Dynamical corrections to density-functional theory for quasiparticles in ferromagnetic 4f systems. I. $T=0$ results for EuO

W. Nolting
Institut für Theoretische Physik II, Westfälische Wilhelms Universität, D-4400 Münster Domagkstrasse 75, Federal Republic of Germany

G. Borstel
Fachbereich Physik der Universität Osnabrück, D-4500 Osnabrück, Federal Republic of Germany

W. Borgiel*
Institut für Theoretische Physik II, Westfälische Wilhelms Universität, D-4400 Münster Domagkstrasse 75, Federal Republic of Germany

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A theory for the electronic quasiparticle spectrum of ferromagnetic 4f systems is presented and applied to the semiconductor EuO. The starting point is a d-f exchange model, which we solve exactly for $T=0$. One of the results is a simple relationship between the spin-up quasiparticle energies and the "free" Bloch energies $\varepsilon_m(\mathbf{k})$, which we use to fix the $\varepsilon_m(\mathbf{k})$ in a highly realistic manner by performing a new self-consistent spin-polarized band-structure calculation based on density-functional theory. With the so-determined Bloch energies we investigate the spin-down quasiparticle spectrum, which exhibits even at $T=0$ strong many-body effects as a consequence of spin-exchange processes between localized magnetic 4f moments and itinerant conduction electrons. We discuss in detail the spin-down quasiparticle spectral density for the ΓL direction, which should be observable in an inverse photoemission experiment. The shape of this function is strongly $\mathbf{k}$ dependent, revealing different types of quasiparticles. The prominent quasiparticle peaks in the spin-down quasiparticle spectral density are used to construct a quasiparticle band structure, which shows some striking deviations from the one-particle solution of the density-functional theory. Furthermore, results for the electronic self-energy and the quasiparticle density of states are presented.

I. INTRODUCTION

Typical magnetic semiconductors such as the europium chalcogenides EuX ($X=$O, S, Se, Te) (Ref. 1) and the chalcogenide spinels $M\mathrm{Cr}_2\mathrm{Y}_4$ ($M=$Hg, Cd; Y=S, Se) (Refs. 2 and 3) have been the subject of extensive experimental as well as theoretical research work. In particular, the EuX have attracted scientific interest as have very few other groups of solid compounds. Theoretical treatments of these magnetic 4f systems are exclusively based on the so-called s-f model,[4,5] which is commonly accepted as a reasonable starting point, being, however, exactly solvable only for some limiting cases. For a direct comparison with experimental data the up-to-now proposed approaches bear some serious deficiencies. For mathematical simplicity the conduction band is normally considered as an s band, while that of the EuX, e.g., has mainly d character. Furthermore, the Bloch density of states (B-DOS) of the noninteracting electron system has turned out to be a decisive model parameter. The usually taken, simply-shaped functions (rectangular, triangular, semielliptic,...) can help us to a good understanding of the general physics of the s-f exchange model, but are useless for a concrete evaluation of, say, a photoemission experiment. For that purpose a realistic predetermination of the B-DOS is absolutely necessary. We have therefore developed a theory for the electronic quasiparticle energy spectrum of ferromagnetic semiconductors like EuO and EuS, which combines a reliable many-body treatment of the s-f (better: d-f) exchange model with a new self-consistent band-structure calculation based on density-functional theory. With the use of a very similar method we have recently calculated the quasiparticle density of states (Q-DOS) of the ferromagnetic 4f metal Gd.[6] In this paper we present $T=0$ results for EuO. A theory for finite temperatures wil be published in a following paper.

As to their purely magnetic properties, 4f systems like EuO may be considered as sufficiently well understood. Particularly, the ferromagnet EuO has turned out to be an almost ideal realization of the abstract Heisenberg-Dirac exchange model,[7-9] where the magnetic moments stem from the just half-filled 4f shells of the Eu^{2+} ions. A special kind of superexchange mechanism[10] assures that the moments order ferromagnetically below the Curie temperature $T_c=69.33$ K[1].

The Heisenberg-Dirac model is of course overtaxed in connection with effects for which the conduction band plays an active role. The well-known red shift of the optical absorption edge for electronic $4f\rightarrow(5d,6s)$ transitions, appearing with decreasing temperature below T_c,[1,11,12] is a striking indicator for a strong correlation between the localized magnetic 4f states and the itinerant conduction-band states. As a consequence of a 4f-(5d,6s) exchange interaction, the electronic quasiparticle energy spectrum

gets a strong dependence on the magnetic state of the localized $4f$ moment system, and therewith a remarkable temperature dependence. Other experiments, which have been successfully interpreted in terms of this exchange interaction, are the spin-filter properties of W-EuS junctions,[13–15] the band-filling dependence of the Curie temperature of EuO,[16,17] the insulator-metal transition in Eu-rich EuO,[18] the anomalous spin polarization of $4f$ electrons photoemitted from Gd-doped EuO,[19] and the pressure-induced transition into an intermediate valence phase observed for EuO.[20–22] For a qualitative understanding of all these experiments the already mentioned s-f model[4,5] has turned out to be an excellent theoretical framework. We extend the model in this paper by taking explicitly into account the $5d$ character of the conduction band. The decisive term is an intra-atomic exchange interaction between the localized $4f$ states and the conduction-band states, which provokes a nontrivial many-body problem. For $T=0$, however, the model is exactly solvable.[23–26] It turns out that in this special case the spin-up quasiparticle density of states ($\uparrow$-Q-DOS) is identical in shape with the "free" B-DOS, only rigidly shifted by a constant energy amount. This gives us the possibility to identify this important model parameter with the result of a self-consistent spin-up spin-polarized band-structure calculation, therewith taking automatically into account all the other electron-electron interactions in the ferromagnetic EuO, which are not explicitly covered by the d-f model Hamiltonian. The only remaining model parameter is then the d-f exchange constant, which we fix by fitting the total red shift of the lower conduction-band edge to the experimental data for the corresponding shift of the optical absorption edge.[1,16]

The paper is organized as follows. In Sec. II we introduce the model, which we believe to be realistic for a description of the electronic quasiparticle spectrum of the ferromagnetic semiconductor EuO. Section III presents the exact $T=0$ solution. The self-consistent spin-up band-structure calculation based on density-functional theory, by which we get the B-DOS, is described in Sec. IV. The results for the $T=0$ spin-down quasiparticle spectrum are discussed in Sec. V in terms of self-energies, spectral densities, and quasiparticle densities of states. We present as a final result the full "many-body corrected" spin-down band structure showing some striking deviations from the predictions of the density-functional theory.

II. THE MODEL

It is well known that many characteristic properties of $4f$ systems like EuO are caused by an intimate correlation between two well-defined electronic subsystems. The first is built up by the seven $4f$ electrons of the Eu^{2+} ions. The just half-filled $4f$ shell lies deeply within the core, and is therefore so totally screened by the outer, completely filled shells ($5s$,$5p$), that the overlap of $4f$ wave functions centered at different sites is rather negligible.[27] The $4f$ electrons are to be regarded as strictly localized forming magnetic moments with $J=S=\frac{7}{2}$ according to Hund's rule. As a consequence of a superexchange coupling[1,10] they order ferromagnetically below $T_c=69.33$ K. This subsystem is surely very realistically described by the Heisenberg-Dirac exchange model

$$H_f=-\sum_{i,j} J_{ij}\mathbf{S}_i\cdot\mathbf{S}_j \ , \tag{1}$$

where J_{ij} are the corresponding exchange integrals. Since EuO crystallizes in the rocksalt structure, and the spins $\mathbf{S}_i$ are confined to Eu^{2+} ions, the double sum runs over all sites of an fcc lattice.

The other subsystem is that of the conduction electrons. According to previous band-structure calculations of Cho[27] the conduction-band consists of hybridized $5d$- and $6s$-bands, all being built up by original Eu states.

$$H_c=\sum_{\substack{\mathbf{k}\sigma\\ m}} \varepsilon_m(\mathbf{k})c^{\dagger}_{\mathbf{k}m\sigma}c_{\mathbf{k}m\sigma} \tag{2}$$

$$=\sum_{\substack{ij\sigma\\ m}} T_{ij}^{(m)}c^{\dagger}_{im\sigma}c_{jm\sigma} \ , \tag{3}$$

where $c^{\dagger}_{\mathbf{k}m\sigma}$ ($c_{\mathbf{k}m\sigma}$) is the creation (annihilation) operator of a conduction electron with wave vector $\mathbf{k}$, band index m, and spin σ ($\sigma=\uparrow,\downarrow$). $\varepsilon_m(\mathbf{k})$ are considered to be renormalized Bloch energies, formulated in a suitable one-particle basis. As already mentioned in the Introduction we shall use the results of a new self-consistent band-structure calculation, in order to take into account in the best possible way the interaction of the conduction electron with all the other electrons of the solid. Details are explained in Sec. IV. The hopping integrals $T_{ij}^{(m)}$ are connected with the Bloch energies $\varepsilon_m(\mathbf{k})$ by

$$T_{ij}^{(m)}=\frac{1}{N}\sum_{\mathbf{k}} \varepsilon_m(\mathbf{k})e^{i\mathbf{k}\cdot(\mathbf{R}_i-\mathbf{R}_j)} \ , \tag{4}$$

where N is the number of unit cells in the crystal. $c^{\dagger}_{im\sigma}$ and $c_{jm\sigma}$ used in (3), are the construction operators in Wannier representation:

$$c_{im\sigma}=\frac{1}{\sqrt{N}}\sum_{\mathbf{k}} c_{\mathbf{k}m\sigma}e^{i\mathbf{k}\cdot\mathbf{R}_i} \ . \tag{5}$$

A Coulomb term, which describes the electron-electron interaction within the conduction band, is not necessary for the semiconductor EuO. It is sufficient in the following to consider a single electron in an otherwise empty band.

The most decisive part of our model Hamiltonian is the (d,s)-f exchange interaction, which is expected to provoke a sensitive reaction of the conduction-band density of states on the magnetic state of the f system manifesting itself particularly in a strong temperature dependence. This fundamental exchange interaction is well described by the following effective Hamiltonian:[4,5,28]

$$H_{\mathrm{ex}}=-\frac{1}{2N}\sum_{i,\sigma}\sum_{\mathbf{k},\mathbf{q}}\sum_{n,m} g_{nm}(\mathbf{k},\mathbf{k}+\mathbf{q})e^{-i\mathbf{q}\cdot\mathbf{R}_i}$$
$$\times\left(z_{\sigma}S_i^z c^{\dagger}_{\mathbf{k}n\sigma}c_{\mathbf{k}+\mathbf{q}m\sigma}\right.$$
$$\left.+S_i^{\sigma}c^{\dagger}_{\mathbf{k}n-\sigma}c_{\mathbf{k}+\mathbf{q}m\sigma}\right) . \tag{6}$$

Here we have written for abbreviation

$$S_i^\sigma = S_i^x + i z_\sigma S_i^y \ , \tag{7}$$

$$z_\sigma = \begin{cases} +1 & \text{if } \sigma = \uparrow \\ -1 & \text{if } \sigma = \downarrow \end{cases} \ . \tag{8}$$

$g_{n,m}(\mathbf{k},\mathbf{k}+\mathbf{q})$ denotes the exchange coupling constant, where, however, its wave-vector dependence as well as its off-diagonal elements are usually neglected:[28]

$$g_{nm}(\mathbf{k},\mathbf{k}+\mathbf{q}) \rightarrow g_m \delta_{nm} \tag{9}$$

The total model Hamiltonian, which we apply in this paper to the ferromagnetic semiconductor EuO, is finally given by the sum of the three above-defined partial operators:

$$H = H_c + H_f + H_{\text{ex}} \ . \tag{10}$$

This operator provokes a complicated many body problem,[5] which in general cannot be treated rigorously. For $T=0$, however, an exact solution is possible. This is presented in Sec. III.

III. $T=0$ SOLUTION

All information, which we need, can be derived from the (retarded or advanced) one-electron Green-function:

$$G_{ij\sigma}^{(m)}(E) = \langle\langle c_{im\sigma}; c_{jm\sigma}^\dagger \rangle\rangle_E \ , \tag{11}$$

$$G_{\mathbf{k}\sigma}^{(m)}(E) = \langle\langle c_{\mathbf{k}m\sigma}; c_{\mathbf{k}m\sigma}^\dagger \rangle\rangle_E = \frac{1}{N} \sum_{i,j} G_{ij\sigma}^{(m)} e^{-i\mathbf{k}(\mathbf{R}_i - \mathbf{R}_j)} \ . \tag{12}$$

We determine this function for the special situation of a single electron in an otherwise empty conduction band (band occupation $n=0$), and that at $T=0$;[23-26] a situation which corresponds to the ferromagnetically saturated semiconductor EuO. In this case the required averaging processes in (11) and (12) can be performed with the electron and magnon vacuum $|0\rangle$, so that the problem becomes exactly solvable. A further simplification is possible since we are mainly interested in the *electronic* excitation spectrum, only. Then it is surely allowed to neglect the term H_f in the total Hamiltonian (10), because magnon energies are smaller by some orders of magnitude than typical electronic quantities like the d-f coupling g_m or the Bloch band width W_m. Although in principle not necessary, the neglect of H_f simplifies the procedure somewhat.

We use the equation of motion method for the determination of $G_{\mathbf{k}\sigma}^{(m)}(E)$. A straightforward calculation yields the following equation of motion

$$E G_{ij\sigma}^{(m)}(E) = \hbar \delta_{ij} + \sum_k T_{ik}^{(m)} G_{kj\sigma}^{(m)}(E) - \tfrac{1}{2} g_m [z_\sigma D_{ii,j\sigma}^{(m)}(E) + F_{ii,j\sigma}^{(m)}(E)] \ , \tag{13}$$

where D and F are "higher" Green functions

$$D_{ik,j\sigma}^{(m)}(E) = \langle\langle S_i^z c_{km\sigma}; c_{jm\sigma}^\dagger \rangle\rangle_E \ , \tag{14}$$

$$F_{ik,j\sigma}^{(m)}(E) = \langle\langle S_i^{-\sigma} c_{km-\sigma}; c_{jm\sigma}^\dagger \rangle\rangle_E \ . \tag{15}$$

Because of the presumption ($T=0$, $n=0$) we can write for D,

$$D_{ik,j\sigma}^{(m)}(E) = S G_{kj\sigma}^{(m)}(E) \tag{16}$$

while F defines the electronic self-energy $M_{ij\sigma}^{(m)}(E)$:

$$-\tfrac{1}{2} g_m F_{ii,j\sigma}^{(m)}(E) = \sum_k M_{ik\sigma}^{(m)}(E) G_{kj\sigma}^{(m)}(E) \ . \tag{17}$$

This ansatz formally solves the equation of motion (13), if one still uses a Fourier transformation on wave vectors:

$$G_{\mathbf{k}\sigma}^{(m)}(E) = \hbar [E - \varepsilon_m(\mathbf{k}) + \tfrac{1}{2} g_m z_\sigma S - M_{\mathbf{k}\sigma}^{(m)}(E)]^{-1} \ . \tag{18}$$

According to Eq. (17) the electronic self-energy is practically identical with the Green function $F_{ii,j\sigma}^{(m)}$ therefore being mainly determined by spin-exchange processes between the localized moment system and the conduction electron. Such exchange processes are of course impossible for a spin-up electron and the totally parallel aligned f system; $F_{ii,j\uparrow}^{(m)}(E)$ therefore vanishes. Formally this follows from Eq. (15) because of $S_i^\dagger |0\rangle = \langle 0| S_i^- = 0$. For $T=0$, $n=0$ the spin-up self-energy is zero, and the quasiparticle spectrum

$$E_{m\uparrow}^{(T=0)}(\mathbf{k}) = \varepsilon_m(\mathbf{k}) - \tfrac{1}{2} g_m S \tag{19}$$

leads to a quasiparticle density of states (Q-DOS), which is identical in shape with the original Bloch density of states (B-DOS) $\rho_0^{(m)}(E)$, only shifted by a constant energy amount:

$$\rho_{m\uparrow}^{(T=0)}(E) = \rho_0^{(m)}(E + \tfrac{1}{2} g_m S) \ . \tag{20}$$

This exact result is of decisive importance for our further procedure, because it provides us with a direct possibility to fix the B-DOS. $\rho_0^{(m)}$ is an essential model parameter and should therefore be determined as realistically as possible. As far as ground-state properties are concerned, self-consistent band-structure calculations, which are based on density-functional theory are commonly accepted as highly reliable. Since our theory tells us that at $T=0$ the spin-up spectrum of the interacting system is quasi-identical with the Bloch spectrum $\varepsilon_m(\mathbf{k})$, it suggests using the result of a spin-up spin-polarized band-structure calculation for the input parameter $\rho_0^{(m)}(E)$. This we have actually done. Details are explained in Sec. IV.

Contrary to the spin-up spectrum the spin-down spectrum is nontrivial, because a spin-down electron has even at $T=0$, the possibility of exchanging its spin with the f system, e.g., by emitting a magnon. The spin-flip function $F_{ik,j\downarrow}^{(m)}(E)$ is therefore rather complicated. Its equation of motion read as

$$E F_{ik,j\downarrow}^{(m)}(E) = \sum_p T_{kp}^{(m)} F_{ip,j\downarrow}^{(m)}(E) - \tfrac{1}{2} g_m [H_{ikk,j\downarrow}^{(m)}(E) + L_{ikk,j\downarrow}^{(m)}(E)] \ . \tag{21}$$

In the limit $n=0$, $T=0$ the "higher" Green functions on the right-hand side decouple exactly:

$$H^{(m)}_{ikk,j\downarrow}(E)=\langle\langle S_i^+ S_k^z c_{km\uparrow};c^\dagger_{jm\downarrow}\rangle\rangle_E \underset{T=0,n=0}{\rightarrow}(S-\delta_{ik})F^{(m)}_{ik,j\downarrow}(E)\ , \tag{22}$$

$$L^{(m)}_{ikk,j\downarrow}(E)=\langle\langle S_i^+ S_k^- c_{km\downarrow};c^\dagger_{jm\downarrow}\rangle\rangle_E \underset{T=0,n=0}{\rightarrow}2S\delta_{ik}G^{(m)}_{ij\downarrow}(E)\ . \tag{23}$$

Equations (13)–(17) and (21)–(23) build a closed system of equations for the spin-down self-energy, which can be solved by Fourier transformation. As a consequence of the neglected magnon dispersion ($H_f\equiv 0$) the resulting self-energy is wave-vector independent:

$$M^{(m)}_\downarrow(E)=R^{(m)}_\downarrow(E)+iI^{(m)}_\downarrow(E) =\tfrac{1}{2}g_m^2 S\frac{B^{(m)}_{0\uparrow}(E)}{1-\tfrac{1}{2}g_m B^{(m)}_{0\uparrow}(E)}\ , \tag{24}$$

$$B^{(m)}_{0\uparrow}(E)=\frac{1}{N}\sum_{\mathbf{p}}[E-\varepsilon_m(\mathbf{p})+\tfrac{1}{2}g_m S]^{-1}\ . \tag{25}$$

It is interesting that the imaginary part of $B^{(m)}_{0\uparrow}$ is nothing else than the ↑-Q-DOS:[20]

$$\mathrm{Im}B^{(m)}_{0\uparrow}(E)=-\pi\rho^{(m)}_0(E+\tfrac{1}{2}g_m S) =-\pi\rho^{(T=0)}_{m\uparrow}(E)\ . \tag{26}$$

The imaginary part $I^{(m)}_\downarrow(E)$ of the spin-down self-energy is therefore unequal to zero just in that energy region, where the ↑-Q-DOS appears.

Physical quantities of special importance are the one-electron spectral density

$$A^{(m)}_{\mathbf{k}\downarrow}(E)=-\frac{1}{\pi}\mathrm{Im}G^{(m)}_{\mathbf{k}\downarrow}(E+i0^+) =\frac{\hbar}{\pi}\frac{0^+-I^{(m)}_\downarrow(E)}{[E-R^{(m)}_\downarrow(E)-\tfrac{1}{2}g_m S-\varepsilon_m(\mathbf{k})]^2+[0^+-I^{(m)}_\downarrow(E)]^2} \tag{27}$$

and the ↓-Q-DOS:

$$\rho^{(m)}_\downarrow(E)=\frac{1}{N\hbar}\sum_{\mathbf{k}}A^{(m)}_{\mathbf{k}\downarrow}(E)\ . \tag{28}$$

By use of the B-DOS

$$\rho^{(m)}_0(E)=\frac{1}{N}\sum_{\mathbf{k}}\delta[E-\varepsilon_m(\mathbf{k})] \tag{29}$$

we can replace the $\mathbf{k}$ summation by an energy integration:

$$\rho^{(m)}_\downarrow(E)=\rho^{(m)}_0[E-R^{(m)}_\downarrow(E)-\tfrac{1}{2}g_m S]\quad \text{if } I^{(m)}_\downarrow=0\ , \tag{30}$$

$$\rho^{(m)}_\downarrow(E)=-\frac{1}{\pi}I^{(m)}_\downarrow(E)\int_{-\infty}^{+\infty}dx\frac{\rho^{(m)}_0(x)}{[E-R^{(m)}_\downarrow(E)-\tfrac{1}{2}g_m S-x]^2+I^{(m)2}_\downarrow(E)}\quad \text{if } I^{(m)}_\downarrow\neq 0\ . \tag{31}$$

These results give evidence how sensitively the concrete conclusions of our model will depend on a proper choice of the B-DOS, for which we present a self-consistent band-structure calculation in Sec. IV.

IV. SELF-CONSISTENT $T=0$ BAND-STRUCTURE CALCULATIONS

The theory presented in Sec. III resulted in a simple relationship (19) between the spin-up quasiparticle spectrum $E^{(T=0)}_{m\uparrow}(\mathbf{k})$ and the one-particle spectrum $\varepsilon_m(\mathbf{k})$. It has already been mentioned that this fact suggests to use the results of a band-structure calculation as input of our many-body procedure. Since $\varepsilon_m(\mathbf{k})$ and the B-DOS $\rho^{(m)}_0$ should be fixed as realistic as possible and existing band calculations for EuO (Refs. 27 and 29) are neither self-consistent nor treat exchange and correlation effects on modern grounds, we decided to perform for EuO a new self-consistent spin-polarized band calculation based on the density-functional theory (DFT). The effective single-particle states $\varphi_m(\mathbf{k})$ of DFT obtained from such a calculation then form an optimal renormalized one-particle basis set for the many-body procedure which takes into account those interactions which are not explicitly contained in the d-f model.

Using the experimental lattice constant $a=5.141$ Å [1] and the prescription of Moruzzi *et al.*[30] for the spin-polarized exchange and correlation potential V_{xc} of DFT, we obtained from a nonrelativistic augmented spherical wave (ASW) calculation[31] the majority spin bands $\varepsilon_{m\uparrow}(\mathbf{k})$ as shown in Fig. 1. The corresponding results for the down-spin system will be greatly modified by many body effects, even for $T=0$, and therefore will be presented in Sec. V.

Relevant band-structure data of our calculation are given in Table I and are compared with the results of oth-

TABLE I. Band-structure data (eV) for EuO.

	Ref. 27	Ref. 29	Present results
$4f(\uparrow)$ bandwidth	0.57	0.31	0.55
$2p$ bandwidth	2.12	1.19(↑), 1.39 (↓)	2.11
$4f(\uparrow)$-$2p$ separation	1.41	5.40(↑)	2.56
$5d(\uparrow)$ bandwidth	~9.1	~7.6	8.67
$L_3(\uparrow)$-$X_3(\uparrow)$			
$X_3(\uparrow)$-$2p$ gap	3.61	6.88(↑), 7.26(↑)	3.18
$X_3(\uparrow)$-$4f(\uparrow)$ gap	1.12	1.17	0.07

er authors. With the exception of the sizes of the two band gaps we find all our energy data lying in between the corresponding results of Cho[27] and those of Farberovich and Vlasov.[29] The fact that our present self-consistent $X_3(\uparrow)$-$4f(\uparrow)$ band gap of 0.07 eV is much smaller than the reported experimental value of 1.12 eV,[1] shows, dramatically, the failure of DFT to predict accurate band gaps for semiconductors and insulators.[32] Since our calculated result for the $X_3(\uparrow)$-$2p$ gap (3.18 eV) is not as bad, when compared with the experimental value of 3 eV,[1] we conclude that the unknown gap correction to the DFT gap[33] increases drastically with the degree of localization of the states forming the gap. We note in passing that this result puts a serious question mark on the recent findings of Vlasov and Farberovich[34] concerning the insulator-metal transition in EuO under pressure.

Figure 2 shows the calculated one-electron density of states (B-DOS) for the first five conduction bands and the resulting total B-DOS for the EuO majority spin system. In what follows we will concentrate on these five conduction bands, which are mainly Eu-5d-like, and disregard all higher-lying bands. The sixth conduction band is predominantly Eu-6s-like and would produce a smooth background in Fig. 2. This and the fact that the s-f exchange constant should be small compared to the d-f constant[28] justifies its neglect in the many-body procedure.

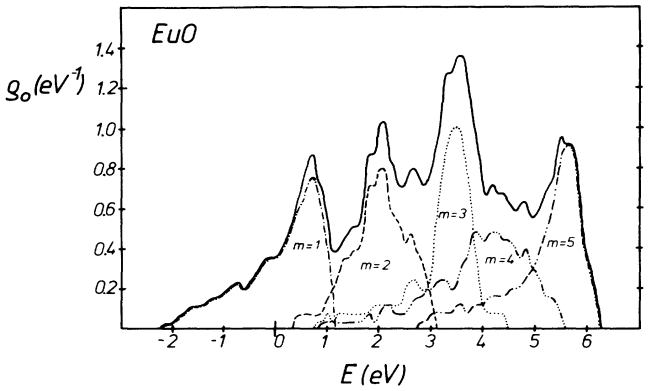

FIG. 2. Total Bloch density of states ρ_0 per atom (solid line) of EuO as a function of energy, calculated for the first five conduction bands, which are mainly of Eu^{2+}-5d character. The partial densities of states ($m = 1, \ldots, 5$) are also indicated. The energy zero is chosen to coincide with the center of gravity of the low-energy $m = 1$ subband.

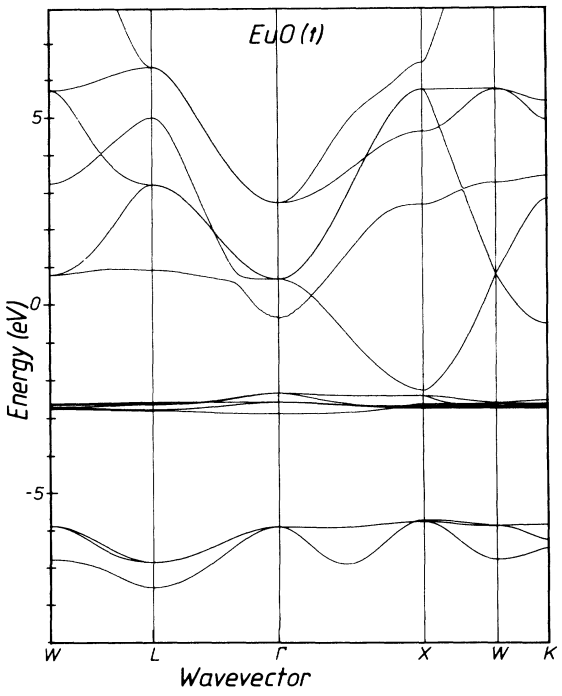

FIG. 1. Ground-state spin-up band structure of EuO as a function of the wave vector, obtained from a nonrelativistic ASW calculation (Ref. 31).

V. DISCUSSION OF THE RESULTS

Our investigation focuses on the electronic quasiparticle spectrum of the ferromagnetic semiconductor EuO. Basic framework for our study is the exchange model (10), the solution of which needs a many-body theoretical treatment. In this paper we concentrate ourselves on the $T = 0$ solution, which can be found rigorously. The model contains some important parameters, one of which is the Bloch density of states $\rho_0^{(m)}(E)$. Via the exact relation (19) we were able to construct a direct connection between our many-body theory and a spin-polarized band-structure calculation, which permitted a highly realistic determination of the B-DOS. A further important model parameter is the exchange coupling constant g_m. One of the present authors has shown[35] that for the "normal" s-f model[5] this exchange-quantity together with the Bloch bandwidth decisively determines the red shift of the optical absorption edge. For the semiconductor EuO the latter is of course defined by the lower edge of the conduction band, more strictly by the lower edge of the $m = 1$ subband. We shall show in our next paper, which will deal with the finite temperature results, that the value of 0.2 eV for the exchange constant, previously proposed and applied in Refs. 5, 15, 17, 22, and 35, indeed leads to a temperature-dependent edge shift in EuO, which has an excellent fit with the experimental data.[16] In this respect we have a realistic choice for g_1, the coupling constant for

the exchange between the f states and the lowest d subband. We see, however, no direct possibility to fit the exchange constants of the other d subbands. All g_m will surely be of the same order of magnitude. Somewhat arbitrarily, we have therefore assumed all g_m to be equal:

$$g_m = 0.2 \text{ eV}, \quad m = 1, \ldots, 5 . \tag{32}$$

In Fig. 3 we have plotted the total spin-polarized Q-DOS as a function of energy. A strong exchange splitting between the spin-up and the spin-down spectrum is observed, prominent peaks are shifted by energies up to 1 eV. The energy-shift is, however, not at all rigid. The spin-exchange processes between the f level and conduction band, which according to Eqs. (15) and (17) mainly influence the spin-up self-energy, take care for substantial deformations on the original B-DOS. It is an interesting detail that the lower edges of the spin-up and the spin-down spectrum exactly coincide, cf. inset in Fig. 3. The spin-down density is, however, very much smaller near the lower edge than the spin-up state density. The details of the Q-DOS will be discussed in connection with Figs. 4–8. For comparison, we have inserted into Fig. 3 the results of the spin-density-functional theory (see Sec. IV). The spin-up spectra of DFT and of our theory of course coincide because of (19). The spin-down spectra, however, exhibit striking differences. One main reason is the presence of the $f\downarrow$ peak near 2 eV in the d region, as predicted by the DFT, which according to our many-body ansatz does not exist. The $d\downarrow$-$f\downarrow$ hybridization in DFT obviously pushes the lower part of the $d\downarrow$ spectrum to lower energies and the upper part to higher energies with the net effect of decreasing the exchange splitting of the d-bands in the lower part. This is clearly demonstrated by the insert, where we have plotted the Q-DOS only for the lowest ($m = 1$) subband. The exchange shift between spin-up and spin-down spectra is, in our theory, very much stronger than predicted by the DFT.

In Figs. 4–8 we analyze the five d subbands ($m = 1, \ldots, 5$) in detail. The upper parts show the one-electron spectral density for five different $\mathbf{k}$ values, which are chosen representatively from the ΓL direction. This function $A^{(m)}_{\mathbf{k}\uparrow\downarrow}(E)$ should be observable directly in an inverse photoemission experiment.[36] The spin-up spectral density is always a sharp δ function at the energy $[\varepsilon_m(\mathbf{k}) - \frac{1}{2}gS]$ representing a quasiparticle with infinite lifetime. More interesting is the behavior of the spin-down spectral density ($\downarrow$-SD), which introduces different types of quasiparticles. A first general features is that the $\downarrow$-SD becomes unequal to zero in the same rather broad energy region, where the $\uparrow$-Q-DOS is finite. This part of the $\downarrow$-SD results from the fact, that a spin-down electron, being excited into the conduction band, can emit a magnon, thereby reversing its own spin and becoming a spin-up electron. Such a process is of course possible only if there are spin-up states within reach. That is the physical reason, why this "scattering part" of $A^{(m)}_{\mathbf{k}\downarrow}(E)$ exactly coincides with $\rho^{(m)}_{\uparrow}(E)$, as can be seen in the lower parts of Figs. 4–8. If we did not neglect from the very beginning

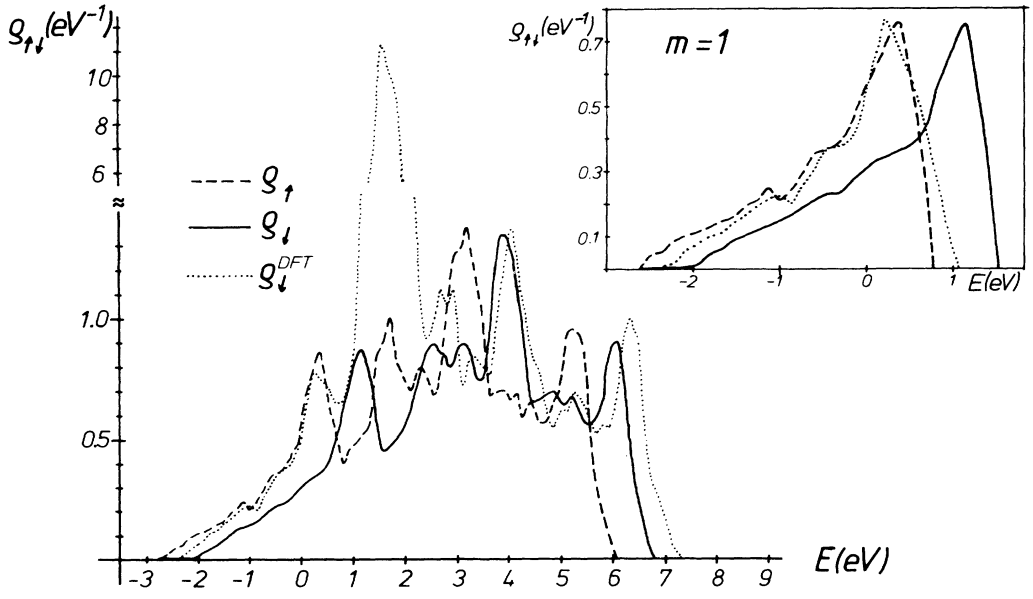

FIG. 3. Total quasiparticle densities of states $\rho_{\uparrow}$ and $\rho_{\downarrow}$ per atom as functions of energy for ferromagnetic EuO at $T = 0$. The solid line is $\rho_{\downarrow}$, as it follows from our many body theory. The corresponding result $\rho^{\mathrm{DFT}}_{\downarrow}$ of the density-functional theory is indicated by the dotted line. Note the change of scale for the prominent $f\downarrow$ peak, appearing only in DFT. $\rho_{\uparrow}$ is given by the dashed line. The inset shows the partial quasiparticle densities of states for the lowest ($m = 1$) subband, only.

the magnon dispersion there would of course appear a slight energy shift between the "scattering part" of $A_{k\downarrow}^{(m)}$ and $\rho_{\uparrow}^{(m)}$ of about the maximum magnon energy (order of magnitude: 10^{-3} eV). According to our exact solution, (24) and (26), the imaginary part $I_{\downarrow}^{(m)}(E)$ of the spin-down self-energy may be written as

$$I_{\downarrow}^{(m)}(E)=\rho_{\uparrow}^{(m)}(E)F(E)\ , \tag{33}$$

where $F(E)$ is a more or less complicated function of energy. Nevertheless, $I_{\downarrow}^{(m)}(E)$ is predominantly determined by $\rho_{\uparrow}^{(m)}(E)$, since the spin-flip scattering probability of the original spin-down electron is of course higher the more spin-up states are within reach.

In some cases (e.g., in Fig. 4.), however, the equation

$$E-R_{\downarrow}^{(m)}(E)-\tfrac{1}{2}gS-\varepsilon_m(\mathbf{k})=0 \tag{34}$$

has a solution outside the region $\rho_{\uparrow}^{(m)}\neq 0$. The corresponding imaginary part of the self-energy is then zero. In this case the spin-down electron cannot emit a magnon because there are no spin-up states, on to which it could be scattered. The result is a quasiparticle with infinite lifetime ("bound state"), which we shall call in the following the "magnetic polaron." The ↓-SD consists in such a case of two nonconnected parts, namely a broad scattering part and a sharp polaron peak. The cases in Fig. 4 or in Figs. 5–8, showing this SD behavior, belong to $\varepsilon_m(\mathbf{k})$ values from the upper part of the mth subband. On the other hand, the other examples in Figs. 4–8 refer to Bloch energies closer to the center of the respective mth subband. The polaron peak has now been shifted into the scattering region with the consequence of a *finite* lifetime of the magnetic polaron. A measure of the lifetime is the inverse halfwidth of the still rather prominent SD peak. There also appear to be situations (e.g., in Figs. 5 and 6), where the ↓-SD is so much deformed that the definition of a quasiparticle becomes meaningless.

In the lower halves of Figs. 4–8 we have plotted the spin-up and spin-down Q-DOS, belonging to the mth subband, and the real and imaginary parts of the electronic self-energy, all as function of energy E. $\rho_{\uparrow\downarrow}^{(m)}(E)$ concerns

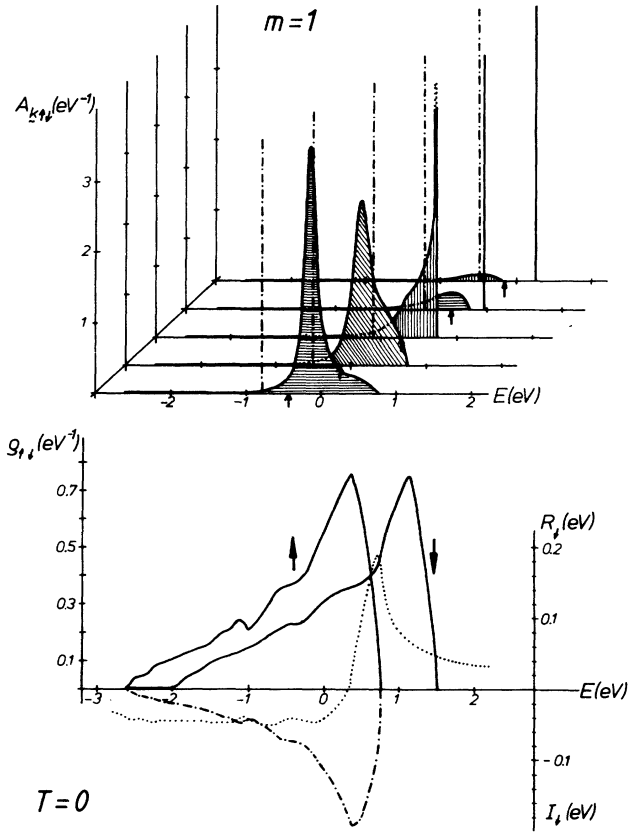

FIG. 4. *Upper half:* One-electron spectral density $A_{\mathbf{k}\uparrow}$ and $A_{\mathbf{k}\downarrow}$ for the $m=1$ subband as functions of energy for five different $\mathbf{k}$ values from the ΓL direction. Solid lines for $A_{\mathbf{k}\downarrow}$; dashed-dotted lines for $A_{\mathbf{k}\uparrow}$. Arrows indicate the position of the corresponding "free" Bloch energy $\varepsilon_1(\mathbf{k})$. *Lower half:* Subband-quasiparticle densities of states per atom $\rho_{\uparrow}$, $\rho_{\downarrow}$ (solid lines) real part $R_{\downarrow}$ (dotted lines), and imaginary part $I_{\downarrow}$ (dashed-dotted line) of the electronic spin-down self-energy as function of energy E.

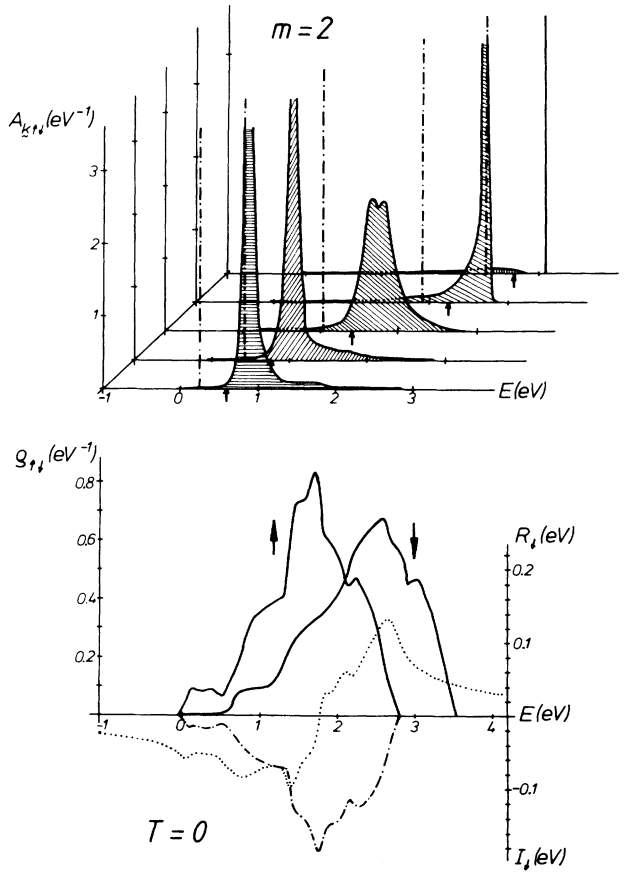

FIG. 5. The same as in Fig. 4, but for the $m=2$ subband.

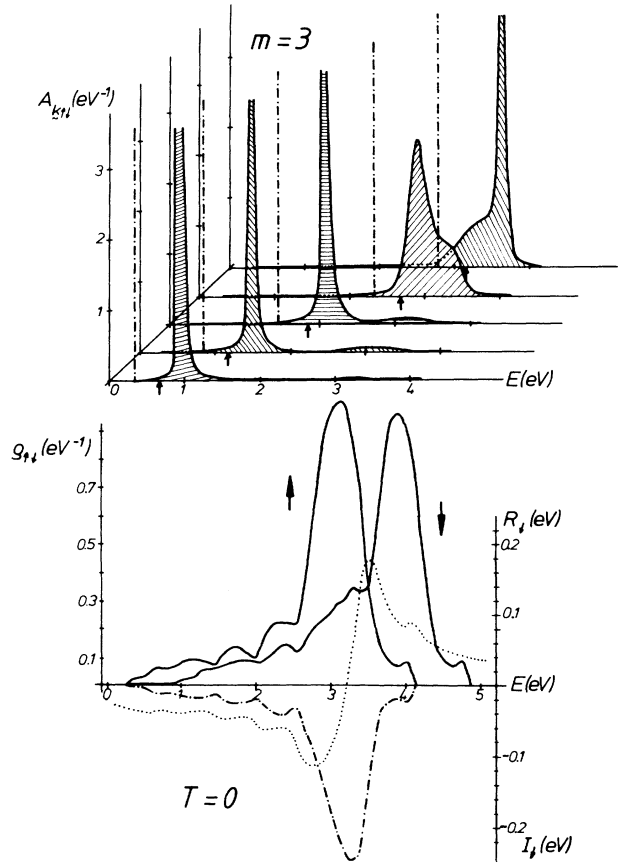

FIG. 6. The same as in Fig. 4, but for the $m=3$ subband.

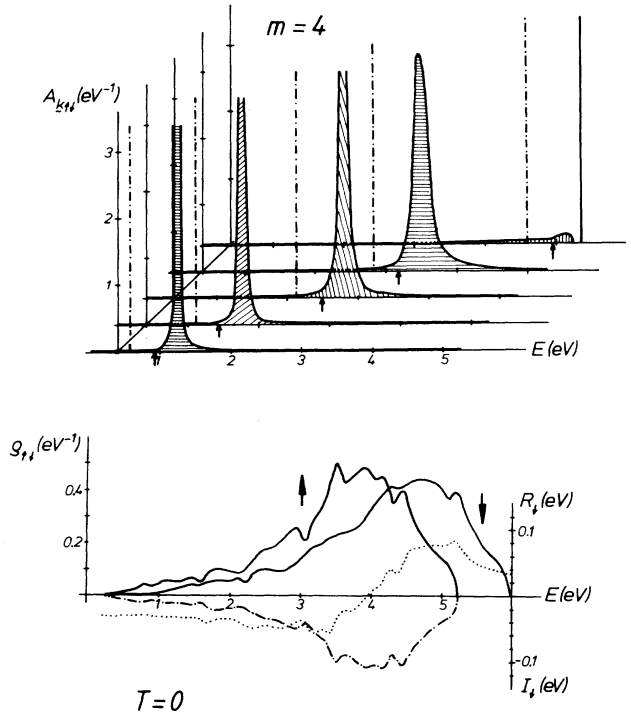

FIG. 7. The same as in Fig. 4, but for the $m=4$ subband.

of course the full mth subband, not only the state density of the ΓL direction. As already mentioned, both Q-DOS $\rho_{\uparrow}^{(m)}$ and $\rho_{\downarrow}^{(m)}$ start at the same energy, where, however, $\rho_{\downarrow}^{(m)}$ is first already rather small. The separation of the more or less pronounced main spin-up and spin-down peaks amounts to about 1 eV. Following the preceding discussion we can read off from the shape of the respective $\downarrow$-SD that the part of $\rho_{\downarrow}^{(m)}$, which lies *above* the upper edge of $\rho_{\uparrow}^{(m)}$ consists of bound states, only. The magnetic polaron has for such energies an infinite lifetime. The part, which overlaps with $\rho_{\uparrow}^{(m)}$ is built up by finitely living quasiparticles as well as by scattering states.

The detailed results for the $\downarrow$-SD allows the construction of a quasiparticle bandstructure, which is pictured in Fig. 9. We identify the quasiparticle energies with the peak position of the spectral density being very often, but not always, identical to the solution of Eq. (34). The dispersion relations following from our theory, consist of pieces with different quasiparticle character. Near the L point the quasiparticles are stable, except for the $m=3$ subband. Near the Γ point we always find quasiparticles with finite lifetimes. In the middle there are regions where a quasiparticle definition becomes doubtful, because the $\downarrow$-SD peaks are too broad. The comparison of our results with those of the DFT calculation reveals some remarkable discrepancies, being particularly large in be-

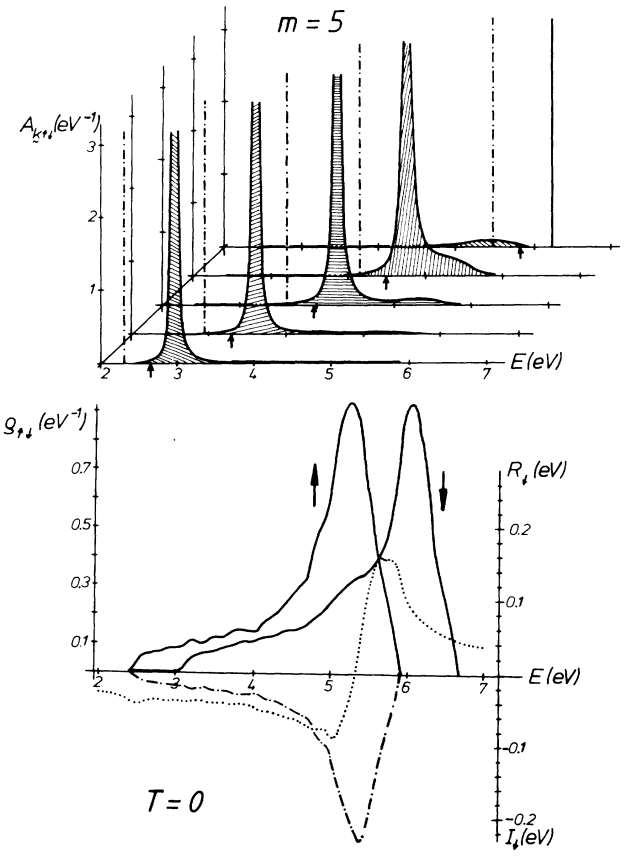

FIG. 8. The same as in Fig. 4, but for the $m=5$ subband.

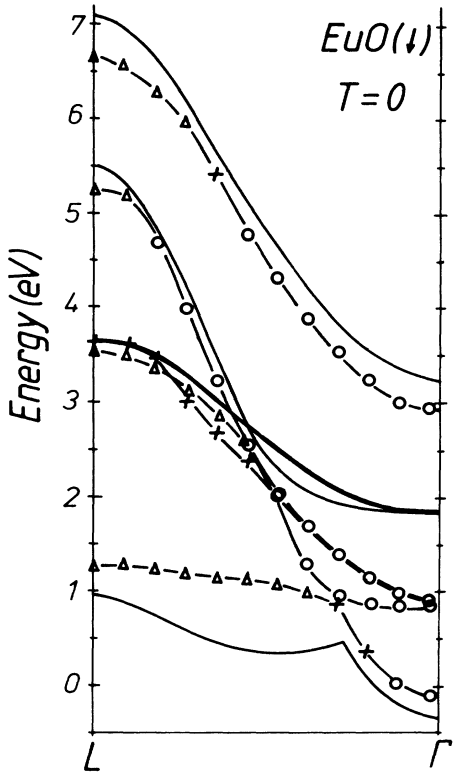

FIG. 9. Full spin-down quasiparticle band structure for EuO at $T=0$ in the ΓL direction. The DFT result is shown for comparison by solid lines. Triangles indicate quasiparticles with infinite lifetimes ("magnetic polaron"), circles correspond to well-defined quasiparticles, but with finite lifetimes, and crosses represent not well-defined quasiparticles.

tween the $m=1$ and $m=2$ subband. In this region the DFT locates the $4f\downarrow$ band, which we did not plot in Fig. 9. It is an interesting detail that the degeneracy at the L_3 point, as predicted by DFT, is removed by our many-body theory. According to our general theory in Sec. III this is due to the propagator $B_{0\uparrow}^{(m)}(E)$, defined in Eq. (25), which has to be calculated by use of the partial density of states $\rho_0^{(m)}(E)$ of the mth subband. $\rho_0^{(2)}(E)$ is different from $\rho_0^{(3)}(E)$, as can be seen in Fig. 2, and the same holds therefore for $B_{0\uparrow}^{(2)}$ and $B_{0\uparrow}^{(3)}$. The propagator $B_{0\uparrow}^{(m)}$, however, determines the electronic self-energy $M_{\downarrow}^{(m)}(E)$, Eq. (24), which is thus different for $m=2$ and $m=3$.

Let us finally point out that the triangles in Fig. 9 mark infinitely living quasiparticles, i.e., sharp δ functions in the respective SD. It goes without saying that this statement should be handled with care. An experiment will of course not at all show δ peaks, which are smeared out by impurities, phonons, not being directly involved in our theory. The simplification (9) also favors the existence of bound magnetic polarons. We shall show in the following paper, that in the paramagnetic region ($T>T_c$) bound states completely disappear.

VI. SUMMARY

We have presented in this paper a theory for the electronic quasiparticle spectrum of ferromagnetic $4f$ systems with a special application to the semiconductor EuO. Basic framework for our study was a d-f exchange model, the most important part of which is an intra-atomic interaction between localized magnetic $4f$ moment and itinerant conduction electrons. The related many-body problem could exactly be solved for $T=0$. The key point of our procedure lies in the fact that for $T=0$ a very simple relationship exists between the spin-up quasiparticle density of states and the one-particle Bloch density of states, which we used to fix the latter as realistic as possible. We performed a new self-consistent spin-polarized band-structure calculation based on density-functional theory. The mentioned simple relationship allowed us to identify the spin-up result with the Bloch density of states, a decisive parameter of our model. Contrary to spin-up spectrum, the spin-down solution exhibits strong many-body effects which we discussed in terms of the spectral density, the real and the imaginary part of the electronic self-energy, and the quasiparticle density of states. The resulting spin-down quasiparticle band structure show some striking deviations from the one-particle result of the density-functional theory.

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*Permanent address: Institute of Physics, Silesian University, Katowice, Poland.

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