s-f model in magnetic semiconductors

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In the s-f model electrons in the conduction band are coupled with the lattice of localized magnetic moments by exchange interaction. In this paper the influence of the exchange interaction on electron bands and electrical resistivity is studied. An intermediate-coupling theory is presented, valid up to the region where the level broadening exceeds the average thermal energy but is small compared to the bandwidth. Using the functional-derivative method the spin correlations are expressed in terms of the connected correlation functions. The functional-derivative method also provides a decoupling recipe for the electron Green's functions. Concentrating on the two-spin correlations the single-particle Green's function is derived by a decoupling method and is shown to be equivalent to a perturbation expansion. The finite lifetime is obtained for all band energies. The absorption edge, derived from density of states, shows in ferromagnetic semiconductors the familiar red shift and in addition a blue shift of magnetic origin in the paramagnetic region. Mobility is derived from the two-particle Green's function calculated by a decoupling method. Level broadening affects both the acceleration and the scattering part, resulting generally in smaller mobilities than predicted by the weak-coupling theory. In addition, corrections to the ordinary acceleration term are obtained. Results for the ferromagnetic semiconductors EuS and EuO are presented. The narrow mobility minimum occurring at T_c in the weak-coupling case is considerably broadened in temperature. The minimum mobilities are around 3 cm²/Vsec in both materials. The low mobility near T_c is partly caused by the intense scattering and partly by the decrease in the acceleration term. The results compare reasonably well with experiments.

I. INTRODUCTION

The s-f model for magnetic semiconductors describes a case in which a relatively small number of charge carriers interact with the lattice of magnetic moments. The charge carriers, called s electrons, are assumed to occupy a broad conduction band. The magnetic moments, on the other hand, are formed by the localized electrons, say, f electrons. The Hamiltonian for the system is

$$H = \sum_{\nu} E_{\nu} a_{\nu}^{\dagger} a_{\nu} - \sum_{\nu,\nu',i} \vec{J}(\nu,\nu',i) \cdot \vec{S}_{i} a_{\nu}^{\dagger} a_{\nu'} + H_{M}.$$
 (1)

The first part represents conduction electrons. The index ν stands for the wave vector \mathbf{k} and the spin index σ of a band electron. E_{ν} is the electron band energy and a_{ν}^{\dagger} and a_{ν} are the electron creation and annihilation operators. The second term in the Hamiltonian is the s-f interaction part. It arises from the exchange coupling between the conduction electron spin $\mathbf{\tilde{s}}$ and the localized spin $\mathbf{\tilde{S}}_i$ at the lattice site $\mathbf{\tilde{R}}_i$. The quantity $\mathbf{\tilde{J}}$ is given by

$$\vec{J}(\nu,\nu',i) = \vec{J}(\vec{k}\sigma,\vec{k}'\sigma',i)
= J(\vec{k}'-\vec{k})e^{i(\vec{k}-\vec{k})\cdot\vec{R}}i2\langle\sigma|\vec{s}|\sigma'\rangle,$$
(2)

where $J(\vec{k}' - \vec{k})$ denotes the wave-vector-dependent exchange parameter. The term H_M in Eq. (1) is the usual Heisenberg Hamiltonian for the magnetic lattice.

The s-f interaction influences the optical, electrical transport, and magnetic properties of the material. A large number of papers have been published concerning these subjects. Excellent reviews have been given, e.g., by Methfessel and Mattis,¹ Haas,² Kasuya,³ Wachter,⁴ Leroux-Hugon,⁵ and Nagaev.⁶ The basic difficulty in the theory of the s-f interaction is associated with the magnetic system itself. Any higherorder perturbation treatment of the s-f interaction involves the dynamic many-spin correlation functions, which are not sufficiently understood. At low temperatures, below the magnetic-ordering temperature, the spin dynamics can be described by the spin waves. Several treatments of the influence of the electron-magnon interaction on electron bands⁷⁻⁹ and electrical resistivity¹⁰⁻¹² have been given. At intermediate temperatures, around the magnetic-ordering temperature, the spin-wave approximation is no longer valid. A simplification results from the fact that the magnetic excitation energies are in general small compared with the electron energies. Thus the spin lattice can be treated as a static system. The static many-spin correlation functions are easier to approximate, although in practice only the first few correlations can be taken into consideration. The first-order correction to electron energies is caused by the long-range magnetic order. In ferromagnets this splits the band into

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two spin-polarized subbands proposed by Vonsovskii and Izyumov.¹³ The second-order correction, treated by Haas¹⁴ and by Rys *et al.*,¹⁵ depends on the two-spin correlation function presenting the role of the short-range order. These first two correlations were sufficient to explain the large red shift of the absorption edge typical of ferromagnetic semiconductors.^{1-4,14,15} The influence of the two-spin correlations on the electrical resistivity was first analyzed by de Gennes and Friedel.¹⁶ The model was further developed by Fisher and Langer¹⁷ for metals and applied to semiconductors by Haas.¹⁴ The critical-temperature region in particular has been studied by Balberg et al.¹⁸ In ferromagnetic semiconductors especially the scattering by spin fluctuations becomes very intense near T_c , a phenomenon which suggests low mobility.¹⁴ This leads to the idea of carrier localization, i.e., to the theory of magnetic-polaron formation proposed by de Gennes¹⁹ and further discussed by several authors.^{6,20,21} At high temperatures the spins become almost uncorrelated. Then the s-f interaction is equivalent to scattering by random short-range potentials, and thus provides a link with the theory of disordered materials. For the s-f interaction treatments in a coherent-potential approximation (CPA) have been presented.^{22,23}

This paper deals with the s-f interaction at intermediate and high temperatures by means of the static-spin approximation. The paper is motivated by the observation that, for instance, in ferromagnets the many-spin correlation functions are divergent in the long-wavelength limit at T_{C} . Consequently the s-f interaction cannot be considered as a weak coupling in this temperature range. Furthermore, the mobilities limited by the spin-disorder scattering are calculated in Refs. 14-18 from the ordinary Boltzmann equation. In ferromagnetic semiconductors the mobilities are of the order of $10-100 \text{ cm}^2/\text{V}$ sec in the paramagnetic region and even smaller near T_{c} .¹⁴ This means that the level broadening Δ due to the finite lifetime is about 0.1-0.01 eV. However, the necessary condition for the validity of the Boltzmann equation is that $\Delta \ll k_B T$ in nondegenerate semiconductors.^{24,25} Thus the condition is violated in most parts of the relevant temperature range and a higher-order treatment of mobility is needed. This paper presents an intermediatecoupling theory for mobility extending to region Δ $>k_BT$ by means of Green's-function techniques. The higher-order spin correlations are expressed in terms of connected correlation functions. Then a class of terms depending on the two-spin correlations is treated to infinite order. One-particle Green's functions are considered in Sec. II. Mobility is calculated from the two-particle Green's function derived in Sec. III. As an application, results for the broad-band ferromagnetic semiconductors EuS and EuO are presented.

II. ONE-PARTICLE GREEN'S FUNCTION

The electronic properties are calculated by means of the Zubarev double-time Green's functions.^{26,27} Constructed from operators A and B, the retarted and advanced Green's functions are

$$G_{r,a}(t,t') = \langle \langle A(t); B(t') \rangle \rangle_{r,a}$$
$$= \pm \frac{1}{i\hbar} \Theta(\pm(t-t')) \langle [A(t), B(t')]_{\eta} \rangle.$$
(3)

Here Θ is the unit step function, the angular brackets $\langle \cdots \rangle$ denote the thermal average, and A(t) and B(t') are the operators in the Heisenberg picture. The quantity $\eta = +1$ or -1, denoting the respective commutator or anticommutator of the operators. The time-Fourier-transformed Green's functions $\langle\langle A; B \rangle\rangle_{\omega}$ satisfy the same equation of motion,

$$\hbar\omega\langle\langle A;B\rangle\rangle_{\omega} = \langle [A,B]_{\eta}\rangle + \langle\langle (AH - HA);B\rangle\rangle_{\omega}.$$
(4)

In the complex- ω plane the retarded Green's functions can be continued analytically into the upper and the advanced ones into the lower half plane. Hereafter the type of Green's functions is expressed by specifying the sign of the imaginary part of ω . When not needed, the subscript ω is suppressed.

Consider the one-electron Green's function $\langle \langle a_{\mu}; a_{\mu}^{\dagger} \rangle \rangle$, with $\eta = -1$. Using Eq. (4) and neglecting the magnetic part of the Hamiltonian as a small term, one derives the coupled equations

$$\begin{split} &\langle \hbar \omega - E_{\mu} \rangle \langle \langle a_{\mu}; a_{\mu}^{\dagger} \rangle \rangle \\ &= \delta_{\mu \mu'} - \sum_{\nu, i} \vec{J}(\mu, \nu, i) \cdot \langle \langle \vec{S}_{i} a_{\nu}; a_{\mu'}^{\dagger} \rangle \rangle ,\\ &\langle \hbar \omega - E_{\nu} \rangle \langle \langle \vec{S}_{i} a_{\nu}; a_{\mu'}^{\dagger} \rangle \rangle \\ &= \langle \vec{S}_{i} \rangle \delta_{\nu \mu'} - \sum_{\nu', j} \vec{J}(\nu, \nu', j) \cdot \langle \langle \vec{S}_{j} \vec{S}_{i} a_{\nu'}; a_{\mu'}^{\dagger} \rangle \rangle ,\\ &\langle \hbar \omega - E_{\nu'} \rangle \langle \langle \vec{S}_{j} \vec{S}_{i} a_{\nu'}; a_{\mu'}^{\dagger} \rangle \rangle \\ &= \langle \vec{S}_{j} \vec{S}_{i} \rangle \delta_{\nu' \mu'} - \sum_{\nu'', i} \vec{J}(\nu', \nu'', l) \cdot \langle \langle \vec{S}_{i} \vec{S}_{j} \vec{S}_{i} a_{\nu''}; a_{\mu'}^{\dagger} \rangle \rangle ,\\ &\langle \hbar \omega - E_{\nu''} \rangle \langle \langle \vec{S}_{i} \vec{S}_{j} \vec{S}_{i} a_{\nu''}; a_{\mu'}^{\dagger} \rangle \rangle \\ &= \langle \vec{S}_{i} \vec{S}_{j} \vec{S}_{i} \rangle \delta_{\nu'' \mu'} - \sum_{\nu'', i} \vec{J}(\nu'', \nu'', l) \cdot \langle \langle \vec{S}_{i} \vec{S}_{j} \vec{S}_{i} a_{\nu''}; a_{\mu'}^{\dagger} \rangle \rangle ,\\ &= \langle \vec{S}_{i} \vec{S}_{j} \vec{S}_{i} \rangle \delta_{\nu'' \mu'} \\ &- \sum_{\nu''', m} \vec{J}(\nu'', \nu'', m) \cdot \langle \langle \vec{S}_{m} \vec{S}_{i} \vec{S}_{j} \vec{S}_{i} a_{\nu'''}; a_{\mu'}^{\dagger} \rangle \rangle , \end{split}$$

etc. This system can be solved, for instance, as an power expansion in J or by using some decoupling method. In both cases a systematic way

is needed to classify the fluctuations. Here the functional-derivative method,²⁸ frequently used in theory of disordered materials, is chosen. Consider first its application to the magnetic system: By definition the thermal average of the component of a spin operator is

$$\langle S_l^{\alpha} \rangle = \mathrm{Tr}(e^{-H_M/k_B T} S_l^{\alpha}) / \mathrm{Tr}(e^{-H_M/k_B T}), \qquad (6)$$

where the Hamiltonian for the magnetic system is

$$H_{M} = -\sum_{i,j} I(\vec{\mathbf{R}}_{i} - \vec{\mathbf{R}}_{j}) \vec{\mathbf{S}}_{i} \cdot \vec{\mathbf{S}}_{j} + g\mu_{B} \sum_{i} \vec{\mathbf{S}}_{i} \cdot \vec{\mathbf{B}}_{i}.$$
 (7)

Here *I* is the Heisenberg exchange parameter and \vec{B}_i denotes the flux density of a magnetic field at the point \vec{R}_i . Considering \vec{B}_i as an independent variable, we can express different correlations as derivatives of $\langle S_i^{\alpha} \rangle$ with respect to B_i^{β} . These are called connected correlation functions and defined as

$$C^{(n+1)}S_1^{\alpha}, S_2^{\beta}, \dots, S_{n+1}^{\gamma}) = \left(-\frac{k_B T}{g\mu_B}\right)^n \frac{\partial^n \langle S_1^{\alpha} \rangle}{\partial B_{n+1}^{\gamma} \cdots \partial B_2^{\beta}}.$$
(8)

To simplify the calculation, the spins are treated classically in the following. The first few of the C functions are

$$C^{(1)}(S_{I}^{\alpha}) = \langle S_{I}^{\alpha} \rangle,$$

$$C^{(2)}(S_{I}^{\alpha}, S_{m}^{\beta}) = \langle \xi_{I}^{\alpha} \xi_{m}^{\beta} \rangle,$$

$$C^{(3)}(S_{I}^{\alpha}, S_{m}^{\beta}, S_{j}^{\gamma}) = \langle \xi_{I}^{\alpha} \xi_{m}^{\beta} \xi_{j}^{\gamma} \rangle,$$

$$C^{(4)}(S_{I}^{\alpha}, S_{m}^{\beta}, S_{j}^{\gamma}, S_{i}^{\delta})$$

$$= \langle \xi_{I}^{\alpha} \xi_{m}^{\beta} \xi_{j}^{\gamma} \xi_{i}^{\delta} \rangle - (\langle \xi_{I}^{\alpha} \xi_{m}^{\beta} \rangle \langle \xi_{j}^{\gamma} \xi_{i}^{\delta} \rangle + \text{perm.}),$$
(9)

etc. The quantity $\overline{\xi}_i$ is defined as

$$\vec{\xi}_i = \vec{S}_i - \langle \vec{S}_i \rangle , \qquad (10)$$

and the notation "perm." denotes the sum of all the terms obtained by permuting the spin indices but excluding the permutations within the thermal averages.

Equation (9) is suitable for a perturbation treatment of the problem. The functional-derivative method can also be applied to the Green's function itself to obtain a decoupling recipe: When H_M is neglected in the time-dependence of $a_{\mu}(t)$, the magnetic Hamiltonian is included in $\langle \langle a_{\mu}; a^{\dagger}_{\mu} \rangle \rangle$ only in the statistical weight operator analogous to Eq. (6). Again treating the spins classically, we can easily calculate the derivatives of $\langle \langle a_{\mu}; a^{\dagger}_{\mu} \rangle \rangle$ with respect to the field \vec{B}_i . In this way the set of Green's functions is obtained as a set of functional derivatives defined by

$$\langle \langle (S_1^{\alpha} S_2^{\beta} \cdots S_n^{\gamma})_c a_{\mu}; a_{\mu}^{\dagger} \rangle \rangle$$

$$= \left(-\frac{k_B T}{g \mu_B} \right)^n \frac{\partial^n \langle \langle a_{\mu}; a_{\mu}^{\dagger} \rangle \rangle}{\partial B_n^{\gamma} \cdots \partial B_2^{\beta} \partial B_1^{\alpha}} .$$

$$(11)$$

The first few of these are

$$\langle\langle\langle (S_i^{\alpha})_c a_{\mu}; a_{\mu'}^{\dagger} \rangle\rangle = \langle\langle \xi_i^{\alpha} a_{\mu}; a_{\mu'}^{\dagger} \rangle\rangle, \qquad (12a)$$

$$\begin{array}{l} \langle \langle (S_{j}^{\beta} S_{i}^{\alpha})_{c} a_{\mu}; a_{\mu}^{\dagger} \rangle \rangle \\ = \langle \langle \xi_{j}^{\beta} \xi_{i}^{\alpha} a_{\mu}; a_{\mu}^{\dagger} \rangle \rangle - \langle \xi_{j}^{\beta} \xi_{i}^{\alpha} \rangle \langle \langle a_{\mu}; a_{\mu}^{\dagger} \rangle \rangle, \end{array}$$
(12b)

$$\langle\langle\langle (S_I^{\gamma}S_J^{\beta}S_{i}^{\alpha})_{c}a_{\mu};a_{\mu}^{\dagger},\rangle\rangle$$

$$= \langle \langle \xi_{I}^{\gamma} \xi_{j}^{\beta} \xi_{i}^{\alpha} a_{\mu}; a_{\mu}^{\dagger} \rangle \rangle - \langle \xi_{I}^{\gamma} \xi_{j}^{\beta} \xi_{i}^{\alpha} \rangle \langle \langle a_{\mu}; a_{\mu}^{\dagger} \rangle \rangle \\ - [\langle \xi_{I}^{\gamma} \xi_{j}^{\beta} \rangle \langle \langle (S_{i}^{\alpha})_{c} a_{\mu}; a_{\mu}^{\dagger} \rangle \rangle + \text{perm.}], \qquad (12c)$$

$$\langle \langle \langle S_{m}^{6} S_{i}^{x} S_{j}^{\beta} S_{i}^{\alpha} \rangle_{c} a_{\mu}; a_{\mu}^{\dagger} \rangle \rangle$$

$$= \langle \langle \xi_{m}^{6} \xi_{i}^{x} \xi_{j}^{\beta} \xi_{i}^{\alpha} a_{\mu}; a_{\mu}^{\dagger} \rangle \rangle - \langle \xi_{m}^{\delta} \xi_{i}^{x} \xi_{j}^{\beta} \xi_{i}^{\alpha} \rangle \langle \langle a_{\mu}; a_{\mu}^{\dagger} \rangle \rangle$$

$$= [\langle \xi_{m}^{6} \xi_{i}^{x} \xi_{j}^{\beta} \rangle \langle \langle \langle S_{i}^{\alpha} \rangle_{c} a_{\mu}; a_{\mu}^{\dagger} \rangle \rangle + \text{perm.}]$$

$$= [\langle \xi_{m}^{6} \xi_{i}^{x} \rangle \langle \langle \langle S_{j}^{\beta} S_{i}^{\alpha} \rangle_{c} a_{\mu}; a_{\mu}^{\dagger} \rangle \rangle + \text{perm.}], \qquad (12d)$$

etc. The decoupling in the given order means that the functional derivative in that order is neglected.

Approximate C functions can be calculated by means, e.g., of the mean-field theory.²⁹ At the end of the calculation \vec{B} is replaced by the true magnetic field acting on the system. In the ferromagnetic case, considered here, let \vec{B} denote a constant field in the z direction. By the mean-field theory^{29,30} the two-spin correlation function for a crystal with fcc lattice and with nearest neighbor interactions only is

$$\langle \xi_{i}\xi_{j} \rangle = \frac{1}{N} \sum_{\vec{q}} e^{i\vec{q} \cdot (\vec{R}_{i} - \vec{R}_{j})} \vec{\Gamma}(\vec{q}) ,$$

$$\Gamma_{ij} = \frac{c_{0}}{\kappa_{i}^{2} + q^{2}a^{2}} \delta_{ij} , \quad i, j = x, y, z , \quad c_{0} = \frac{4S(S+1)T}{T_{c}} ,$$

$$\kappa_{z}^{2} = 12 \left(\frac{TB'_{S}(0)}{T_{c}B'_{S}(\vec{x})} - 1 \right) , \quad \kappa_{x}^{2} = \kappa_{y}^{2} = 12 \left(\frac{T\overline{x}B'_{S}(0)}{T_{c}B_{S}(\overline{x})} - 1 \right) .$$

$$(13)$$

Here N is the number of unit cells in the crystal, S the spin quantum number, B'_{S} the derivative of the Brillouin function, and a the lattice constant. Higher-order correlation functions can be calculated by the same method,³⁰ but are not needed here.

In the equation of motion the thermal average part of spins is conveniently taken into account by introducing the new spin-polarized bands defined by

$$E_{\nu} = E_{\vec{k}\sigma} = \hbar^2 k^2 / 2m^* + \Delta E_{\sigma} ,$$

$$\Delta E_{\sigma} = -J(0)N\langle S \rangle (\delta_{\sigma, \dagger} - \delta_{\sigma, \dagger}) ,$$
(14)

where m^* denotes the effective mass, $\langle S \rangle$ the

thermal average of S_i^z , and \dagger and \dagger the two different spin orientations. From now on the interaction part contains the spin operators $\xi_i = \vec{S}_i - \langle \vec{S}_i \rangle$. The lowest-order Green's function is obtained when the decoupling is made in Eq. (12a):

$$G_{\mu}^{(0)} = \langle \langle a_{\mu}; a_{\mu'}^{\dagger} \rangle \rangle^{(0)} = \delta_{\mu \mu'} / (\hbar \omega - E_{\mu}).$$
⁽¹⁵⁾

This is the free-electron propagator associated with the spin-polarized bands. Performing the decoupling in Eq. (12b), one gets

$$\langle\langle a_{\mu}; a_{\mu'}^{\dagger} \rangle\rangle^{(1)} = \delta_{\mu \mu'} / \left(\hbar \omega - E_{\mu} - \sum_{\nu, i, j} \frac{\mathbf{\tilde{J}}(\mu, \nu, i) \cdot \langle \mathbf{\tilde{\xi}}_{i} \mathbf{\tilde{\xi}}_{j} \rangle \cdot \mathbf{\tilde{J}}(\nu, \mu, j)}{\hbar \omega - E_{\nu}} \right).$$
(16)

When continuing to higher orders mathematical complications arise. However, concentrating only on the two-spin correlations and on the terms diagonal in the electron indices, one obtains a solution similar to Eq. (16), but the denominator $\hbar \omega - E_{\nu}$ becomes also corrected to second order. When continuing in the same way the solution becomes a continued fraction. Thus the Green's function is finally

$$G_{\mu} = \langle \langle a_{\mu}; a_{\mu}^{\dagger} \rangle \rangle = \delta_{\mu \mu'} / \left(\hbar \omega - E_{\mu} - \sum_{\nu, i, j} \mathbf{J}(\mu, \nu, i) \cdot \langle \mathbf{\xi}_{i} \mathbf{\xi}_{j} \rangle \cdot \mathbf{J}(\nu, \mu, j) \langle \langle a_{\nu}; a_{\nu}^{\dagger} \rangle \rangle \right).$$
(17)

It is interesting to compare this solution with the perturbation expansion in terms of the *C* functions obtained by using Eqs. (5) and (9). The expansion is visualized up to fourth order in Fig. 1. The double horizontal line presents the true Green's function and the single line the zero-order function given in Eq. (15). The dashed lines represents the *s*-*f* interaction. With each cross from which *n* interaction lines are emanating there is associated a factor $C^{(n)}$. When the two-spin correlations only are considered, a solution is obtained by summing the diagrams of types *a*, *d*, and *f* to infinite order. This is done by the self-energy insertion

$$\Sigma_{\mu} = \sum_{\nu, i, j} \overline{\mathbf{j}}(\mu, \nu, i) \cdot \langle \overline{\xi}_i \overline{\xi}_j \rangle \cdot \overline{\mathbf{j}}(\nu, \mu, j) \langle \langle a_{\nu}; a_{\nu}^{\dagger} \rangle \rangle, \quad (18)$$

which gives a solution equivalent to Eq. (17).

After the lattice sums in Eq. (18) have been calculated by means of Eq. (13), an integration over the wave-vector space is left. The integral is relatively slowly converging. Thus it is necessary to ensure the convergence by the fact that $J(\vec{q})$, the Fourier transform of the exchange parameter, must decrease rapidly beyond the I Brillouin zone. The \vec{q} dependence of J is not known. Here a simple form

$$J(\bar{q}) = J/(1+q^2l^2)$$
(19)

is adopted. The parameter l describes the extend of the exchange interaction in real space. Thus lis of the order of the radius of the magnetic shell. Assuming then that in the integral the self-energy is independent of the wave vector, we can perform the integration. The result is

$$\Sigma_{\vec{k}\sigma} = -\frac{J^2 N^2}{16\pi E_0} \frac{1}{ak} \sum_{\sigma'} \frac{c(\sigma, \sigma')}{[1 + (l\Omega_{\sigma'}/a)^2]^2} \left(\arctan \frac{ak}{\kappa(\sigma, \sigma') - i\Omega_{\sigma'}} - \arctan \frac{ak}{\kappa(\sigma, \sigma') + a/l} - \frac{ak}{2} \frac{(a/l)[1 + (l\Omega_{\sigma'}/a)^2]}{[\kappa(\sigma, \sigma') + a/l]^2 + a^2k^2} \right),$$

$$(20)$$



FIG. 1. Diagrams occurring in the expansion of the one-particle Green's function.

where

$$\Omega_{\sigma'} = (\hbar \omega - \Delta E_{\sigma'} - \Sigma_{\sigma'})^{1/2} E_0^{-1/2}, \quad E_0 = \hbar^2 / 2m^* a^2,$$

and

$$c(\sigma, \sigma') = \begin{cases} c_0, & \sigma = \sigma' \\ 2c_0, & \sigma \neq \sigma' \end{cases}, \qquad \kappa(\sigma, \sigma') = \begin{cases} \kappa_z, & \sigma = \sigma' \\ \kappa_x, & \sigma \neq \sigma' \end{cases}.$$

Here $\Sigma_{\sigma'}$ denotes the wave-vector-independent selfenergy. The expression can be further simplified. In semiconductors the states close to the bottom of the band are the most important ones. For these states $|(l/a)\Omega| \ll 1$, in general. Furthermore, the self-energy can be assumed to be independent of spin. Then finally a third-order equation is obtained for the spin- and wave-vector-independent Σ . Typical results for EuS are shown in Fig. 2 for the self-energy and the density of states defined by

$$D_{\sigma}(\omega) = \frac{i}{2\pi V} \sum_{\vec{k}} \left[G_{\vec{k}\sigma}^{\pm}(\omega) - G_{\vec{k}\sigma}^{\pm}(\omega) \right] , \qquad (21)$$

where

$$G_{\mathbf{k}\sigma}^{\pm}(\omega) = G_{\mathbf{k}\sigma}(\omega \pm i\epsilon)$$

Here ϵ is a small positive number and V denotes the volume of the crystal. Using the k-independent self-energy, we find for the density of states



FIG. 2. (a) Self-energy Σ and (b) the density of states per spin, D, for EuS in the paramagnetic region at the temperature $T=1.025T_C$. The parameter values used in the calculation are $S=\frac{7}{2}$, a=5.96 Å, $m^*=m_0$, T_C =16 K, and l=0.3 Å. The exchange parameter J (0) is chosen such that the maximum band-splitting energy $\Delta E=2J(0)NS=0.66$ eV.

$$D_{\sigma}(\omega) = \frac{1}{2\pi^2 a^3 (2E_0)^{3/2}} \frac{\Delta}{\left[-\Lambda + (\Lambda^2 + \Delta^2)^{1/2}\right]^{1/2}}, \quad (22)$$

where

 $\Lambda = \hbar \omega - \Delta E_{\sigma} - \operatorname{Re}\Sigma(\omega), \quad \Delta = |\operatorname{Im}\Sigma(\omega)|.$

As shown in Fig. 2(a) the self-energy is a slowly varying function of ω . Thus the approximate band energies, determined as zeros of the real part of . the Green's-function denominator, have roughly speaking the same k dispersion as originally. However, the new band energies are all well damped because of the nonvanishing imaginary part of Σ . The density of states is similar to the unperturbed curve, but is shifted downwards in energy. Figure 3 shows the absorption edge as a function of temperature in EuS and EuO. The absorption edge is defined as the edge of the density-of-states curve. A large red shift of the edge, typical of ferromagnetic semiconductors,¹⁻⁴ is seen. In the ferromagnetic phase the edge is determined by the long-range order, but near and above T_c the corrections arising from the shortrange order dominate. The transition region appears as a kink in the edge below T_{C} . The kink becomes suppressed by the applied magnetic field, which possibly explains why it is not reported experimentally. Haas¹⁴ calculated the corrections to energy bands coming from spin fluctuations up to second order using the ordinary perturbation theory. The results were good except at T_c , where the second-order correction was divergent in the $\vec{k} \rightarrow 0$ limit. Rys *et al.*¹⁵ calculated the band edge from a Green's function, which was essentially the same as in Eq. (16). Their result was



FIG. 3. Absorption edge in EuS and EuO. For EuO the values a = 5.15 Å and $T_C = 69$ K are used, the other parameters being the same as for EuS.

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in good agreement with experiments, and was not divergent anymore. Our result in Fig. 3 explains also properly the red shift. In addition, as a new property, it shows the blue shift of the edge in the paramagnetic region in EuS. The magnitude of the blue shift compares favorably with the experimental value 2.8×10^{-4} eV/K for EuS.³¹ With the choice of the parameters used in this calculation the magnetic blue shift in EuO occurs at temperatures above 300 K. However, the temperature dependence of the absorption edge is generally more complicated. In ordinary semiconductors the shift is caused by lattice dilatation and by electron-phonon coupling.³² Recently the blue shift in EuSe was explained by Batlogg and Wachter³³ entirely by means of the lattice mechanisms. Our calculation shows clearly that the magnetic part of the blue shift cannot be disregarded.

With the same approximations as used above in the case of the one-electron Green's function, the solutions for the higher-order functions are

$$\langle \langle \langle \mathbf{S}_{i} \rangle_{c} a_{\nu}; a_{\mu}^{\dagger} \rangle \rangle$$

$$= \sum_{i} G_{\nu} (-\mathbf{J}(\nu, \mu, l) \cdot \langle \mathbf{\xi}_{i} \mathbf{\xi}_{i} \rangle) G_{\mu},$$

$$\langle \langle \langle \mathbf{S}_{j} \mathbf{S}_{i} \rangle_{c} a_{\nu}; a_{\mu}^{\dagger} \rangle \rangle$$

$$= \sum_{\nu', t', l} [G_{\nu} (-\mathbf{J}(\nu, \nu', l') \cdot \langle \mathbf{\xi}_{i'} \mathbf{\xi}_{j} \rangle) \times G_{\nu} (-\mathbf{J}(\nu', \mu, l) \cdot \langle \mathbf{\xi}_{i} \mathbf{\xi}_{i} \rangle) G_{\mu} + (ij)],$$
(23)

etc. Here (ij) means the permutation of i and j.

III. ELECTRICAL CONDUCTIVITY

In an electric field $E_{\alpha}e^{-i\omega t}$ applied in the direction α the electrical current density in the direction β is

$$V_{\beta} = \sigma_{\beta\alpha}(\omega) e^{-i\omega t} E_{\alpha} , \qquad (24)$$

where the conductivity is given by^{26,27}

$$\sigma_{\beta\alpha}(\omega) = -(e^2/V) \langle \langle v_{\beta}; r_{\alpha} \rangle \rangle_{\omega + i\epsilon}, \quad \eta = +1.$$
 (25)

In this equation e denotes the unit charge and v_{β} and r_{α} are the velocity and position operators

$$v_{\beta} = \sum_{\vec{k}\sigma, \vec{k}'\sigma} \frac{\hbar k_{\beta}}{m^{*}} \, \delta_{\vec{k},\vec{k}'} a^{\dagger}_{\vec{k}\sigma} a_{\vec{k}'\sigma} ,$$

$$r_{\alpha} = \sum_{\vec{k}\sigma, \vec{k}'\sigma} \left(\frac{\partial}{i\partial k'_{\alpha}} \langle \vec{k} | \vec{k}' \rangle \right) a^{\dagger}_{\vec{k}\sigma} a_{\vec{k}'\sigma} .$$
(26)

The conductivity depends on the retarted twoparticle Green's function of the type $\langle\langle a_{\nu}^{\dagger}a_{\nu'}; r_{\alpha}\rangle\rangle$. This is calculated by the same method as the oneparticle function in Sec. II with repeated use of the equation of motion. The decoupling principle given in Eqs. (11)-(12) can also be applied in the present case. With the same approximations as in the one-particle case an equation for $\langle\langle a_{\nu}^{\dagger}a_{\nu'}; r_{\alpha}\rangle\rangle$ is obtained:

$$(\hbar\omega - E_{\nu'} + E_{\nu})\langle\langle a_{\nu}^{\dagger}a_{\nu'}; r_{\alpha}\rangle\rangle = T(\nu, \nu') + \sum_{\substack{\nu'', \nu''', i \\ \mu'', \mu''', j}} \tilde{J}(\nu'', \nu''', i) \cdot \langle \tilde{\xi}_{i}\tilde{\xi}_{j}\rangle \cdot \tilde{J}(\mu'', \mu''', j)\langle\langle [\{a_{\nu}^{\dagger}a_{\nu'}, a_{\nu''}^{\dagger}a_{\nu'''}], a_{\mu''}^{\dagger}a_{\mu'''}]; r_{\alpha}\rangle\rangle.$$

$$(27)$$

The commutator with curly brackets means

$$\{a_{\nu}^{\dagger}a_{\nu'}, a_{\nu''}^{\dagger}a_{\nu'''}\} = a_{\nu}^{\dagger}a_{\nu'''}\delta_{\nu'\nu''}F(\omega, \nu, \nu'\, \nu') - a_{\nu''}^{\dagger}a_{\nu'}\delta_{\nu\nu'''}F(\omega, \nu\, \nu'\, \nu'), \qquad (28)$$

where the function F is to be derived from the equation

$$-\sum_{\mu,i,j} \left[\overline{\mathbf{J}}(\nu',\mu,i) \cdot \langle \overline{\mathbf{\xi}}_i \overline{\mathbf{\xi}}_j \rangle \cdot \overline{\mathbf{J}}(\mu,\nu',j) F(\omega,\nu,\mu) + \overline{\mathbf{J}}(\mu,\nu,i) \cdot \langle \overline{\mathbf{\xi}}_i \overline{\mathbf{\xi}}_j \rangle \cdot \overline{\mathbf{J}}(\nu,\mu,j) F(\omega,\mu,\nu') \right].$$
(29)

The quantity T is given by the expression

 $F^{-1}(\omega,\nu,\nu') = \hbar\omega + E_{\nu} - E_{\nu'}$

$$T(\nu, \nu') = \langle [a_{\nu}^{\dagger}a_{\nu'}, r_{\alpha}] \rangle + \langle [\{a_{\nu}^{\dagger}a_{\nu'}, H'\}, r_{\alpha}] \rangle$$
$$+ \langle [\{\{a_{\nu}^{\dagger}a_{\nu'}, H'\}, H'\}, r_{\alpha}] \rangle + \cdots .$$
(30)

Here H' denotes the s-f interaction part, modified in the following way: After the curly-bracket commutators have been calculated, the spin operators are replaced by the same combination that occurs in the functional derivative of that order. This takes care of the decoupled terms forming the last part in Eq. (27).

Formally Eq. (27) is a transport equation: The term on the left-hand side represents the free evolution of the system, T arises from the acceleration by a (unit) electric field, and the last term represents scattering by spin fluctuations. When the lowest-order terms only are considered, i.e., when T is replaced by the first term.

$$T^{(0)}(\nu,\nu') = T^{(0)}(\vec{\mathbf{k}}\sigma,\vec{\mathbf{k}}'\sigma') = \left(-\frac{\partial n_{\vec{\mathbf{k}}\sigma}}{i\partial k_{\alpha}}\right)\delta_{\vec{\mathbf{k}}\sigma,\vec{\mathbf{k}}'\sigma'},$$
(31)

and F is similarly approximated by

$$F^{(0)}(\omega, \nu, \nu') = (\hbar \omega + E_{\nu} - E_{\nu'})^{-1}, \qquad (32)$$

it is seen that Eq. (27) becomes formally equivalent to the Boltzmann equation for the case $\nu = \nu'$. In the following, Eq. (27) is treated more carefully in order to obtain a proper intermediatecoupling result.

Consider the solution of Eq. (29). The role of the function F is to produce in the weak-coupling limit the energy-conserving δ functions in the scattering part. In the higher-order calculation the δ functions become broadened. An approximation to F is obtained by introducing a finite damping term into Eq. (32), i.e., by inserting

$$F(\omega, \nu, \nu') = (\hbar \omega + E_{\nu} - E_{\nu}, + i \Delta_{\nu\nu})^{-1}$$

Substituting this into Eq. (29) and assuming that $\Delta_{\nu\nu}$, is slowly varying in its indices, one can carry out the integrations and obtain an equation for $\Delta_{\nu\nu}$, which can be solved by iteration. There is also another solution, which relates F to the Green's functions: Consider the quantity

$$I(\omega, \nu, \nu') = \frac{-i\hbar}{2\pi} \int_{-\infty}^{\infty} d\omega' G_{\nu}(\omega') G_{\nu}^{+}(\omega'+\omega) , \qquad (33)$$

where $G^{\pm}_{\nu}(\omega') = G_{\nu}(\omega' \pm i\varepsilon)$. Substituting the Green's functions from Eq. (17) and using the sum rule

$$\frac{-i\hbar}{2\pi}\int_{-\infty}^{\infty}d\omega' [G_{\nu}^{-}(\omega') - G_{\nu}^{+}(\omega')] = 1 ,$$

one obtains

$$I(\omega, \nu, \nu') = \frac{1}{\hbar\omega + E_{\nu} - E_{\nu'}} \left(1 - \frac{i\hbar}{2\pi} \sum_{\nu'', i, j} \int_{-\infty}^{\infty} d\omega' [\mathbf{\bar{J}}(\nu', \nu'', i) \cdot \langle \mathbf{\bar{\xi}}_{i} \mathbf{\bar{\xi}}_{j} \rangle \cdot \mathbf{\bar{J}}(\nu'', \nu', j) G_{\nu''}^{+}(\omega' + \omega) - \mathbf{\bar{J}}(\nu, \nu'', i) \cdot \langle \mathbf{\bar{\xi}}_{i} \mathbf{\bar{\xi}}_{j} \rangle \cdot \mathbf{\bar{J}}(\nu'', \nu, j) G_{\nu''}^{-}(\omega')] G_{\nu}^{-}(\omega') G_{\nu'}^{+}(\omega' + \omega) \right).$$
(34)

The first part of the ω' integration can be written

$$\int_{-\infty}^{\infty} d\omega' G_{\nu}^{+}(\omega'+\omega)G_{\nu}^{-}(\omega')G_{\nu}^{+}(\omega'+\omega)$$
$$=\int_{-\infty}^{\infty} d\omega' G_{\nu}^{+}(\omega'+\omega)G_{\nu}^{+}(\omega'+\omega) \left[G_{\nu}^{-}(\omega') - G_{\nu}^{+}(\omega')\right].$$

This follows from the analytical properties of the Green's functions: the part coming from $G^+G^+G^+$ is zero, since the integration can be performed over a contour in the upper half plane. Then in the integral the approximation

$$\begin{aligned} \left[G_{\nu}^{*}(\omega') - G_{\nu}^{*}(\omega')\right] \left[G_{\nu}^{*}(\omega'') - G_{\nu}^{*}(\omega'')\right] \\ &\simeq \left(2\pi i/\hbar\right)\delta(\omega'' - \omega') \left[G_{\nu}^{*}(\omega'') - G_{\nu}^{*}(\omega'')\right] \end{aligned} \tag{35}$$

is made. This is allowed if the spectral density $G^- - G^+$ shows a peaked structure. Then the integral becomes

$$\int_{-\infty}^{\infty} d\omega' G_{\nu''}^{*}(\omega'+\omega)G_{\nu}^{-}(\omega')G_{\nu'}^{*}(\omega'+\omega)$$

$$\simeq \frac{-i\hbar}{2\pi} \int_{-\infty}^{\infty} d\omega' G_{\nu''}^{*}(\omega'+\omega)G_{\nu}^{-}(\omega')$$

$$\times \int_{-\infty}^{\infty} d\omega'' G_{\nu'}^{*}(\omega''+\omega)G_{\nu}^{-}(\omega''). \quad (36)$$

Similarly one can treat the second integral in Eq. (34) involving the product of three Green's functions. As a result one obtains for $I(\omega, \nu, \nu')$ an equation which turns out to be equivalent to Eq. (29) for *F*. Hence

$$F(\omega,\nu,\nu') \simeq \frac{-i\hbar}{2\pi} \int_{-\infty}^{\infty} d\omega' G_{\nu}(\omega') G_{\nu}^{*}(\omega'+\omega) . \quad (37)$$

When the scattering part in Eq. (27) is written out for the case $\nu = \nu'$, the familiar terms representing the forward and backward scattering are recognizable. By neglecting the latter contribution, one obtains the solution

$$\langle \langle a_{\nu}^{\dagger} a_{\nu}^{}; r_{\alpha} \rangle \rangle = F(\omega, \nu, \nu) T(\nu, \nu) .$$
(38)

Introducing a complex, frequency-dependent relaxation time τ_{ν} defined by

$$\langle\langle a_{\nu}^{\dagger}a_{\nu};r_{\alpha}\rangle\rangle = (1/i\hbar)[\tau_{\nu}/(1-i\omega\tau_{\nu})]T(\nu,\nu), \quad (39)$$

we see that

$$1/\tau_{\nu} = (1/i\,\hbar) [F^{-1}(\omega,\nu,\nu) - \hbar\omega].$$
(40)

The use of Eqs. (37), (29), and (18) then gives

$$\frac{1}{\tau_{\nu}} = \frac{1}{2\pi} \int_{-\infty}^{\infty} d\omega' \left[\Sigma_{\nu}^{*}(\omega' + \omega) G_{\nu}^{-}(\omega') + \Sigma_{\nu}^{-}(\omega') G_{\nu}^{*}(\omega' + \omega) \right], \qquad (41)$$

where $\Sigma^{\pm}(\omega) = \Sigma(\omega \pm i\varepsilon)$. By applying the spectral representations for the Green's functions^{26, 27}

$$G^{\pm}(\omega) = \frac{1}{\pi} \int_{-\infty}^{\infty} d\omega' \frac{\operatorname{Im} G^{+}(\omega')}{\omega' - \omega \mp i\varepsilon}$$
$$= \pm \frac{1}{2\pi i} \int_{-\infty}^{\infty} d\omega' \frac{G^{\pm}(\omega')}{\omega' - \omega \mp i\varepsilon}$$
(42)

and analogous representations for the self-energy,

Eq. (41) becomes

$$\frac{1}{\tau_{\nu}} = \frac{i}{\pi} \int_{-\infty}^{\infty} d\omega' [G_{\nu}^{+}(\omega' + \omega) - G_{\nu}^{-}(\omega' - \omega)] |\operatorname{Im}\Sigma_{\nu}(\omega')| .$$
(43)

When it is again assumed that the spectral density shows a peaked structure, the relaxation time in the dc case becomes

$$\frac{1}{\tau_{\nu}^{(0)}} = 2\Delta_{\nu}/\hbar, \quad \Delta_{\nu} = \left| \operatorname{Im}\Sigma_{\nu}(\vec{\omega}) \right|.$$
(44)

Here $\hbar \overline{\omega}$ means the new band energy, i.e., the energy at which $\hbar \overline{\omega} - E_{\nu} - \operatorname{Re} \Sigma_{\nu}(\overline{\omega}) = 0$. Equation (44) expresses the known relation between the momentum relaxation time and the level broadening, originally derived in the weak scattering limit.³⁴ As seen above, the result is more general. When Eq. (44) was derived, the assumption of a peaked spectral-density structure has been utilized. At first sight this seems to imply weak interaction, but in fact it can be replaced by an assumption of a slowly varying $\Sigma(\omega)$, which is valid in the present case.

When the zero-order acceleration term given in Eq. (31) is used, the dc conductivity becomes

$$\sigma_{\lambda}^{(0)} = \frac{-e^2}{Vm^*} \sum_{\vec{k}} k_{\alpha} \tau_{\vec{k}\lambda}^{(0)} \frac{\partial n_{\vec{k}\lambda}}{\partial k_{\alpha}} , \qquad (45)$$

where λ denotes the spin index. This is formally the same as the result of the Boltzmann transport equation, but includes the effect of level broadening both in the relaxation time and in the momentum distribution. Thus the validity of Eq. (45) is not restricted by the condition $\Delta \ll k_B T$ as in the case of the Boltzmann equation. There are two consequences of the level broadening in Eq. (45): (i) The momentum distribution defined by^{26,27}

$$n_{\vec{k}\sigma} = \langle a^{\dagger}_{\vec{k}\sigma} a_{\vec{k}\sigma} \rangle$$
$$= \frac{i\hbar}{2\pi} \int_{-\infty}^{\infty} d\omega [G^{*}_{\vec{k}\sigma}(\omega) - G^{-}_{\vec{k}\sigma}(\omega)] f(\omega) , \qquad (46)$$

where

$$f(\omega) = (e^{(\hbar\omega - E_F)/k_BT} + 1)^{-1}$$

and E_F denotes the Fermi energy, differs con-

siderably from the Fermi-Dirac distribution: In the region $\Delta \geq k_B T$, $n_{\vec{k}\sigma}$ is a slowly decreasing function of \vec{k} compared to the exponential decrease in the $\Delta \rightarrow 0$ limit in the nondegenerate case. Thus $(1/n_{\vec{k}\sigma})(\partial n_{\vec{k}\sigma}/\partial k_{\alpha})$, and consequently the mobility, decreases. In the limit $\Delta \gg k_B T$ the mobility decreases, as an order-of-magnitude estimate, by a factor $k_B T/\Delta$ from the ordinary result.

(ii) The relaxation time $\tau_{k\sigma}^{(0)}$ given by Eq. (44) is usually smaller than the result obtained by the lowest-order theory. Thus the mobility decreases further relative to the ordinary result.

Next, consider the corrections to the acceleration term. Using Eq. (23), we find the second term in the expansion to be

$$T^{(1)}(\nu, \nu') = \langle [\{a^{\dagger}_{\nu}a_{\nu}, H'\}, r_{\alpha}] \rangle$$

= $\delta_{\nu\nu}, \sum_{\mu'} W(\nu, \mu') [F(\omega, \nu, \mu')$
 $-F(\omega, \mu', \nu)] D^{\alpha}_{\nu\mu'} (G_{\nu}G_{\mu'}),$ (47)

where

$$W(\nu, \mu') = W(\vec{\mathbf{k}}\sigma, \vec{\mathbf{k}}'\sigma') = NJ^{2}(\vec{\mathbf{k}} - \vec{\mathbf{k}}') 2\langle \sigma | \vec{\mathbf{s}} | \sigma' \rangle$$
$$\cdot \vec{\Gamma}(\vec{\mathbf{k}} - \vec{\mathbf{k}}') \cdot \langle \sigma' | \vec{\mathbf{s}} | \sigma \rangle 2$$

(48)

and D denotes in general

$$D^{\alpha}_{\vec{k}_{1}\sigma_{1}\cdots\vec{k}_{n}\sigma_{n}}(G_{\vec{k}_{1}\sigma_{1}}\cdots G_{\vec{k}_{n}\sigma_{n}})$$

$$= -\frac{\hbar}{2\pi} \left(\frac{\partial}{\partial k_{\alpha 1}} + \cdots + \frac{\partial}{\partial k_{\alpha n}}\right)$$

$$\times \int_{-\infty}^{\infty} d\omega' [G^{+}_{\vec{k}_{1}\sigma_{1}}(\omega')\cdots G^{+}_{\vec{k}_{n}\sigma_{n}}(\omega') - \text{c.c.}]f(\omega').$$
(49)

In higher orders the calculation becomes tedious. In order to pick up the large terms in the expansion the following hint is used: In the case of scattering by short-range potentials the Fourier transform of the potential is independent of the wave vector. Then all terms of the type $\Sigma_{\mu}, \cdots D_{\mu}^{\alpha}, (G_{\mu}, \cdots) = 0$, by symmetry. Thus only terms proportional to $D_{\nu}^{\alpha}(G_{\nu})$ with ν fixed contribute. The following term in the expansion is

$$T^{(2)}(\nu, \nu') = \langle [\{\{a_{\nu}^{\dagger}a_{\nu}, H'\}, H'\}, r_{\alpha}] \rangle$$

$$= -\delta_{\nu\nu} \sum_{\mu', \mu''} \{W(\mu'', \mu')W(\mu', \nu)[F(\omega, \mu', \nu)F(\omega, \mu'', \nu) + F(\omega, \nu, \mu')F(\omega, \nu, \mu'')] \} D^{\alpha}_{\nu}(G_{\nu}G_{\mu}G_{\mu'})$$

$$- W(\mu', \nu)W(\mu'', \nu)[F(\omega, \mu', \nu)F(\omega, \mu', \mu'') + F(\omega, \nu, \mu')F(\omega, \mu'', \mu'')] \} D^{\alpha}_{\nu}(G_{\nu}G_{\mu}G_{\mu'}) .$$
(50)

It turns out that a class of terms in the series of T can be summed to yield

$$T(\vec{\mathbf{k}}\sigma,\vec{\mathbf{k}}'\sigma') = -\frac{\hbar}{2\pi} \delta_{\vec{\mathbf{k}}\sigma,\vec{\mathbf{k}}'\sigma'} \times \int_{-\infty}^{\infty} d\omega' \left(X^*(\omega',\vec{\mathbf{k}}\sigma,\vec{\mathbf{k}}'\sigma') \frac{\partial G^*_{\vec{\mathbf{k}}\sigma}(\omega')}{\partial k_{\alpha}} - X^-(\omega',\vec{\mathbf{k}}\sigma,\vec{\mathbf{k}}'\sigma') \frac{\partial G^*_{\vec{\mathbf{k}}\sigma}(\omega')}{\partial k_{\alpha}} \right) f(\omega'),$$
(51)

where X satisfies the integral equation

$$X^{\pm}(\omega', \nu, \nu') = 1 + \sum_{\mu} G^{\pm}_{\mu}(\omega') [W(\mu, \nu)F(\omega, \nu', \mu)X^{\pm}(\omega', \mu, \nu') - W(\mu, \nu')F(\omega, \mu, \nu)X^{\pm}(\omega', \nu, \mu)].$$
(52)

When iterated for solution, it is observed that approximatively Eq. (52) consists of two geometrical series provided the wave-vector dependence of W is weak. With these approximations the solution is

$$X^{\pm}(\omega', \nu, \nu) = 1 + \frac{K^{\pm}_{\nu}(\omega')}{1 - K^{\pm}_{\nu}(\omega')} - \frac{L^{\pm}_{\nu}(\omega')}{1 + L^{\pm}_{\nu}(\omega')}, \qquad (53)$$

where

$$K_{\nu}^{\pm}(\omega') = \sum_{\mu} G_{\mu}^{\pm}(\omega') W(\mu, \nu) F(\omega, \nu, \mu) ,$$

$$L_{\nu}^{\pm}(\omega') = \sum_{\mu} G_{\mu}^{\pm}(\omega') W(\mu, \nu) F(\omega, \mu, \nu) .$$
(54)

The last two terms in Eq. (53) give the corrections to the ordinary acceleration term. In the weak-coupling limit the corrections vanish. In the intermediate-coupling case the corrections tend to reduce the ordinary acceleration term at small-k values. When the wave vector increases, the corrections again vanish. Consequently, the corrections to the acceleration term tend to further decrease the mobility.

Figure 4 shows the average dc mobility in EuS and EuO. The mobility is calculated from Eq. (45), taking into account the acceleration-term corrections given in Eqs. (51)-(54). The average mobility is defined by

$$\mu = (\sigma_{\dagger} + \sigma_{\downarrow})/e(n_{\dagger} + n_{\downarrow}), \qquad (55)$$

where $n_{t,+}$ denotes the concentration of spin-up (down) electrons. For the sake of comparison the results derived from the Boltzmann equation are also shown. As seen, the intermediate-coupling theory gives in general smaller mobilities than the weak-coupling theory, even by a factor 0.1 above T_c in EuS. Furthermore, the narrow minimum occurring at T_c in the weak-coupling case becomes considerably broadened in temperature. The minimum mobilities are about $3 \text{ cm}^2/\text{V}$ sec in EuS and EuO. Then the mobility increases with increasing temperature, reaching values of the order of $20 \text{ cm}^2/\text{V}$ sec at room temperature in both materials. In the ferromagnetic region the mobility increases rapidly owing to the onset of the long-range order, which effectively quenches the spin fluctuations.

IV. DISCUSSION

The functional-derivative method provides a straightforward procedure for treatment of the s-f interaction. The intermediate-coupling theory presented above successfully explains the behavior of the absorption edge. In addition to the red shift of the edge, a blue shift of magnetic origin was found in the paramagnetic region. Discussion of mobility is more complicated. Ferromagnetic semiconductors typically show a resistivity peak near T_c . The height of the peak increases by orders of magnitude when doping decreases. The key question is to what extent the resistivity peak is caused by mobility changes compared to changes in carrier concentration. Experimentally the mobility is usually determined by the combined conductivity and Hall-effect measurements. Unfortunately, the resistivity peak is sensitive to magnetic field, which introduces complications.



FIG. 4. dc mobility in EuS and EuO calculated by means of the intermediate-coupling theory (full lines) and by the weak-coupling theory (dashed lines).

Shapira et al.^{35,36} succeeded in measuring the Hall-effect at all temperatures in EuS and EuO in samples where the carrier concentration exceeded 10^{18} cm⁻³. The data are more complete for EuO.³⁶ Typically $\mu(300 \text{ K}) = 30 \text{ cm}^2/\text{V} \text{ sec}$, the minimum mobility is about 10 cm^2/V sec slightly above T_c , and then the mobility increases rapidly in the ferromagnetic region. Owing to the utilization of the Hall effect, the reported mobilities are limited to region where the external magnetic field B is at least of the order of 0.5 T. The zero-field mobility can differ considerably from the previous results. Still, it can be concluded (Ref. 36, Figs. 3 and 7) that the minimum zero-field mobility in EuO is of the order of 1 cm²/V sec for $n > 10^{18}$ cm⁻³. The mobility in EuS is similar to that for previous case, but the minimum values can be of the order of 10^{-2} cm²/V sec (Ref. 35, Fig. 1). Recently Kajita et al.37 have obtained mobility results for pure EuO using fast-photopulse techniques. Their results are limited to $T \leq T_c$ and B > 0.5 T.

Extension of the mobility results to the zerofield case requires that the Hall-effect measurements be replaced by another experiment. Oliver *et al.*³⁸ were able to measure the free-carrier absorption in the infrared region at all temperatures in EuO where $n > 10^{18}$ cm⁻³. Their mobility

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results follow the behavior described above. Similar results were obtained by Schoenes and Wachter³⁹ in EuO from combined free-carrier absorption, Faraday-rotation, and plasma-resonance experiments for a sample with $n > 10^{19}$ cm⁻³.

Photoconductivity has also been utilized to determine carrier concentration and mobility.⁴ In general, the photoconductivity is a quantum efficiency-mobility-lifetime product $\beta\mu\tau$. The estimation of the recombination lifetime τ is particularly difficult. However, the transient-photoconductivity experiments seem to indicate that $\beta\tau$ is almost constant for temperatures near T_c .⁴⁰ When this is adopted, it is concluded from the photoconductivity data,⁴ that the minimum mobilities near T_c are in undoped samples of the order of 1 cm²/V sec in EuO and 10⁻¹-10⁻² cm²/Vsec in EuS.

The behavior of the mobility described above compares reasonably well with the calculated results in Fig. 4. However, according to the photoconductivity experiments, the calculated mobility minimum in EuS should be deeper.

ACKNOWLEDGMENT

The author would like to thank the Academy of Finland for financial support.

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