

Conduction band behaviour in EuTe

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Abstract

The variation of the electronic density of states with temperature in antiferromagnetic semiconductors is investigated theoretically based on the s - f model, taking into account multiple scattering at the same site. It is suggested that the conduction band in an antiferromagnetic semiconductor splits into two subbands when spontaneous magnetization arises below the Néel temperature (T_N). Based on the calculated result for the bottom of the conduction band, we consistently explain not only the magnetic redshift of the optical absorption edge observed in EuO, EuS and EuSe, but also the magnetic blue shift in EuTe.

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1. Introduction

In the previous paper¹⁾, we showed that the anomalous redshift of the optical absorption edge and the temperature dependence of the electron-spin polarization, which were experimentally observed in EuO and EuS, can be explained consistently by applying the single-site approximation to the s - f model in a ferromagnetic semiconductor. In this work, extending the previous method to antiferromagnetic semiconductors, we intend to explain the magnetic blue shift of the optical absorption edge observed in EuTe.

Compared with that for ferromagnetic semiconductors, the theoretical treatment for antiferromagnetic semiconductors is rather complicated²⁻⁶⁾ because the spontaneous magnetization develops in opposite orientations for different sublattices below T_N . Here we assume that magnetic moments (hereafter referred to as f spins) are situated regularly on two interpenetrating ferromagnetic sublattices, A and B. We take the $+z$ direction as the orientation of the spontaneous magnetization of the f spin at the sites of the A sublattice, and we assume that no external field is applied. Thus, when the spontaneous magnetization of sublattice A is $\langle S_z^A \rangle_{av} (= \langle S_z \rangle_{av})$, that of sublattice B is $\langle S_z^B \rangle_{av} (= -\langle S_z \rangle_{av})$, where $\langle \rangle_{av}$ represents a thermal average.

When a single electron (hereafter referred to as an s electron) is injected into an otherwise empty conduction band, it moves in the crystal while interacting with f spins through the s - f exchange interaction. This situation can be described well by the s - f exchange model.^{1,7)} In this model, the total Hamiltonian, H_t , consists of H_s , H_f and H_{sf} which represent the translational energy of an s electron, the Heisenberg exchange interaction between f spins, and the s - f exchange interaction between an s electron and f spins, respectively. The notation used in this work is the same as that used in Ref. 1. However, H_f here represents antiferromagnetic ordering, and thus the lattice index m , n must be distinguished according to the sublattice species to which the lattice belongs.

When spontaneous magnetization arises, the effective potential to which an s electron is subjected at each site differs according to the orientation (i.e., \uparrow or \downarrow) of the electron's spin and to the sublattice species (i.e., A or B) of the site.

Pioneering work on the density of states in antiferromagnetic semiconductors was performed using a mean-field approach ("Zener model") by von der Linden and Nolting²⁾. In this approach, the effective potentials to which s electrons with up-spin are subjected are

assumed to be $-I\langle S_z \rangle_{\text{av}}$ for the A sublattice sites and $+I\langle S_z \rangle_{\text{av}}$ for the B sublattice sites ; no spin-flip process is taken into account. The energy of a conduction electron with up-spin is obtained as

$$\varepsilon(k, \uparrow) = \varepsilon_a(k) \pm \sqrt{|\varepsilon_2(k)|^2 + (I\langle S_z \rangle_{\text{av}})^2}. \quad (1.1)$$

The Bloch energies due to electron transfer between similar sublattice sites, $\varepsilon_1(k)$ ($=\varepsilon^{AA}(k)$ $=\varepsilon^{BB}(k)$), and between different sublattice sites, $\varepsilon_2(k)$ ($=\varepsilon^{AB}(k)=\varepsilon^{BA}(k)$), are defined respectively, as

$$\varepsilon_1(k) = \varepsilon^{AA}(k) = \frac{1}{N} \sum_{m \in A, n \in A} \varepsilon_{mn} e^{ik(m-n)}, \quad (1.2)$$

$$\varepsilon_2(k) = \varepsilon^{AB}(k) = \frac{1}{N} \sum_{m \in A, n \in B} \varepsilon_{mn} e^{ik(m-n)}, \quad (1.3)$$

where ε_{mn} is the nearest-neighbor hopping integral. Equation (1.1) represents splitting of the Bloch band into two subbands below T_N ; the splitting disappears in the paramagnetic region $T \geq T_N$. Each of the energy levels, $\varepsilon(k, \uparrow)$ and $\varepsilon(k, \downarrow)$, is spin-degenerated at least as long as no external field is applied. Note that the mean field approach gives an energy of

$$\varepsilon(k, \uparrow) = \varepsilon_1(k) + \varepsilon_2(k) \quad (1.4)$$

at $T \geq T_N$, which is equal to the Bloch energy for $IS=0$, and

$$\varepsilon(k, \uparrow) = \varepsilon_1(k) \pm \sqrt{|\varepsilon_2(k)|^2 + (IS)^2} \quad (1.5)$$

at $T=0$. Therefore, the mean-field approach predicts a distinct redshift of the lower band edge upon cooling below T_N . This contradicts the experimental observation that a magnetic blue shift is observed in antiferromagnetic semiconductors. This discrepancy may arise because multiple scattering is not taken into account by the mean field approach. The aim of the present study is to investigate the effect of multiple scattering on the conduction electrons in an antiferromagnetic semiconductor, by applying the single-site approximation¹ to the s - f model.

2. Basic Considerations

We first consider an effective medium described by two complex potentials, Σ_p and Σ_a . $\Sigma_p(\Sigma_a)$ represents the short-range potential to which a s electron is subjected when the orientation of its spin is parallel (antiparallel) to the orientation of the spontaneous magnetic moment at that site. (The mean-field approach corresponds to setting $\Sigma_p = -I\langle S_z \rangle_{\text{av}}$ and $\Sigma_a = +I\langle S_z \rangle_{\text{av}}$. Thus, an s electron moving in this effective medium is described by the (unperturbed) reference Hamiltonian K :

$$K = \sum_{m\mu} \varepsilon_{m\mu} a_{m\mu}^\dagger a_{n\mu} + \sum_{m\mu} \Sigma_m^\mu a_{m\mu}^\dagger a_{n\mu}, \quad (2.1)$$

with

$$\Sigma_m^\mu = \begin{cases} \Sigma_p & \text{when } \mu = \uparrow \text{ and } m \in A, \text{ or, when } \mu = \downarrow \text{ and } m \in B \\ \Sigma_a & \text{when } \mu = \downarrow \text{ and } m \in A, \text{ or, when } \mu = \uparrow \text{ and } m \in B, \end{cases} \quad (2.2)$$

where $m \in A(B)$ means that site m belongs to sublattice A(B). Since K includes no spin-flip term, the orientation of the spin of an electron moving in the effective medium remains unchanged. Furthermore, the formula is symmetrical for both \uparrow and \downarrow spins.

Next, we set the Bloch electron state with up-spin in the effective medium as

$$|k, \uparrow\rangle = c_A |Ak, \uparrow\rangle + c_B |Bk, \uparrow\rangle. \quad (2.3)$$

Here, $|Ak, \uparrow\rangle$ ($|Bk, \uparrow\rangle$) is the Bloch state related to the Wannier states on the m site of the A(B) sublattice, $|m \uparrow\rangle$, as

$$|Ak, \uparrow\rangle = \sqrt{\frac{2}{N}} \sum_{m \in A} e^{ik \cdot m} |m \uparrow\rangle, \quad (2.4)$$

$$|Bk, \uparrow\rangle = \sqrt{\frac{2}{N}} \sum_{m \in B} e^{ik \cdot m} |m \uparrow\rangle, \quad (2.5)$$

Note that the number of each sublattice is $N/2$, and so these functions satisfy the following orthonormalities:

$$\langle Ak, \uparrow | Ak, \uparrow \rangle = \langle Bk, \uparrow | Bk, \uparrow \rangle = 1, \quad (2.6)$$

$$\langle Ak, \uparrow | Bk, \uparrow \rangle = \langle Bk, \uparrow | Ak, \uparrow \rangle = 0. \quad (2.7)$$

Next, we consider the solution of the eigenvalue equation

$$K |k, \uparrow\rangle = E(k) |k, \uparrow\rangle. \quad (2.8)$$

This leads to the simultaneous equations

$$c_A [K_{AA} - E(k)] + c_B K_{AB} = 0, \quad (2.9)$$

$$c_A K_{BA} + c_B [K_{BB} - E(k)] = 0, \quad (2.10)$$

with

$$K_{AA} = \langle Ak, \uparrow | K | Ak, \uparrow \rangle = \varepsilon_1(k) + \Sigma_p, \quad (2.11)$$

$$K_{BB} = \langle Bk, \uparrow | K | Bk, \uparrow \rangle = \varepsilon_1(k) + \Sigma_a, \quad (2.12)$$

$$K_{AB} = \langle Ak, \uparrow | K | Bk, \uparrow \rangle = \varepsilon_2(k), \quad (2.13)$$

$$K_{BA} = \langle Bk, \uparrow | K | Ak, \uparrow \rangle = \varepsilon_2(k). \quad (2.14)$$

By solving the secular equation, we obtain two energy eigenvalues, $E_p(k)$ and $E_a(k)$. Since K is not Hermitian, the energy eigenvalues are generally complex and the two eigenfunctions are not orthogonal. Nevertheless, for further calculation we assume here that the eigenfunctions corresponding to the two eigenvalues, $E_p(k)$ and $E_a(k)$, are orthogonal to each other. This assumption simplifies the calculation considerably. (Note that this problem does not arise for ferromagnetic semiconductors. Since the two eigenfunctions are characterized by the spin of the electrons, they are orthogonal to each other. See also later discussion.)

Now, we consider the f spin located at the 0-site (A sublattice) in the effective medium described by K . Defining the reference Green's function P as

$$P(\omega) = \frac{1}{\omega - K}, \quad (2.15)$$

the diagonal matrix elements of P in Wannier representations are calculated as $F_+(\omega) = \langle 0 \uparrow | P | 0 \uparrow \rangle$ and $F_-(\omega) = \langle 0 \downarrow | P | 0 \downarrow \rangle$.

In order to determine Σ_p and Σ_a , we apply the condition that the average scattering by a single f spin in the medium is zero, as in Ref. 1. For the elements of the t matrix of the s - f exchange interactions,

$$\langle 0 \uparrow | t | 0 \uparrow \rangle_{\text{av}} = 0, \quad (2.16)$$

$$\langle 0 \downarrow | t | 0 \downarrow \rangle_{\text{av}} = 0. \quad (2.17)$$

When the above conditions are applied, only the replacements Σ_p for Σ_\uparrow and Σ_a for Σ_\downarrow are needed. It is easily verified that our treatment corresponds to the coherent potential approximation (CPA) at $T \geq T_N$ ⁸⁾.

In order to calculate the density of states, the semicircular band with a half-bandwidth of Δ is introduced as an undisturbed density of states (i.e., for $IS=0$),⁸⁾

$$\rho(\varepsilon) = \frac{2}{\pi\Delta} \sqrt{1 - \left(\frac{\varepsilon}{\Delta}\right)^2}. \quad (2.18)$$

For $T < T_N$, we need the Bloch energies $\varepsilon_1(k)$ and $\varepsilon_2(k)$, which are related to the crystal structure. In this study, we consider the following cases.

(i) sc-structure (von der Linder and Nolting²⁾ called it the ABAB-structure) and bcc-structure. Since the nearest neighbors of each site are located on the other sublattice, $\varepsilon_1(k)$ is 0. Thus, we simply replace $\varepsilon_2(k)$ by ε . The results for this case are summarized as follows:

$$E_p(\varepsilon) = \frac{\Sigma_p + \Sigma_a - \sqrt{4\varepsilon^2 + (\Sigma_p - \Sigma_a)^2}}{2}, \quad (2.19)$$

$$E_a(\varepsilon) = \frac{\Sigma_p + \Sigma_a + \sqrt{4\varepsilon^2 + (\Sigma_p - \Sigma_a)^2}}{2}, \quad (2.20)$$

$$|c_A(\varepsilon)|^2 = \frac{\varepsilon^2}{\varepsilon^2 + |E_p - \Sigma_p|^2}, \quad (2.21)$$

$$|c_B(\varepsilon)|^2 = \frac{|E_p - \Sigma_p|^2}{\varepsilon^2 + |E_p - \Sigma_p|^2}. \quad (2.22)$$

(ii) fcc-structure (von der Linden and Nolting called it the NSNS-structure). Since the nearest neighbors for each site consist of equal numbers of sites from both sublattices, we simply replace $\varepsilon_1(k)$ and $\varepsilon_2(k)$ by $\varepsilon/2$. The results for the fcc-structure are summarized as follows:

$$E_p(\varepsilon) = \frac{\varepsilon + \Sigma_p + \Sigma_a - \sqrt{\varepsilon^2 + (\Sigma_p - \Sigma_a)^2}}{2}, \quad (2.23)$$

$$E_a(\varepsilon) = \frac{\varepsilon + \Sigma_p + \Sigma_a + \sqrt{\varepsilon^2 + (\Sigma_p - \Sigma_a)^2}}{2}, \quad (2.24)$$

$$|c_A(\varepsilon)|^2 = \frac{\varepsilon^2}{\varepsilon^2 + |2(E_p - \Sigma_p) - \varepsilon|^2}, \quad (2.25)$$

$$|c_B(\varepsilon)|^2 = \frac{|2(E_p - \Sigma_p) - \varepsilon|^2}{\varepsilon^2 + |2(E_p - \Sigma_p) - \varepsilon|^2}. \quad (2.26)$$

In both cases, the summation over k is replaced by the integration with respect to ε , using model state density $\rho(\varepsilon)$. Accordingly,

$$F_\uparrow(\omega) = 2 \int_{-\Delta}^0 d\varepsilon \rho(\varepsilon) \frac{|c_A(\varepsilon)|^2}{\omega - E_p(\varepsilon)} + 2 \int_0^\Delta d\varepsilon \rho(\varepsilon) \frac{|c_B(\varepsilon)|^2}{\omega - E_a(\varepsilon)} \quad (2.27)$$

$$F_\downarrow(\omega) = 2 \int_{-\Delta}^0 d\varepsilon \rho(\varepsilon) \frac{|c_B(\varepsilon)|^2}{\omega - E_p(\varepsilon)} + 2 \int_0^\Delta d\varepsilon \rho(\varepsilon) \frac{|c_A(\varepsilon)|^2}{\omega - E_a(\varepsilon)} \quad (2.28)$$

In all of the present numerical calculations, the total density of states should satisfy

$$\int_{-\infty}^{\infty} D_\mu(\omega) d\omega = -\frac{1}{\pi} \text{Im} \int_{-\infty}^{\infty} F_\mu(\omega) d\omega = 1.0 \quad (2.29)$$

for both $\rho = \uparrow$ and \downarrow .

3. Results and Discussion

In this work, the f spin is treated as a classical spin (i.e., $1/S \rightarrow 0$). Thus, $D(\omega) = D(-\omega)$, and the values of $\langle S_z \rangle_{\text{av}}/S$, calculated using the molecular field theory, are 0.0, 0.39, 0.79 and 1.0 for $T = T_N$, $0.09 T_N$, $0.05 T_N$ and 0, respectively.

Figure 1 shows the result for the sc- (or bcc-) structure with $IS/2\Delta = 0.1$. The product of the density of states and the half-bandwidth, $D(\omega)\Delta (= D_+(\omega)\Delta + D_-(\omega)\Delta)$, is shown as a function of the reduced energy, ω/Δ , for various temperatures. At paramagnetic temperatures, $T \geq T_N$, the present result agrees with that obtained using the CPA.⁸ Below T_N the conduction band splits into two subbands; the low (high)-energy subband corresponds to coupling of the electron spin parallel (antiparallel) to the orientation of the f spins. At $T = 0$, Eqs. (2.16) and (2.17) give $\Sigma_p = -IS$ and $\Sigma_a = +IS$, and so we obtain

$$D(\omega)\Delta = \frac{4|\omega|}{\pi\Delta} \left\{ \frac{\Delta^2 + (IS)^2 - \omega^2}{\omega^2 - (IS)^2} \right\}^{\frac{1}{2}} \quad (3.1)$$

for $IS < |\omega| < \sqrt{\Delta^2 + (IS)^2}$, and 0 otherwise. It is verified that the density of states given by Eq. (3.1) satisfies the condition (2.29), although Eq. (3.1) diverges at $\omega = \pm IS$.

A similar result for the fcc-structure with $IS/2\Delta = 0.1$ is shown in Fig. 2. At $T = 0$, we obtain

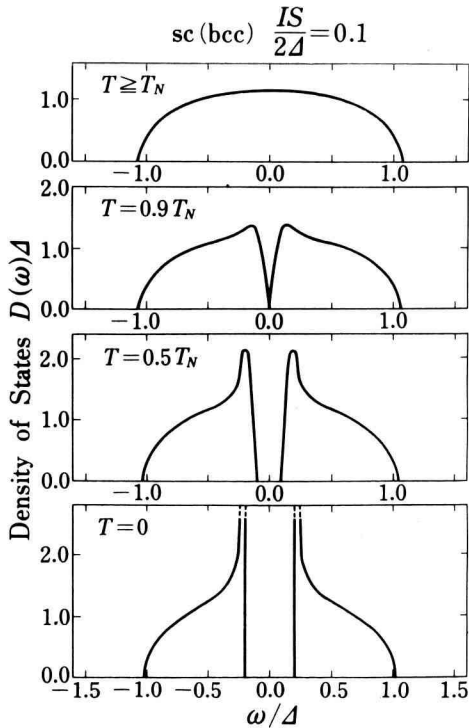


Fig. 1 The density of states in an sc (bcc) antiferromagnetic semiconductor with $IS/2\Delta = 0.1$ for $T \geq T_N$, $T = 0.9 T_N$, $0.5 T_N$ and 0.

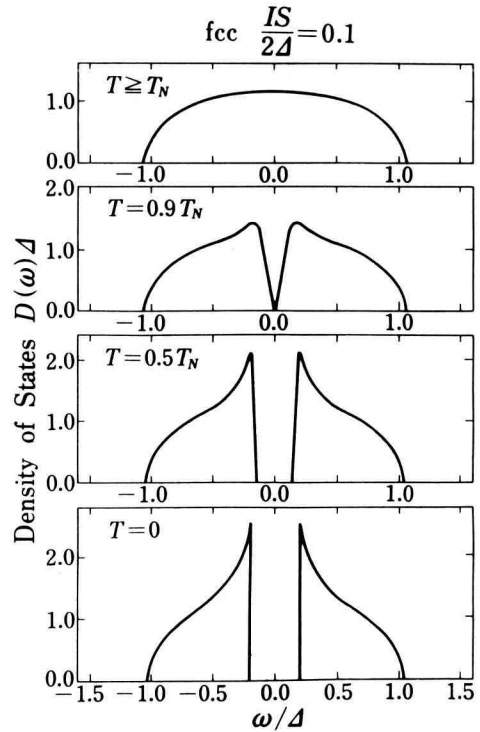


Fig. 2 The density of states in an fcc antiferromagnetic semiconductor with $IS/2\Delta = 0.1$ for $T \geq T_N$, $T = 0.9 T_N$, $0.5 T_N$ and 0.

$$D(\omega)\Delta = \frac{4}{\pi} \left(\frac{\Delta}{\omega} \right)^2 \left[\left(\frac{\omega}{\Delta} \right)^2 + \left(\frac{IS}{\Delta} \right)^2 \right] \left\{ 1 - \left(\frac{\Delta}{\omega} \right)^2 \left[\left(\frac{IS}{\Delta} \right)^2 - \left(\frac{\omega}{\Delta} \right)^2 \right] \right\}^{\frac{1}{2}} \quad (3.2)$$

for $IS < |\omega| < (\Delta + \sqrt{\Delta^2 + 4(IS)^2})/2$, and 0 otherwise.

It is worth noting that at $T=0$ both Σ_p and Σ_a are real and so K is Hermitian. Thus, the two energy eigenfunctions are orthogonal to each other.

For $0 < T < T_N$, when $IS/2\Delta = 0.1$, the discrepancy between the numerically estimated values for the left-hand side in Eq. (2.29) and 1.0 is within the numerical error range. However, when the exchange interaction becomes stronger, such as $IS/2\Delta \geq 0.2$, the discrepancy exceeds the numerical error range. This is probably due to the assumption that the eigenfunctions corresponding to E_p and E_a are approximately orthogonal to each other. This is not true for $0 < T < T_N$.

In Fig. 3, the energy decrease due to the s - f exchange interaction, $\delta\omega_b$, is shown as a function of $IS/2\Delta$ for $T \geq T_N$ and $T=0$. The energy decrease of the bottom of the band (from $\omega = -\Delta$), $\delta\omega_b$, is calculated using the CPA⁸⁾ for $T \geq T_N$ as

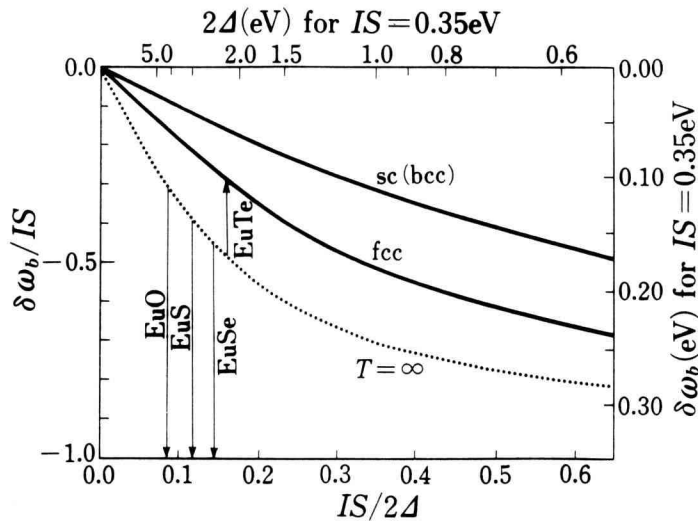


Fig. 3 The (reduced) energy decrease of the bottom of the conduction band, $\delta\omega_b/IS$, as a function of $IS/2\Delta$. The dotted line is for $T=\infty$ and the solid lines are for the sc (bcc) antiferromagnetic semiconductor at $T=0$, and fcc antiferromagnetic semiconductor at $T=0$. In each case, classical spin is assumed for the f spins. The arrows show the magnetic redshift for EuO, EuS and EuSe and the blue shift for EuTe.

Table 1

	2Δ (eV)	$IS/2\Delta$	Theory (eV)	Experiment (eV)
EuO	4.2	0.083	0.24	0.27
EuS	3.0	0.117	0.21	0.18
EuSe	2.5	0.140	0.19	0.13
EuTe	2.2	0.159	-0.067	-0.03

($IS = 0.35$ eV)

$$\frac{\delta\omega_b}{IS} = -\frac{1}{8}\left(\frac{2\mathcal{A}}{IS}\right)\left\{\sqrt{1+64\left(\frac{IS}{2\mathcal{A}}\right)^2}-1\right\}. \quad (3.3)$$

In an antiferromagnetic semiconductor, the results for $T < T_N$ differ according to the crystal structure. In particular, when $T=0$, for the sc- (bcc-) structure,

$$\frac{\delta\omega_b}{IS} = -\frac{1}{2}\left(\frac{2\mathcal{A}}{IS}\right)\left\{\sqrt{1+4\left(\frac{IS}{2\mathcal{A}}\right)^2}-1\right\}, \quad (3.4)$$

and for the fcc-structure,

$$\frac{\delta\omega_b}{IS} = -\frac{1}{4}\left(\frac{2\mathcal{A}}{IS}\right)\left\{\sqrt{1+16\left(\frac{IS}{2\mathcal{A}}\right)^2}-1\right\}. \quad (3.5)$$

Using the results in Fig. 3, we explain the temperature dependence of the optical absorption edge observed in Eu-chalcogenides. In a ferromagnetic semiconductor, when the temperature T decreases from ∞ to 0, the energy of the bottom of the band shifts from $\delta\omega_b$ given by Eq. (3.3) to $-IS$. This is the magnetic redshift of the optical absorption edge observed in EuO, EuS and EuSe. On the other hand, the energy of the bottom of the band in an antiferromagnetic semiconductor at $T=0$ is given by Eq. (3.4) or (3.5). Thus, the optical absorption edge in an antiferromagnetic semiconductor shows a magnetic blue shift, as observed in EuTe.

In Table 1, we show a comparison of the present theoretical results and the experimental data. Using the bandwidth $2\mathcal{A}$ taken from Wachter^{9,10}, we calculate the shift in the energy of the bottom of the band. The agreement with the experimental results is satisfactory.

We also investigate the quantum effect of the f spin. For a finite value of S , instead of Eq. (3.3), we use

$$\frac{\delta\omega_b}{IS} = -\frac{1}{8}\left(\frac{2\mathcal{A}}{IS}\right)\left\{\sqrt{1+\frac{8}{S}\left(\frac{IS}{2\mathcal{A}}\right)+64\left(\frac{IS}{2\mathcal{A}}\right)^2\left(1+\frac{1}{2S}\right)}-1-\frac{4}{S}\left(\frac{IS}{2\mathcal{A}}\right)\right\} \quad (3.6)$$

at $T \geq T_N$ ¹¹. Thus, when $S=7/2$, the theoretical results for the magnetic redshift are 0.224 eV (EuO), 0.193 eV (EuS) and 0.175 eV (EuSe). Agreement with the experimental data is reasonable. (Note that when the free-electron band with bandwidth W is used instead of a semicircular band, $IS/2\mathcal{A}$ is replaced by $(3/4)(IS/W)$ in Eq. (3.6)).

4. Concluding remarks

We aim to devise an improved s - f model for antiferromagnetic semiconductors, which is applicable to wide ranges of $IS/2\mathcal{A}$ and temperature.

For this purpose, we first studied the energy eigenvalues and eigen-functions for an s electron moving in an effective medium described by the complex potentials, Σ_p and Σ_a . Next, we calculated the density of states, using Σ_p and Σ_a determined under the condition that the average scattering by a single f spin in the medium is zero, together with the assumption that the energy eigenfunctions are orthogonal to each other. The multiple scattering at one site is taken into consideration within the t matrix of the s - f exchange interaction. Thus, the present treatment agrees with the CPA at $T \geq T_N$. Furthermore, assuming a semicircular model density for $IS=0$, we present the analytical results at $T=0$ for an antiferromagnetic semiconductor with sc- (bcc-) and fcc-structures in the classical spin limit. The variation of the electric density of states with temperature is demonstrated for $IS/2\mathcal{A}=0.1$.

The results showed that with decreasing temperature, the single band at $T \geq T_N$ splits into two subbands below T_N , which are characterized by coupling of the electron spin parallel and antiparallel to the orientation of the f spin.

We also show the energy decreases of the bottom of the band as a function of $IS/2J$ at $T=0$ and ∞ . Based on these results, we showed that not only the magnetic redshift of the optical absorption edge observed in EuO, EuS and EuSe, but also magnetic blue shift in EuTe, can be explained consistently using the single-site approximation.

However, when $0 < T < T_N$, our method is not applicable for a strong exchange interaction such as $\frac{IS}{2J} \gtrsim 0.2$. This is probably due to the assumption that the eigenfunctions for K are orthogonal to each other. Since K is not Hermitian, another improved approach is necessary, especially for strong exchange interaction. This problem will be treated elsewhere.¹¹⁾

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