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Quasiparticle density of states and edge shifts of doped ferromagnetic semiconductors

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We use a new spectral-density approach to find the temperature behavior of the quasiparticle density of states for the conduction band of a ferromagnetic semiconductor. Because of the exchange interaction between conduction electrons and localized spin system (4*f* moments in the case of Eu chalcogenides), the conduction band splits into several quasiparticle subbands. The famous red shift of the optical absorption edge is almost quantitatively explained as a bandwidth effect of the lower ↑ subband. The striking band-filling dependence of the red shift, first observed by Schoenes and Wachter for $Eu_{1-x}Gd_xO$, finds a natural interpretation by the existence and temperature dependence of quasiparticles in the conduction band of a ferromagnetic semiconductor EuO.

I. INTRODUCTION

Since their discovery about twenty years ago ($CrBr_3$) (Ref. 1) the magnetic semiconductors, and among these especially the Eu chalcogenides,[2] have been studied with a huge effort, both experimentally and from a theoretical point of view. The main reason for this effort was that these materials have turned out to be excellent model substances for certain basic phenomena of magnetism. With respect to their purely magnetic properties the EuX (X=O, S, Se, Te) compounds are surely the best known realizations of the Heisenberg model, defined by the Hamiltonian

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

where the spin operator $\vec{S}_i$, acting at lattice site $\vec{R}_i$, originates from the seven quasilocalized electrons of the half-filled 4*f* shell of the Eu^{2+} ion ($S=\frac{7}{2}$ according to Hund's rule). J_{ij} are the direct exchange integrals. There is some experimental evidence[2] that the nearest-neighbor coupling is ferromagnetic for all Eu compounds, but decreasing from the oxide to the telluride. On the other hand, the next-nearest-neighbor coupling is antiferromagnetic, and that increasing from the oxide to the telluride. As a result, EuO and EuS are ferromagnets, while EuSe is a metamagnet[3] and EuTe is an antiferromagnet.

The simple Heisenberg model (1) is, of course, unable to explain such properties of the magnetic semiconductors for which the conduction band plays a nonnegligible role. A striking example is the famous red shift of the optical absorption edge, appearing upon cooling below T_C. This effect has been observed for *all* ferromagnetic semiconductors so far, so that it must be regarded as a typical attribute of this class. The red shift is caused by a drastic shift of the lower conduction-band edge to smaller energies with decreasing temperature. Such effects are to be ascribed to a characteristic exchange interaction between the localized *f*-spin system (1) and the excited itinerant conduction electrons. The corresponding theoretical model is known in literature as an *s*-*f* model,[4,5] defined by the following Hamiltonian:

$$H = H_f + H_s + H_{sf} \,. \tag{2}$$

H_f is the same as in Eq. (1), while H_s is the operator of kinetic energy of the conduction electrons

$$H_s = \sum_{ij\sigma} T_{ij} a^\dagger_{i\sigma} a_{j\sigma} \tag{3}$$

$$= \sum_{\vec{k},\sigma} \epsilon(\vec{k}) a^\dagger_{\vec{k}\sigma} a_{\vec{k}\sigma} \,, \tag{4}$$

formulated in (3) in Wannier, and in (4) in Bloch representation, respectively. $a^\dagger, a$ are, as usual, electron creation and annihilation operators. The indices i, j refer to lattice sites, $\vec{k}$ to the electron wave vector, and $\sigma = \uparrow, \downarrow$ denotes the spin. The conduction electron interacts with the localized spin system by an intra-atomic exchange

$$H_{sf} = -g \sum_{i\sigma\sigma'} (\vec{\sigma} \cdot \vec{S}_i)_{\sigma\sigma'} a^\dagger_{i\sigma} a_{i\sigma'} \tag{5}$$

$$= -g \sum_i [S^z_i (a^\dagger_{i\uparrow} a_{i\uparrow} - a^\dagger_{i\downarrow} a_{i\downarrow}) + S^+_i a^\dagger_{i\downarrow} a_{i\uparrow} + S^-_i a^\dagger_{i\uparrow} a_{i\downarrow}] \,, \tag{6}$$

proportional to the intra-atomic *s*-*f* coupling constant g.

The many-body problem connected with the total Hamiltonian H in Eq. (2) could not exactly be solved up to now. Many of the proposed approximations[4,6-11] mainly aim at a reasonable explanation of the above-mentioned red-shift effect. This task has been enlarged to the more general question after the real temperature behavior of the whole conduction band. Because of s-f correlation (5), the conduction band will possibly split into several quasiparticle subbands. Their structure may exhibit strong dependence on temperature T as well as on typical model parameters, as degree of band filling n, coupling strength g, f-spin value S, lattice structure, and so on. The simplest picture has been brought about by first-order perturbation theory, or mean-field decouplings in Green-function techniques, respectively, showing a splitting of the conduction band into two spin-polarized subbands, one for $\uparrow$ and one for $\downarrow$ electrons.[4] The energetic distance between both subbands is predicted to be proportional to the product of coupling constant g times long-range magnetization $\langle S^z\rangle$ of the localized f-spin system. It is therefore temperature dependent and disappears in the paramagnetic region ($T > T_C$, where $\langle S^z\rangle \equiv 0$).

In a previous paper,[12] hereafter referred to as I, we have applied a spectral density approach (SDA) which severely questions that simple and widely accepted picture of the conduction band, outlined by perturbation theory. The main differences are as follows. (1) The splitting is more complicated and not due to the onset of ferromagnetism, but happens in the paramagnetic region, too (see Fig. 1 in I). (2) The various subbands consist of complicated mixed-spin states, generally preventing the electron spin from being totally polarized. (3) The temperature dependence manifests itself first of all in modification of the bandwidth. The famous red shift is caused by a broadening of the lowest $\uparrow$ quasiparticle subband with decreasing temperature. This last fact has been checked in I for the three ferromagnetic Eu compounds EuO, EuS, and EuSe, leading to almost quantitative fits of the experimental data (see Fig. 2 in I). Within the framework of the s-f model these three compounds are all characterized by the same lattice structure (Eu^{2+} ions occupy fcc sites), the same spin value $S=\frac{7}{2}$, and the same coupling constant g, which is a purely intra-atomic property of Eu^{2+} ions. Thus, the only different quantity for the three Eu compounds is the lattice constant d, or equivalently the width W of the "free" ($g=0$) Bloch band. In the authors' opinion the SDA, developed in I, is the only approach which is able to explain the strikingly different red shifts of EuO, EuS, and EuSe by use of the same set of parameters (g, S, fcc lattice structure). The differences are due to different values of W. Note that the only adjustable model parameter, the coupling constant g, is fixed by one of the three compounds, so that there is no further adjustable parameter for the other two.

The aim of this paper is twofold. First we want to continue and to complete our considerations in I. Strictly speaking, the quasiparticle energy spectrum, plotted as a function of temperature T in Fig. 1 of I, does not yet contain complete information about the structure of conduction band, as nothing is said about spectral weights and finite lifetimes of the various quasiparticles. These quantities can account for the fact that for certain combinations of the parameters, the one or the other subband may be unobservable because of too low spectral weights or too large dampings of the respective quasiparticles. A proper quantity, which can provide us with really complete information in this respect, is the quasiparticle density of states $\rho_\sigma(\omega)$ of the interacting s-f system, which will be discussed for the three cubic lattices in Sec. II.

The second aim concerns a further striking experiment performed with ferromagnetic semiconductors. Schoenes and Wachter have measured and reported[13] the red-shift values for Gd-doped EuO. Such a replacement of some divalent Eu^{2+} ions by trivalent Gd^{3+} ions leads to finite fillings of the conduction band. The experiment shows a steady decrease of the red shift with increasing carrier concentration. Less than 1% of free electrons per magnetic ion are already sufficient to reduce the red shift to half the value of the undoped material.[13] Here lies a clear challenge to the theory because for such small dopings the presumptions of the s-f model should work as well as for the undoped material. The carrier-concentration dependence of the red shift seems to indicate a *decreasing* ferromagnetic coupling with increasing number of charge carriers. It is especially surprising when compared to the huge increase of T_C with concentration,[13] which suggests thus on the contrary an *increasing* ferromagnetic coupling. These experimental data find, however, a direct explanation by the quasiparticle band structure outlined by the SDA, as will be shown in Sec. III. In Sec. IV we give our conclusions.

II. QUASIPARTICLE DENSITY OF STATES

We start with some general remarks about the spectral density approach (SDA) without presenting mathematical details, for which the reader may be referred to I. A basic function is the one-electron spectral density $A_{\vec{k}\sigma}(\omega)$,

$$A_{\vec{k}\sigma}(\omega)=\frac{1}{2\pi}\int_{-\infty}^{+\infty} d(t-t')e^{i\omega(t-t')}\langle[a_{\vec{k}\sigma}(t),\, a^{\dagger}_{\vec{k}\sigma}(t')]_{+}\rangle\,, \tag{7}$$

from which all one-electron equilibrium properties of the underlying s-f system may be derived.[14] If we neglect from the very beginning the Heisenberg exchange H_f equation (1), because of its negligible influence on the electronic quasiparticle spectrum of a ferromagnetic semiconductor (since $J_{ij} \ll g, W$), then we know the exact shape of $A_{\vec{k}\sigma}(\omega)$ for some important limiting cases.[15] In the atomic limit, as well as in the strongly coupled system ($gS \gg W$) at $T=0$, the spectral density $A_{\vec{k}\sigma}(\omega)$ consists of two (or one for $\sigma=\uparrow$ and $T=0$) δ-function peaks.[15] For moderate coupling constants and finite temperatures a broadening of the peaks occurs, according to finite lifetimes of respective quasiparticles. This broadening is properly taken into account by a Gaussian *Ansatz* for $A_{\vec{k}\sigma}(\omega)$ with two maxima, as introduced in I. Free parameters of this *Ansatz* are next fitted to exactly calculated spectral moments. It turns out, however, that $A_{\vec{k}\sigma}(\omega)$ cannot be determined self-consistently only by its own spectral moments. So we introduce two "higher" spectral densities,

$$B_{\vec{k}\sigma}(\omega)=\int_{-\infty}^{+\infty}\frac{d(t-t')}{2\pi}e^{i\omega(t-t')}\frac{1}{N}\sum_{i,j}e^{i\vec{k}\cdot(\vec{R}_i-\vec{R}_j)}\langle[S_i^z(t)a_{i\sigma}(t),\, a^{\dagger}_{j\sigma}(t')]_{+}\rangle\,, \tag{8}$$

$$C_{\vec{k}\uparrow(\downarrow)}(\omega)=\int_{-\infty}^{+\infty}\frac{d(t-t')}{2\pi}e^{i\omega(t-t')}\frac{1}{N}\sum_{i,j}e^{i\vec{k}\cdot(\vec{R}_i-\vec{R}_j)}\langle[S_i^{-(+)}(t)a_{i\downarrow(\uparrow)}(t),\, a^{\dagger}_{j\uparrow(\downarrow)}(t')]_{+}\rangle\,, \tag{9}$$

which must have the same quasiparticle structure as $A_{\vec{k}\sigma}(\omega)$ (concrete reasons are given in I). Consequently, analogous Gaussian *Ansätze* have to be chosen for these functions, too. By calculating a sufficient number of exact A, B, and C moments we finally get a closed system of equations for the free parameters in the *Ansätze* for $A_{\vec{k}\sigma}(\omega)$, $B_{\vec{k}\sigma}(\omega)$, and $C_{\vec{k}\sigma}(\omega)$, which can be solved self-consistently.

The results of this very simple, in principle, procedure (SDA) have been extensively discussed in I. Summarizing, we can say that the general behavior of the fundamental one-electron spectral density $A_{\vec{k}\sigma}(\omega)$ is essentially determined by the following five model parameters: (a) temperature T, (b) wave vector $\vec{k}$, (c) ratio of s-f coupling constant to Bloch bandwidth, g/W, (d) f-spin value S, and (e) lattice structure.

The spectral density $A_{\vec{k}\sigma}(\omega)$ itself is not directly observable. Much more important for the experiment is the density of states $\rho_\sigma(\omega)$, which is simply related to $A_{\vec{k}\sigma}(\omega)$ by a $\vec{k}$ summation over the first Brillouin zone:

$$\rho_\sigma(\omega)=\frac{1}{N}\sum_{\vec{k}}A_{\vec{k}\sigma}(\omega)\,. \tag{10}$$

To avoid confusion it should be stressed that $\rho_\sigma(\omega)$ is the density of states available for an excited electron with an original spin σ. That does not at all mean that this electron retains its spin after being excited into the conduction band.

In the SDA the $\vec{k}$ dependence of the spectral density is only indirect, namely due to the Bloch energies $\epsilon(\vec{k})$ (see I). That means that we can replace (10) by

$$\rho_\sigma(\omega)=\frac{1}{N}\sum_{\vec{k}}A_\sigma(\omega;\epsilon(\vec{k}))=\int_{-\infty}^{+\infty}d\epsilon\,\rho_0(\epsilon)A_\sigma(\omega;\epsilon)\,, \tag{11}$$

where

$$\rho_0(\epsilon)=\frac{1}{N}\sum_{\vec{k}}\delta(\epsilon-\epsilon(\vec{k})) \tag{12}$$

is the density of states of the noninteracting system, known for instance in the tight-binding approximation.[16]

Figures 1 and 2 show the density of states resulting from (11) for an fcc lattice, an f-spin value $S=\frac{7}{2}$, and a coupling constant $g=0.2$ eV. These parameters are all appropriate for the Eu chalcogenides. The ↑ and ↓ quasiparticle densities of states, plotted as functions of energy for various temperatures T, show both for $T\neq 0$ two slightly overlapping subbands. The degree of overlapping decreases with increasing g/W. The upper ↑ subband disappears in the ferromagnetic saturation ($T=0$) since then the localized f-spin system is completely aligned. An ↑ electron has therefore no possibility of spin exchange and moves as a quasifree particle through the lattice. The corresponding density of states is identical to $\rho_0(\omega)$, except for a constant shift of all band states by $-\frac{1}{2}gS$:

$$\rho_\uparrow(\omega;T=0)=\rho_0(\omega+\tfrac{1}{2}gS)\,. \tag{13}$$

With increasing temperature the upper ↑ subband enlarges at cost of the lower ↑ subband. In the ↓ spectrum the trend is quite the opposite. But here both subbands are present for all temperatures. Note that we have in the ferromagnetically

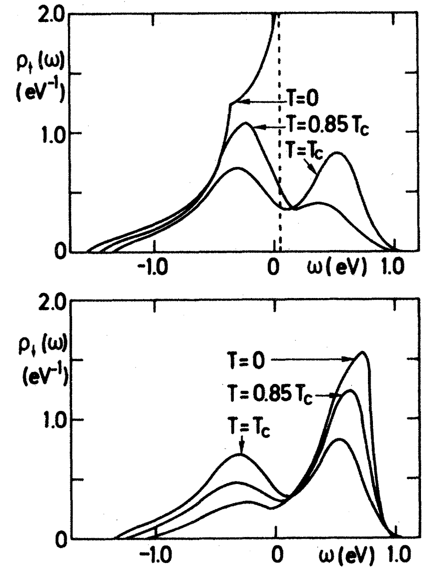

FIG. 1. Quasiparticle density of states $\rho_\sigma(\omega)$ as a function of energy for various temperatures T. The upper part shows $\rho_\uparrow(\omega)$, the lower part $\rho_\downarrow(\omega)$. Parameters: $g=0.2$ eV, $S=\frac{7}{2}$, $W=1.6$ eV, fcc lattice.

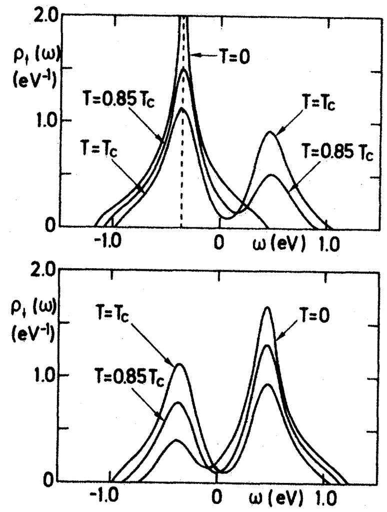

FIG. 3. The same as in Fig. 1, but for bcc lattice.

saturated system *three* subbands: one ↑ and two ↓ bands. Therefore, the SDA fulfills the exact result for $T=0$, derived in Ref. 15.

Practically the same temperature behavior of the quasiparticle density of states is observed for the two other cubic lattices (sc in Fig. 3, bcc in Fig. 4). It is an important fact that in all cases (Figs. 1–4) ↑ and ↓ spectra occupy almost the same energy. A distinct exchange splitting between ↑ and ↓ spectra, as expected from the perturbation theory,[4] does not happen. The observed red shift is due to the ↑ spectrum, while the ↓ spectrum is shifting blue upon cooling from $T=T_C$ to $T=0$. In this connection it is an interesting fact that the total red shift of the lower band edge between $T=T_C$ and $T=0$ is remarkably different for the three cubic lattices. Figures 1, 3, and 4, which present the density of states for the same parameters except for the lattice structure ($g=0.2$ eV, $S=\frac{7}{2}$, $W=1.6$ eV), yield the following red shift (s_r) values:

$$s_r(\text{fcc})=0.22\ \text{eV},\quad s_r(\text{bcc})=0.19\ \text{eV},\quad s_r(\text{sc})=0.13\ \text{eV}\,. \tag{14}$$

III. RED SHIFT OF DOPED FERROMAGNETIC SEMICONDUCTORS

We now discuss the striking carrier-concentration dependence of the red shift, as first observed

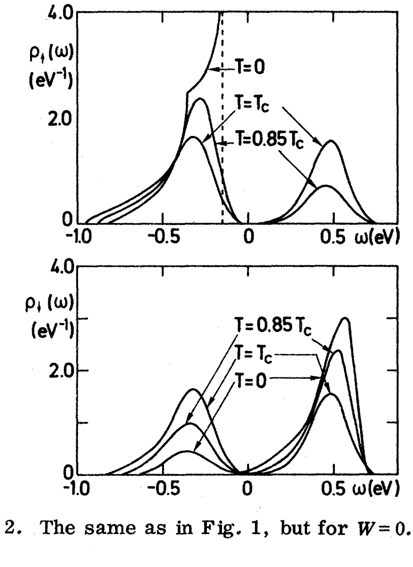

FIG. 2. The same as in Fig. 1, but for $W=0.8$ eV.

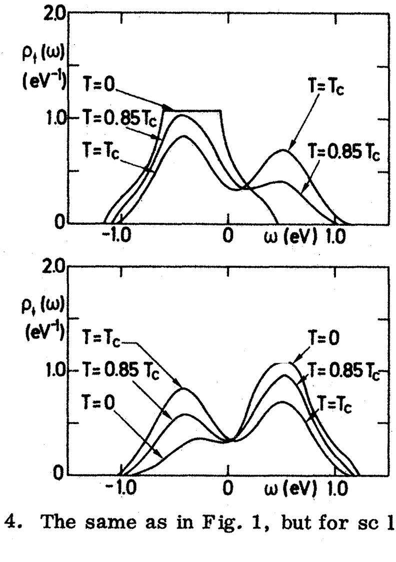

FIG. 4. The same as in Fig. 1, but for sc lattice.

by Schoenes and Wachter[13] for Gd-doped EuO. Gd is trivalent, so that in principle each Gd ion replacing a divalent Eu ion can give one electron to the conduction band. It is an interesting experimental observation that for weak dopings the number of conduction electrons is substantially smaller than the number of dopants, i.e., not all excess electrons are really completely delocalized. Thus the real concentration n of free charge carriers and not the Gd concentration x in $\mathrm{Eu}_{1-x}\mathrm{Gd}_x\mathrm{O}$ is important for what follows. n is the number of conduction electrons per magnetic ion. Table I, which has been taken from Ref. 13, shows that already for very small concentrations n of at most 2% the red shift (s_r) goes down to less than half the value of the undoped material. Note, that s_r denotes here the total shift of the optical absorption edge between room temperature and $T=0$.

Within the SDA this striking red-shift effect is a consequence of the specific temperature behavior of the various quasiparticle subbands. Figure 5 shows a schematic plot for the energy spectrum corresponding to the low-energy $\uparrow$ and $\downarrow$ subbands (see also Fig. 1 in I), which shall remind us once more that the red shift is a bandwidth effect.

Two mechanisms play an important role.

(1) The observation of the red shift needs an absorption experiment. For finite band fillings the excitation of the $4f$ electron occurs in such an experiment no longer onto the lower band edge, but onto the Fermi level μ, which is, of course, a function of the carrier concentration n. The quasiparticle spectrum in Fig. 5, however, shows that the temperature shift of the band states will be strongly $\vec{k}$ dependent. The states near the lower edge of the lower $\uparrow$ subband will shift red; the states near the upper edge, however, are shifting blue. In the middle of this subband there is a "neutral" state, which does not shift at all with temperature. That means, the higher the chemical potential μ, say, the larger the band filling n, the weaker the actual red shift of the optical absorption edge becomes.

TABLE I. Curie temperature T_C, number of free carriers $N_{\text{free carrier}}$, and red shift s_r of different $\mathrm{Eu}_{1-x}\mathrm{Gd}_x\mathrm{O}$ samples, after Schoenes and Wachter (Ref. 13). The last column gives the number n of free electrons per magnetic ion.

Sample	T_C (K)	$N_{\text{free carrier}}$ (10^{19} cm^{-3})	s_r (eV)	n (10^{-2})
0	69	<<0.1	0.260	0
1	80	3.8	0.236	0.127
2	80	6.6	0.224	0.223
3	83	16	0.188	0.532
4	115	60	0.124	2.016

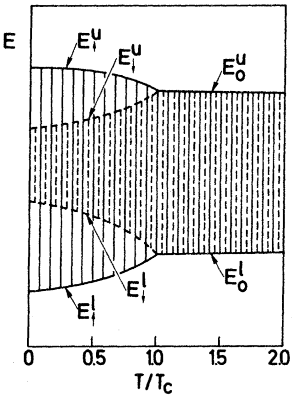

FIG. 5. Schematic plot of the lower $\uparrow$ and $\downarrow$ quasiparticle subbands (— for $\sigma=\uparrow$ --- for $\sigma=\downarrow$, l—lower edge, u—upper edge) as a function of reduced temperature T/T_C (according to Fig. 1 in Ref. 12).

(2) According to Fig. 5 and Table I, it becomes clear that for the considered small band occupations each state can be doubly occupied for $T>T_C$, but only singly occupied for $T=0$. That means an additional reduction of the red shift occurs because the Fermi edge μ is relatively deeper within the band for $T=0$ than for $T>T_C$.

Figure 6 shows the reduction of the red shift

$$\Delta s_r = s_r(n=0) - s_r(n) \tag{15}$$

as a function of band occupation n for various Bloch bandwidths W, calculated with the SDA. The other chosen parameters ($S=\frac{7}{2}$, $g=0.2$ eV, fcc lattice) are realistic for the Eu chalcogenides. As discussed in the Introduction (in detail in I), the only distinctive mark for the various ferromagnetic Eu chalcogenides is the Bloch bandwidth W. Since there is a one-to-one relation between and the red shift $s_r(n=0)$ of the undoped material (see Fig. 2 in I), we can derive W from the measured $s_r(n=0)$. Possibly because of experimental complications connected with an unambiguous definition of a real band edge, there have been reported in literature slightly different red-shift values for the undoped sample of EuO, which lead in our theory to different values of W. In Fig. 6 we have plotted curves for three different bandwidths, belonging to $s_r(n=0)=0.27$ eV (Ref. 2) (i.e., $W=2.35$ eV) and $s_r(n=0)=0.26$ eV (Ref. 13) (i.e., $W=2.09$ eV), both reported for EuO, and $s_r(n=0)=0.18$ eV (i.e., $W=0.9$ eV), appropriate for EuS.[2] The experimental data for the reduction of the red shift, given by Schoenes and Wachter[13] and Wachter[2] for Gd-doped EuO, also plotted in Fig. 6, are encouragingly well explained by the presented theory.

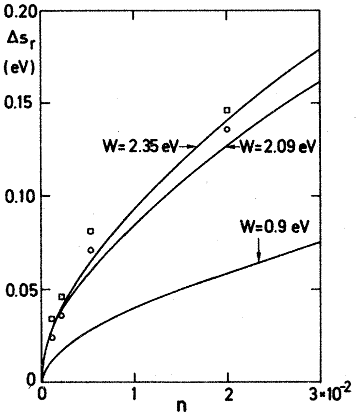

FIG. 6. Red shift reduction Δs_r, caused by doping, as a function of free-carrier concentration n for various Bloch bandwidths W. Inserted are experimental data reported by Schoenes and Wachter (Ref. 13) for Gd-doped EuO samples. Squares represent the experimental result if $s_r(n=0)=0.27$ eV (Ref. 2), circles if $s_r(n=0)=0.26$ eV (Ref. 13) (to be compared with the calculated s_r for $W=2.35$ and 2.09 eV, respectively). Parameters: $g=0.2$ eV, $S=\frac{7}{2}$, fcc lattice.

IV. CONCLUSIONS

The main point of our investigation, reported in this and the previous paper,[12] concerned the influence of the s-f exchange interaction between a conduction electron and the localized Heisenberg spin system on the electronic properties of a ferromagnetic semiconductor. We have especially been interested in finding a realistic picture of the true conduction-band structure and its dependencies on typical model parameters as temperature T, lattice structure, s-f coupling strength g, degree of band filling n, and f-spin value S. Perturbation theory and other previous approximations have led to the widespread assumption that the conduction band is split below T_C into two subbands, one for each spin direction, which should be shifted against one another by an amount of about coupling constant times magnetization. One of us has shown in an earlier paper[15] that this picture contradicts some exactly calculable limiting cases which, on the contrary, point to a much more complicated band splitting. The presented spectral density approach (SDA) fulfills all known limiting cases. It shows that the quasiparticle energy spectrum of a ferromagnetic semiconductor indeed consists of various subbands are generally built up by complicated mixed-spin states, preventing the electron spin from being totally polarized. The famous red shift for doped as well as undoped samples is almost quantitatively explained as a bandwidth effect of the lowest $\uparrow$ subband and not at all by an exchange splitting in the usual sense. The general band splitting is not due to the onset of ferromagnetism, but happens for temperatures $T > T_C$, too, where the $\uparrow$ and $\downarrow$ subbands coincide. We would like to stress that the proposed SDA can be tested by the above-mentioned characteristic experiments, giving satisfactory results and, furthermore, that it exactly reproduces all the known rigorously calculable limiting cases, which both strongly support the SDA in a general case, too.

In this paper and in I we have restricted our considerations to the *electronic* quasiparticle spectrum of a *ferro*magnetic semiconductor. The question how a partially filled conduction band influences the magnon spectrum of such materials has been beyond the scope of this work. Especially the carrier-concentration dependence of the Curie temperature T_C (Ref. 13) is still waiting for a quantitative explanation. Moreover, a generalization of the theory to antiferromagnetic materials (EuTe) could lead to further interesting results.

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