E_{m\uparrow}(\mathbf{k}; T=0) = \epsilon_m(\mathbf{k}) - \frac{1}{2}g_m S, \quad (2.29)$$

$$M_{\uparrow}^{(m)}(E; T=0) = -\frac{1}{2}g_m S. \quad (2.30)$$

The spin-down spectrum is, however, more complicated. The self-energy reads as

$$M_{\downarrow}^{(m)}(E; T=0) = \frac{1}{2}g_m S \left[ 1 + \frac{g_m B_{0\uparrow}^{(m)}(E; T=0)}{1 - \frac{1}{2}g_m B_{0\uparrow}^{(m)}(E; T=0)} \right]. \quad (2.31)$$

$$B_{0\sigma}^{(m)}(E; T) = \frac{1}{N} \sum_{\mathbf{p}} [E - \epsilon_m(\mathbf{p}) - M_{\sigma}^{(m)}(E; T)]^{-1} \quad (2.32)$$

The simple result (2.30) is of decisive importance for our further procedure, because it shows that at  $T=0$ , the spin-up spectrum of the interacting system is quasi-identical with the ‘‘free’’ Bloch energies  $\epsilon_m(\mathbf{k})$ . ‘‘Free’’ means here without  $d$ - $f$  exchange interaction. One could

argue that the terms  $H_f$  and  $H_v$ , not included in  $\bar{H}_c$  (2.28), will place some  $f$  states within the conduction-band region, which then substantially deform the  $\epsilon_m(\mathbf{k})$  by hybridization. This is indeed the case for finite temperatures, but not for  $T=0$ . In this special case  $M_{f\uparrow}^{(\alpha)}$  vanishes identically, so that according to (2.9) all  $4f \uparrow$  states are located far below (1.12 eV) the conduction band.

When performing a self-consistent spin-polarized band-structure calculation on the basis of density-functional theory (DFT), it is obviously allowed to identify the spin-up result with the ‘‘free’’ one-particle energies  $\epsilon_m(\mathbf{k})$ . The DFT takes into account, in principle, all interactions, i.e., in a certain sense the  $d$ - $f$  exchange  $H_{\text{ex}}$  as well. The latter leads, however, in the  $T=0$  spin-up spectrum (2.29), to a trivial additive term, only, which is meaningless because of the free choice of the energy zero. On the other hand, the effective single-particle energies of DFT contain all other interactions which are not explicitly considered within the model (2.6). By this method, we surely get highly realistic input parameters  $\epsilon_m(\mathbf{k})$  for our  $T=0$  many-body procedure. Strictly speaking, we have used the experimental<sup>7</sup> lattice constant of EuO  $a = 5.141 \text{ \AA}$ , and the prescription of Moruzzi *et al.*<sup>14</sup> for the spin-polarized exchange and correlation potential of DFT to perform a nonrelativistic augmented spherical wave (ASW) calculation.<sup>15</sup> We note that the inclusion of relativistic effects would surely shift the relative  $4f$ - $5d$  position, but since, within our procedure, the value of the  $4f$ - $5d$  gap is fitted to the experimental value, this fact cannot significantly alter our final results. The actual calculation of the single-particle energies  $\epsilon_m(\mathbf{k})$  and the corresponding densities of states is based on a discrete mesh of 182  $\mathbf{k}$  points in the irreducible wedge of the first Brillouin zone.

The spin-up density of states is plotted in Fig. 2. For our model we need the unoccupied part of the spectrum [Fig. 2(b)]. The first five conduction bands ( $m=1, \dots, 5$ ) are mainly  $5d$ -like, the sixth subband ( $m=6$ ) is predominantly built up by Eu  $6s$  states. Higher-lying bands are disregarded.

While the model (2.28) is exactly solvable for  $T=0$ , approximations must be tolerated for finite temperatures. In a preceding paper<sup>2</sup> we have proposed an approach for the temperature dependence of the conduction-electron self-energy, which treats the decisive spin-exchange processes between  $f$  moments and conduction electrons with special care and turns out to be exact for  $T=0$ . The latter fact is to consider a weighty support for our approximate theory. Details of the mathematical derivation are presented in Ref. 2. We cite here only the final result for the self-energy:

$$M_{\sigma}^{(m)}(E; T) = -\frac{1}{2}g_m z_\sigma \langle S^z \rangle + \frac{\frac{1}{4}g_m^2 \langle S_i^{-\sigma} S_i^{\sigma} \rangle B_{0-\sigma}^{(m)}(E; T)}{1 - [\frac{1}{2}g_m (1 + z_\sigma \langle S^z \rangle) - M_{-\sigma}^{(m)}(E; T)] B_{0-\sigma}^{(m)}(E; T)}. \quad (2.33)$$

The exchange coupling constants  $g_m$  are model parameters.  $g_1$  may be fixed by the spectacular red shift of the optical absorption edge for the electronic  $4f$ - $5d$  transition, which is observed with decreasing temperature

below  $T_C$ . The total edge shift between room temperature and  $T=0$  amounts to 0.27 eV in EuO.<sup>7</sup> It results of course from a respective shift of the lower conduction-band edge being correctly reproduced within our model

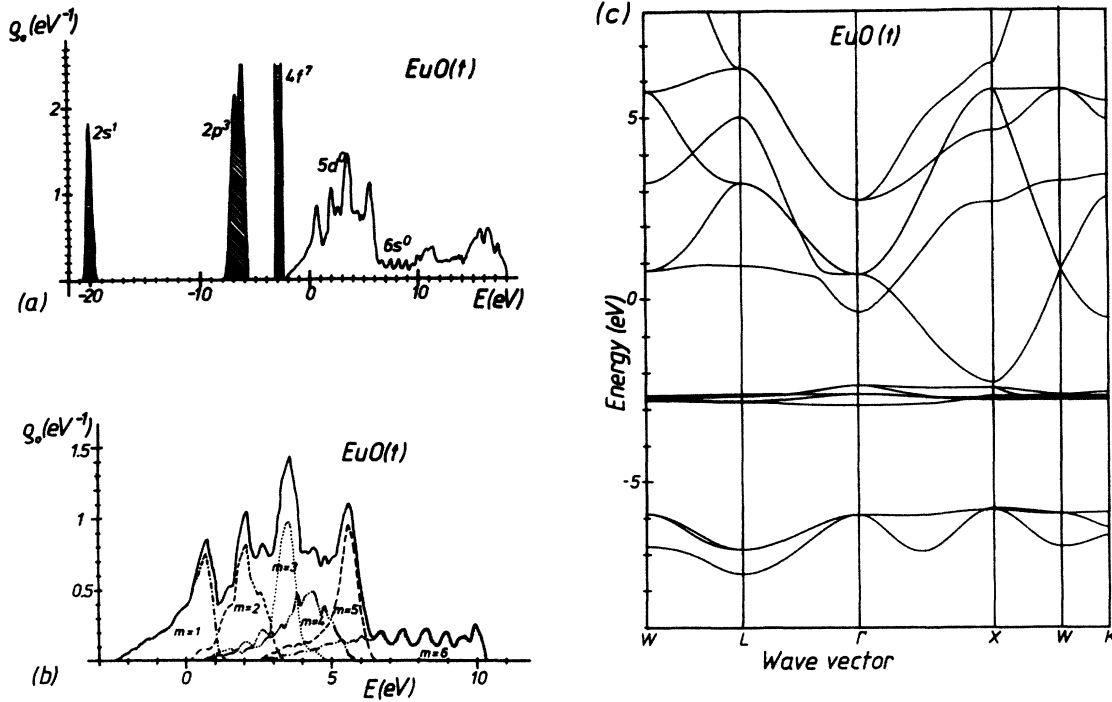


FIG. 2. Ground-state spin-up results for EuO, obtained from a nonrelativistic ASW calculation. (a) Bloch density of states (BDOS)  $\rho_0$  per atom as a function of energy. Shaded region marks the occupied part of the spectrum. (b) Partial densities of states ( $m = 1, \dots, 6$ ) of the first six conduction bands as functions of energy. The energy zero coincides with the center of gravity of the  $m = 1$  subband. The  $m = 1, \dots, 5$  subbands are mainly of Eu  $5d$  character; the  $m = 6$  subband of Eu  $6s$  character. (c) Band structure as a function of wave vector.

by taking  $g_1 = 0.2$  eV. Unfortunately, we do not see similar possibilities to fix the other  $g_m$ , also, by experimental data. It is, however, plausible that the exchange constants  $g_m$ , which couple  $4f$  and  $5d$  states of the  $\text{Eu}^{2+}$  ion, are all of the same order of magnitude. On the other hand, the coupling between  $4f$  and  $6s$  states is surely much weaker because the overlap of  $4f$  and  $6s$  wave functions is substantially smaller than that of  $4f$  and  $5d$  wave functions, as can be checked by Fig. 5 of Ref. 16. Similar arguments have been used by Harmon and Freeman,<sup>17</sup> in order to derive for Gd  $g(4f - 6s) = 0.4 \times g(4f - 5d)$ . We therefore choose:

$$g_m = 0.2 \text{ eV for } m = 1, \dots, 5; \quad g_6 = 0.1 \text{ eV}. \quad (2.34)$$

### E. $f$ -spin correlation functions

Our results (2.27) for the  $f$ -electron self-energy and (2.33) for the conduction-electron self-energy, respectively, contain the  $f$  magnetization  $\langle S^z \rangle$  and the  $f$ -spin correlation  $\langle S_i^\sigma S_i^{-\sigma} \rangle$  being mainly responsible for the temperature dependence of the quasiparticle energy spectrum. As explained in Sec. II A, these correlation functions may be derived from the bare Heisenberg model, because the conduction band of the insulator EuO is empty, so that there cannot be any influence of itinerant electrons on the localized moment system.

We have proposed in Ref. 2 a self-consistent spectral-moment method for the Dyson-Malëev transformed Heisenberg model, by which we get  $\langle S^z \rangle$  in quantitative agreement with experimental EuO data, at least for  $0 \leq T \leq 0.8T_C$  (see Fig. 2 in Ref. 2). The same method, applied to the transverse spin-correlation function, leads, in this low-temperature region, to the following simple formula:

$$\langle S_i^\sigma S_i^{-\sigma} \rangle = (1 + z_\sigma) \langle S^z \rangle + (S + \langle S^z \rangle) (S - \langle S^z \rangle). \quad (2.35)$$

In the paramagnetic region ( $T > T_C$ ) we take the result of a "local" mean-field approximation:<sup>18,19</sup>

$$\langle S_i^\sigma S_i^{-\sigma} \rangle = \frac{2}{3} S(S+1) \frac{1}{N} \sum_{\mathbf{q}} \left[ 1 - \frac{T_C J(\mathbf{q})}{T J(0)} \right]^{-1}. \quad (2.36)$$

## III. DISCUSSION OF THE RESULTS

Our investigation is concentrated with the electronic quasiparticle spectrum of ferromagnetic  $4f$  insulators. We are mainly interested in the reaction of the quasiparticle density of states (QDOS) on the localized moment ferromagnetism, which should manifest itself in a characteristic temperature dependence. In order to be concrete, we present results for EuO, surely a prototype of ferromagnetic semiconductors. As already mentioned, we

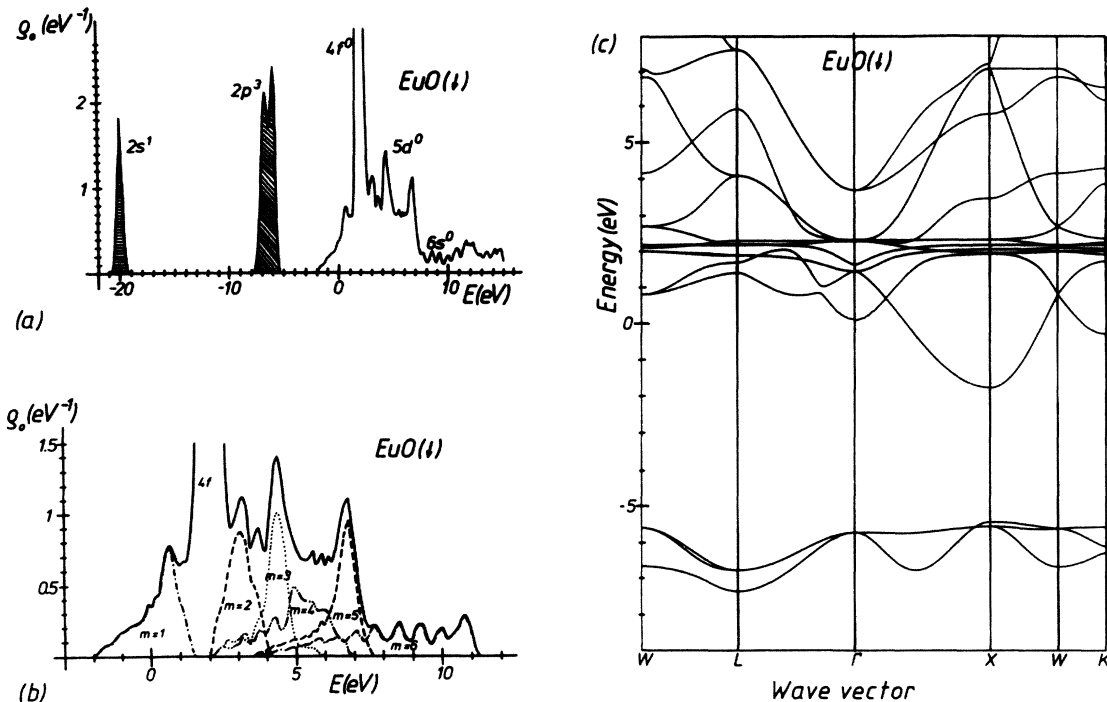


FIG. 3. Same as in Fig. 2, but for spin down.

do not intend to explain the origin of ferromagnetism in EuO, but assume that the localized magnetic  $4f$  moments order collectively below the Curie temperature  $T_C = 69.33$  K (Ref. 7) as a consequence of a special kind of superexchange (Ref. 8). Currently, it is well-established that the EuO exchange integrals are of importance for nearest and next-nearest neighbors, only.<sup>20–23</sup> Using the experimental values  $J_1/k_B = 0.625$  K and  $J_2/k_B = 0.125$  K, we achieved in a previous paper,<sup>2</sup> for the magnetization  $\langle S^z \rangle$ , a quantitative fit with experimental data by applying a self-consistent moment method to the Heisenberg model. We take this  $\langle S^z \rangle$  as input for our present study.

A typical feature of the QDOS of a  $4f$  system is the appearance of two very pronounced peaks. The low-energy peak, consisting of occupied  $4f$  states and therefore being observable by XPS, is localized below the conduction-band region, at least in  $4f$  insulators or semiconductors. The high-energy peak is built up by unoccupied  $4f$  states, which fall into the empty conduction-band region, and therefore hybridize strongly with neighboring band states. Because of this hybridization the high-energy peak appears broader than the low-energy one, and can be interpreted in a certain sense as a quasiparticle state, which may move through the lattice with spin  $\sigma$ , momentum  $\hbar\mathbf{k}$ , and energy  $E_\sigma(\mathbf{k})$ . This can be detected in a BIS experiment according to an electronic  $4f^n \rightarrow 4f^{n+1}$  transition. The energetic separation of the two peaks is a measure of the Coulomb correlation energy, which enters our theory as parameter  $U_f$  in the Hamiltonian  $H_f$  (2.2). As discussed in the Introduction, we believe that the value  $U_f \approx 10$  eV, measured by Lang *et al.*<sup>11</sup> for Eu metal, should be realistic for EuO as well.

Described in detail in Sec. II D is how the one-electron energies  $\varepsilon_m(\mathbf{k})$  are determined by a self-consistent spin-polarized band-structure calculation based on the density-functional theory (DFT). They therewith bring into account, in a highly realistic manner, those interactions which are not explicitly covered by our model Hamiltonian. Furthermore, the exact relation (2.29) guarantees that no interaction is counted twice. Our model is constructed so, that it contains all terms which are thought to be decisive for temperature effects in the QDOS. Other interactions may be important for the general structure of the energy spectra, but do not modify them very much as a function of temperature. The  $T=0$   $\uparrow$  results of DFT therefore represent an optimal renormalized basis set for our many body procedure. Fig-

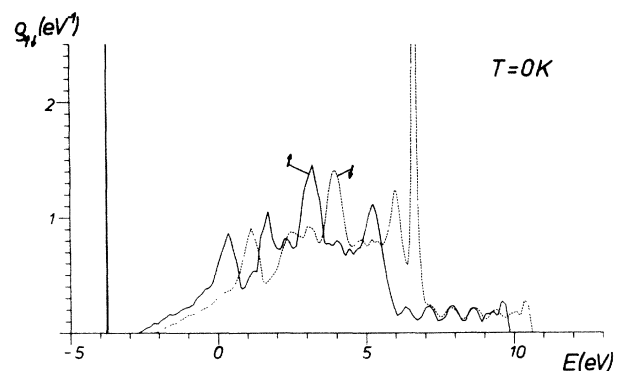


FIG. 4. Quasiparticle densities of states per atom  $\rho_1$  and  $\rho_2$  as a function of energy for EuO at  $T=0$  K. Solid line denotes the spin-up case; dashed line for the spin-down case. The pronounced peaks are Eu  $4f$  like.

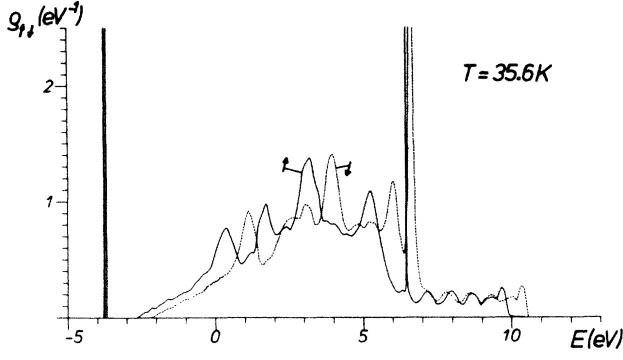


FIG. 5. Same as in Fig. 4, but for  $T = 35.6$  K ( $\langle S^z \rangle / S = \frac{6}{7}$ ).

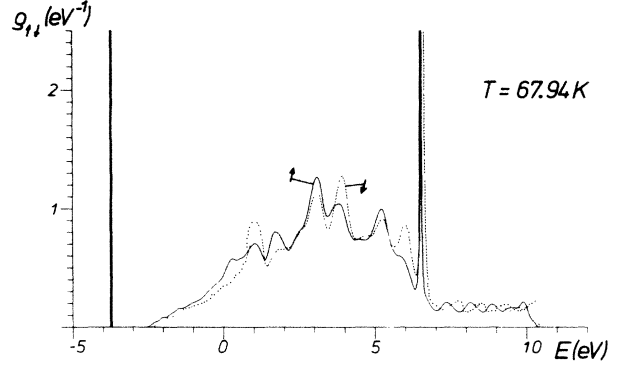


FIG. 7. Same as in Fig. 4, but for  $T = 67.94$  K ( $\langle S^z \rangle / S = \frac{2}{7}$ ).

ure 2(a) shows the calculated one-electron Bloch density of states (BDOS) over a region of some 40 eV. The  $2s$  and  $2p$  peaks stem from the oxygen being no longer considered within our study. The occupied  $4f\uparrow$  peak lies only 0.07 eV below the empty conduction band, while the experimental value is 1.12 eV.<sup>7</sup> This discrepancy is to ascribe to the well-known failure of DFT to predict correct band gaps in insulators. However, this shortcoming of DFT does not affect our procedure, since the  $f$ -peak position is adjusted by the parameter  $E_f$  in the model Hamiltonian (2.2). The decisive input information, which we take from DFT for our many-body treatment following Eq. (2.29), are the first six empty conduction bands [Fig. 2(b)], numbered by  $m = 1, \dots, 6$ .

Figure 3 shows the  $T=0$   $\downarrow$  DFT results. The empty  $f$  peak is located within the region of the  $m = 1$  and  $m = 2$  Bloch band. Therefore, DFT predicts for EuO at  $T=0$  ferromagnetic saturation (totally polarized  $4f$  electrons), and insulating or semiconducting behavior. Although this is just the experimental observation, we do not believe in the correctness of the DFT  $\downarrow$  spectrum because of mainly three reasons. (1) The strong intra-atomic Coulomb correlation is surely not sufficiently well treated in a one-electron band-structure calculation. Therefore, the position of the  $4f\downarrow$  peak in Fig. 3 is doubtful, but on the other hand it is also very decisive because of the strong hybridization with conduction-band states. Lopez-Aguilar and Costa-Quintana<sup>24–26</sup> have tried to solve this so-called “Coulomb gap problem” for paramagnetic  $4f$  systems like SmS by adding a proper pseudopo-

tential to the band Hamiltonian, which simulates the Coulomb correlation effects. By this method, unoccupied  $4f$  states are pushed to higher energies. (2) DFT is dramatically overcharged in what concerns localized moment ferromagnetism as realized in EuO. This problem has already been discussed in the Introduction, and remains the main impetus for our present study. It is clear that for the  $4f$  insulator EuO, only saturated ferromagnetism can be reproduced by band-structure calculations, more or less by chance. A continuous temperature dependence of the magnetization  $\langle S^z \rangle$  would require in the “band picture” a continuous change of the number of occupied and unoccupied  $4f\uparrow$  and  $4f\downarrow$  states per site, but a partial overlapping of  $4f\uparrow$  and  $4f\downarrow$  “bands” is incompatible with the requirement that EuO is an insulator. However, it is of course impossible, even in the improved version of Refs. 24–26, to realize a noninteger number of  $4f\uparrow$  or  $4f\downarrow$  states per site. (3) The exact  $T=0$  evaluation of the  $d$ - $f$  exchange  $H_{ex}$  shows that for the  $\uparrow$  spectrum dynamic spin-flip processes between  $4f$  spins and conduction-electron spins, i.e., magnon emission or absorption are excluded. It therefore follows a very simple relationship (2.29) between quasiparticle and one-electron energies. A  $\downarrow$  electron, however, can exchange its spin with the localized  $4f$  spins even at  $T=0$ . This results in a rather involved  $\downarrow$  self-energy (2.31). Such spin-flip processes, which are responsible for remarkable shifts, deformations and splittings of the original BDOS, are not taken into account by the DFT.

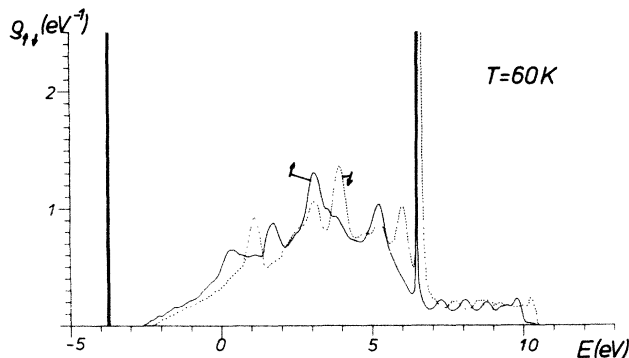


FIG. 6. Same as in Fig. 4, but for  $T = 60$  K ( $\langle S^z \rangle / S = \frac{4}{7}$ ).

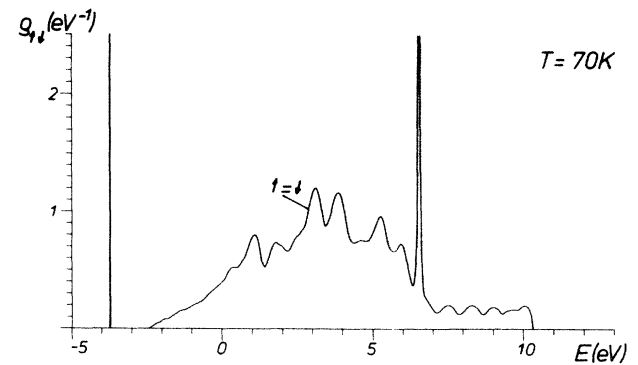


FIG. 8. Same as in Fig. 4, but for  $T = 70$  K ( $\langle S^z \rangle = 0$ ).



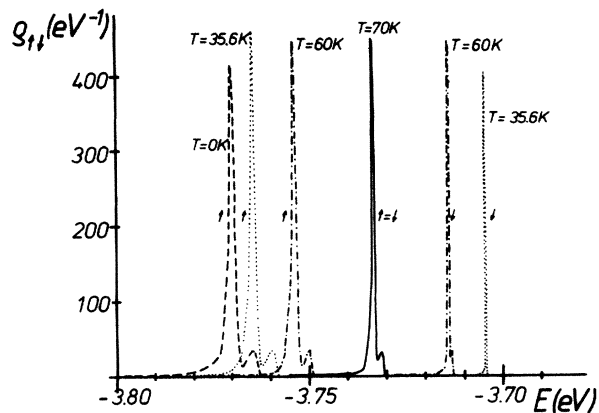


FIG. 9. Low-energy  $4f$ -like peak in the quasiparticle density of states as function of energy for several temperatures and both spin directions. The bare  $4f$  energy is  $E_f = -3.7$  eV.

The  $T=0$  results, which follow from our renormalized many-body procedure (Sec. II), are plotted in Fig. 4. The  $\uparrow$  spectrum is only rigidly shifted relative to the BDOS (Fig. 2), as required by (2.29). The  $4f\uparrow$  peak is very sharp and appears below the conduction-band region. In the  $\downarrow$  spectrum the  $4f$  peak is located within the transition region from  $5d$ -like to  $6s$ -like states ( $m=5,6$ ). Strong hybridization causes that the high-energy  $4f\downarrow$  peak is substantially broader than the low-energy  $4f\uparrow$  peak, both having, however, the same spectral weight. The  $d$ - $f$  exchange  $H_{ex}$  (2.4) takes care of a distinct splitting of  $5d/6s\uparrow$  and  $5d/6s\downarrow$  spectra. Corresponding peaks are shifted against one another up to 1 eV. The relative shift is, however, not at all rigid, mainly because of the dynamic spin-flip terms in (2.4), which describe the spin-exchange processes between the  $4f$ -moment system and  $5d/6s$  conduction electrons. In the lower part of the  $5d/6s$  spectrum ( $m=1,2,3$ ), the influence of the  $4f\downarrow$  peak is negligible. The QDOS in this region is of course identical with that presented in Ref. 1.

Figure 5 shows the QDOS for  $T=35.6$  K, where the magnetization is still rather high  $\langle S^z \rangle / S = \frac{6}{7}$ . We recognize a very sharp high-energy  $4f\uparrow$  peak and also a low energy  $4f\downarrow$  peak, both disappearing at  $T=0$ . Some  $4f\uparrow$  weight has been shifted from the low- to the high-energy peak and some  $4f\downarrow$  weight in opposite direction from the high- to the low-energy peak. The sum of the areas under the two peaks, however, remains constant, and that for both spin directions and all temperatures. Furthermore, the high-energy  $\uparrow$  peak has the same weight as the low-energy  $\downarrow$  peak and vice versa. With increasing temperature (Figs. 6–8), the upper  $4f\uparrow$  peak becomes broader and the upper  $4f\downarrow$  peak narrower. The exchange splitting of the  $5d/6s$  spectrum is roughly proportional to the magnetization  $\langle S^z \rangle$ . In the paramagnetic region  $T \geq T_C$  (see Fig. 8), both spin spectra are of course identical.

In our model the temperature dependence of the localized ferromagnetism manifests itself in the QDOS in a

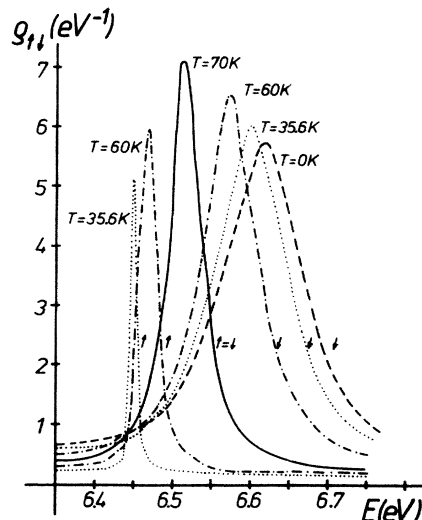


FIG. 10. High-energy  $4f$ -like peak in the quasiparticle density of states as function of energy for several temperatures and both spin directions.

temperature-dependent shift of spectral weight between a low- and a high-energy peak, which both exist for finite temperatures in each spin spectrum,  $T=0$  is a special case with only one peak per spin spectrum. The positions of these peaks are only slightly temperature dependent as can be seen in Figs. 9 and 10, where we have plotted the peaks for various temperatures on a greater scale. If we denote the area under the low-energy  $4f$  peak by  $A_{L\sigma}$  and that under the high-energy  $4f$  peak of the  $\sigma$  spectrum by  $A_{H\sigma}$ , then we can find, that

$$(A_{L\uparrow} - A_{L\downarrow}) / (A_{L\uparrow} + A_{L\downarrow}) = (A_{H\downarrow} - A_{H\uparrow}) / (A_{H\downarrow} + A_{H\uparrow})$$

equals the normalized magnetization  $\langle S^z \rangle / S$ . This interpretation of “localized” ferromagnetism is conceptually different from the “Stoner picture,” by which the demagnetization with increasing temperature is explained by an increasing overlap of  $\uparrow$  and  $\downarrow$  “bands.”

Let us finally point out a special detail, which we already mentioned at the end of Sec. II B. Each of the seven  $4f$  levels per spin direction hybridizes with the conduction-band states with a spectral weight of  $\frac{1}{7}$ , only. The residual weight  $\frac{6}{7}$  does not take part in the mixing. The total area under the two  $4f$ -like peaks in the QDOS, which are plotted in Figs. 4–10, is therefore 1 per spin direction, i.e.,  $A_{L\sigma} + A_{H\sigma} = 1$  for  $\sigma = \uparrow$  and  $\downarrow$ . In addition, there are two sharp  $\delta$  peaks at the energies  $E_f$  and  $E_f + U_f$  with temperature-dependent spectral weights  $3(1 + z_\sigma \langle S^z \rangle / S)$  and  $3(1 - z_\sigma \langle S^z \rangle / S)$ , respectively. These  $\delta$  peaks are not plotted in Figs. 4–10.

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