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Exchange interaction mechanisms in magnetic semiconductors

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Effective exchange interaction in magnetic semiconductors is deduced by taking into account various factors: hybridization of the f-electron states with the valence and conduction bands, total interatomic exchange including the interband interaction of the electrons, Coulomb repulsion effect, and the effect of the finite f-level width. The calculated values of the exchange constants I_1 and I_2 for europium chalcogenides are in good agreement with the experimental values.

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1. In the description of the properties of magnetic dielectrics and semiconductors based on transition and rare-earth metals (REM), various mechanisms of exchange interaction are resorted to. Whereas the Kramers superexchange interaction mechanism[1] suffices to explain the magnetism of magnetic dielectrics, which are antiferromagnetic in most cases, the situation is more complicated in the case of magnetic semiconductors based on REM. Thus, for example, in the series of of europium-chalcogenides, which have a more complicated electronic structure, one observes a broad spectrum of magnetic properties on going from compound to compound or when the lattice parameter is altered by pressure.[2,3] Xavier and de Graaf[4,5] attempted to explain both the ferromagnetic and antiferromagnetic exchange interactions in EuO and EuS by using only the Bloembergen–Rowland interband-interaction mechanism.[6] Their overstated values of the interband-interaction constants can, however, not be regarded as acceptable.

An analogous mechanism, due to excitation of electrons from localized p orbits of anions into the conduction band, was proposed by Berdyshev and Letfulov[7] and by Kazakov.[8] The data they used, however, were taken from experimental papers[9,10] whose authors incorrectly identified the forbidden gap width B with the f-level1) binding energy Δ. The real values of the parameters B (see Table I) in europium chalcogenides are 2–6 times larger than those assumed in the aforementioned theoretical papers,[7,8] so that the obtained exchange integrals are smaller by one or two orders of magnitude than those given in Table II of[7]. Thus, the interband-interaction mechanism alone is not sufficient to obtain good agreement with the experimental data.

It follows from magneto-optical measurements and from calculations of the band structure with allowance for the strong f-electron correlation[12] and the lattice polarization[13] that in europium chalcogenides the localized weakly smeared f level lies in the forbidden gap

TABLE I.

	Δ, eV	B, eV	U, eV	R_0, Å
EuO	1.1	6.3	10	5.15
EuS	1.7	5.2	11 *	5.92
EuSe	1.9	4	12	6.2
EuTe	2.0	3.8 *	13 *	6.6

between the valence and conduction bands.[14] In this case one cannot neglect the mechanisms of the direct and kinetic exchanges via excitations of the f electrons to the $6s$ and $5d-t_{2g}$ levels of europium,[15] say on account of hybridization of the localized f states with empty conduction band[16] or zero-point lattice vibrations.[17] However, the lattice mechanism in europium chalcogenides is apparently strongly suppressed, since the Curie temperature does not reveal any isotopic effects or anomalies in the paramagnetic susceptibility. In addition to the foregoing mechanism, other explanations, frequently quite complicated ones,[3] were also offered for the exchange interaction in magnetic semiconductors.

The presence of a transition integral leads to the onset, besides the usual Kramers–Anderson superexchange interaction, to new indirect exchange interactions connected with the interaction of the f electrons with the empty conducting band and the filled valence band. Estimates show that an appreciable contribution to the exchange interaction is made also by mechanisms with participation of excited $4f^8$ configurations with one excess f electron at the center, so that it is necessary to take into account also excited states that admit of a transition of the magnetic f electrons from cation to cation (directly, $4f^7-4f^8$, or via the empty conduction band).

We obtain below, using the simple model of the band structure of a magnetic semiconductor shown in Fig. 1 (compare with Fig. 24 of [2]), an effective exchange interaction that includes all the mechanisms listed above. Account is taken also of the effects of hybridization of the states of the f electrons and of the local exchange with the valence band.[18] This model provides not only a correct qualitative description of the exchange in magnetic semiconductors, but makes it also possible to obtain good qualitative agreement with the exchange-parameters values obtained experimetally for a large family of magnetic semiconductors of the europium-chalcogenide type.

2. In real magnetic semiconductors, the conduction and valence bands are made up respectively of the states $6s$ and $5d-t_{2g}$ electrons of the metal (cations) and p states of the anions. These states are represented in the figure by the upper band, with a dispersion ϵ_{1k}, separated by a gap Δ from a localized f level of width $\sim W$, and a lower band with a dispersion law ϵ_{2k}, separated by a gap B from the bottom of the conduction band. We consider a case with one localized f electron at the center; we assume that the excitation energy of a system with two f electrons at the center due to Coulomb repulsion, U, is larger than the Fermi energy. All the energies are reckoned from the bottom of the conduction band.

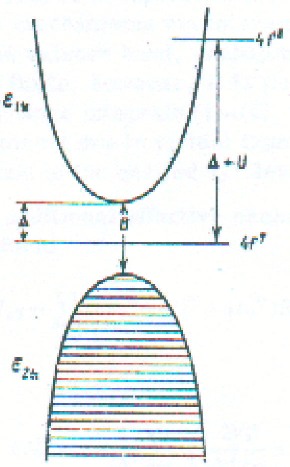

FIG. 1. Model of band structure.

The Hamiltonian of the model is given by

$$H=\sum_{k,\sigma}\varepsilon_{1k}a_{k\sigma}^{+}a_{k\sigma}+\sum_{k,\sigma}\varepsilon_{2k}c_{k\sigma}^{+}c_{k\sigma}+\frac{U}{2}\sum_{j\sigma}b_{j\sigma}^{+}b_{j\sigma}b_{j,-\sigma}^{+}b_{j,-\sigma}$$
$$+(-\Delta)\sum_{j,\sigma}b_{j\sigma}^{+}b_{j\sigma}+\sum_{i,j,\sigma}W_{ij}b_{i\sigma}^{+}b_{j\sigma}+N^{-1/2}\sum_{k,j,\sigma}(V_{1j}(\mathbf{k})a_{k\sigma}^{+}b_{j\sigma}$$
$$+V_{2j}(\mathbf{k})c_{k\sigma}^{+}b_{j\sigma})+\text{э. с.}+\frac{1}{2}\sum_{j,\sigma,\sigma'}\{\mathcal{J}_{1}a_{j\sigma}^{+}a_{j\sigma'}+\mathcal{J}_{2}(a_{j\sigma}^{+}c_{j\sigma'}+c_{j\sigma}^{+}a_{j\sigma'})$$
$$+\mathcal{J}_{3}c_{j\sigma}^{+}c_{j\sigma'}\}b_{j\sigma'}^{+}b_{j\sigma}=\hat{H}_{0}+\hat{W}+\hat{V}+\hat{J}. \qquad ($$

The first two terms of (1) are the kinetic energies of th upper and lower bands respectively; the fourth and fifth terms constitute the energy of the localized f electrons in the site representation with a transition integral equal to W_{ij}. The sixth term represents local hybridization of the f level with both bands, which takes the form $V_{1j}(\mathbf{k})=V_1\exp(i\mathbf{k}\mathbf{R}_j)$, $V_{2j}(\mathbf{k})=V_2\exp(i\mathbf{k}\mathbf{R}_j)$; the seventh term is the local intra-atomic exchange and includes also the interband exchange $\mathcal{J}_2$ due to the interaction of holes and electrons from different bands.[6] Finally, t third term is the intra-atomic repulsion of the f electrons at one center. We note that without the terms J and W the Hamiltonian (1) is a generalization of Anderson's model to the case of two bands.

In the considered substances, the parameters $\mathcal{J}$, V, and W can be regarded as small ($\mathcal{J}$, V, $W\ll\Delta$, B, U), and perturbation theory then yields an effective exchange-interaction Hamiltonian in the form

$$H_{\text{eff}}=\left\langle\hat{V}\frac{1}{E_0-H_0}\hat{V}\frac{1}{E_0-H_0}\hat{V}\frac{1}{E_0-H_0}\hat{V}+\hat{V}\frac{1}{E_0-H_0}\hat{J}\frac{1}{E_0-H_0}\hat{V}\right.$$
$$+\hat{J}\frac{1}{E_0-H_0}\hat{V}\frac{1}{E_0-H_0}\hat{V}+\hat{V}\frac{1}{E_0-H_0}\hat{V}\frac{1}{E_0-H_0}\hat{J}+\hat{J}\frac{1}{E_0-H_0}\hat{J}$$
$$\left.+\hat{V}\frac{1}{E_0-H_0}\hat{W}\frac{1}{E_0-H_0}\hat{V}+\hat{W}\frac{1}{E_0-H_0}\hat{W}\right\rangle. \qquad (2$$

The averaging in (2) is over the 2^N-fold degenerate subspace of the localized functions of the f electrons for a filled valence band and an empty conduction band.

Substituting in (2) the explicit form of the corresponding operators from (1), carrying out the necessary commutations of the operators $a_{k\sigma}(c_{k\sigma})$, $a_{k\sigma}^{+}(c_{k\sigma}^{+})$, and using the usual transition, on the subspace of the homopolar functions ($b_{i+}^{+}b_{i+}+b_{i-}^{+}b_{i-}=1$) from electron operators to spin operators $b_{i\pm}^{+}b_{i\pm} = 1/2 \pm S_i^z$, $b_{i\pm}^{+}b_{i\mp} = S_i^{\pm}$, we obtain in analogy with[16] the expression

$$H_{\text{eff}} = \Delta H_{\text{eff}} + \sum_{i\neq j} (J_{ij}^{(1)} + J_{ij}^{(2)} + J_{ij}^{(3)})\mathbf{S}_i\mathbf{S}_j - 2\sum_{i\neq j} I_1 S_i S_j \tag{3}$$

where

$$J_{ij}^{(1)} = \frac{1}{N^2}\sum_{\mathbf{k}_1,\mathbf{k}_2}\left(\frac{2V_1^2}{\Delta+\varepsilon_{1\mathbf{k}_1}} + \frac{2V_1^2}{\Delta+V} - \mathcal{J}_1\right)\frac{V_1^2\exp\{i(\mathbf{k}_1-\mathbf{k}_2)\mathbf{R}_{ij}\}}{(\Delta+\varepsilon_{1\mathbf{k}_1})(\Delta+\varepsilon_{1\mathbf{k}_2})}, \tag{4}$$

$$J_{ij}^{(2)} = -\frac{1}{2N^2}\sum_{\mathbf{k}_1,\mathbf{k}_2}\left(\frac{2V_1V_2}{\Delta+\varepsilon_{1\mathbf{k}_1}} + \frac{2V_1V_2}{\Delta+U} - \mathcal{J}_2\right)^2\frac{\exp\{i(\mathbf{k}_1-\mathbf{k}_2)\mathbf{R}_{ij}\}}{\varepsilon_{1\mathbf{k}_1}-\varepsilon_{2\mathbf{k}_2}}, \tag{5}$$

$$J_{ij}^{(3)} = -\frac{1}{N}\sum_{\mathbf{k}}\frac{W_{ij}}{\Delta+\varepsilon_{1\mathbf{k}}}\left(\frac{2V_1^2}{\Delta+\varepsilon_{1\mathbf{k}}} + \frac{4V_1^2}{\Delta+U}\right)\exp(i\mathbf{k}\mathbf{R}_{ij}) + \frac{2W_{ij}^2}{\Delta+U} \tag{6}$$

and $\mathbf{R}_{ij} = \mathbf{R} = \mathbf{R}_i - \mathbf{R}_j$ is the distance between the ith and jth atoms.

The first integral here is the contribution made to the exchange interaction by the hybridization and by the local exchange on account of the upper band. The first term in (4) represents the kinetic exchange mechanism and is a direct generalization of the Kramers-Anderson superexchange interaction.

Putting $U \to \infty$ in (4) we arrive a formulas (4) and (5) of[16]. We note that allowance for the Coulomb repulsion U leads to a renormalization of the exchange constant $\mathcal{J}_1$, which does not depend on the momentum. The increment to $\mathcal{J}_1$ is negative here and in the absence of $\mathcal{J}_1$ the term (4) is always antiferromagnetic. The last term in (4) corresponds to the process wherein the electron excited into the empty conduction band interacts in exchange fashion with the remaining magnetic f electrons. The effective constant

$$\frac{2V_1^2}{\Delta+\varepsilon_{1\mathbf{k}}} + \frac{2V_1^2}{\Delta+U} - \mathcal{J}_1$$

can be obtained from Anderson's model by the Schrieffer-Wolff transformation.[19]

The second integral $J^{(2)}$ corresponds to interband exchange ascribed to the upper band. Here, as in (4), the usual constant is replaced by a quantity that depends on the momentum. As seen from (5), at $\mathcal{J}_2 = 0$ the hybridization itself leads to an effective interband interaction. Expression (6) contains three terms: the first two are new exchange-interaction mechanisms due to the integral of the transition with participation of the conduction band and of the excited $4f^8$ level with energy $\Delta + U$, while the last term of (6) is the usual Kramers-Anderson antiferromagnetic exchange.[1]

In the limit of strong repulsion ($U \to \infty$), the term (6) is left with an exchange integral corresponding to a process of third-order perturbation theory, in which the f electron goes from the given center into the conduction band; the vacant place is taken by an f electron from a neighboring atom, and then the electron returns from the band to the center. This process leads to ferromagnetic exchange interaction.

As $U \to \infty$ the strong Coulomb interactions prevents the presence of more than one f electron at the center; in this case the Pauli principle, which excludes electron transitions to occupied states, prevents the onset of exchange mechanisms via interaction of the f electrons with the valence band, analogous to expressions (4)–(6). If U is finite, however, it is possible to have, besides the exchange integrals (4)–(6), also indirect-exchange mechanisms due to virtual transitions of valence-band electrons to the excited $4f^8$ level.

The additional effective exchange interaction in (3) is of the form

$$\Delta H_{\text{eff}} = \sum_{i\neq j}(\Delta J_{ij}^{(1)} + \Delta J_{ij}^{(2)} + \Delta J_{ij}^{(3)})\mathbf{S}_i\mathbf{S}_j,$$

where

$$\Delta J_{ij}^{(1)} = -\frac{1}{2N^2}\sum_{\mathbf{k}_1,\mathbf{k}_2}\left\{\left(\frac{2V_2^2}{\Delta+\varepsilon_{2\mathbf{k}_1}} + \frac{2V_2^2}{\Delta+U} - \mathcal{J}_2\right)^2 - \left(\frac{2V_2^2}{\Delta+\varepsilon_{2\mathbf{k}_1}} + \frac{2V_2^2}{U-\varepsilon_{2\mathbf{k}_1}} - \mathcal{J}_2\right)^2\right\}\frac{\exp\{i(\mathbf{k}_1-\mathbf{k}_2)\mathbf{R}_{ij}\}}{\varepsilon_{2\mathbf{k}_1}-\varepsilon_{2\mathbf{k}_2}}, \tag{7}$$

$$\Delta J_{ij}^{(2)} = -\frac{1}{2N^2}\sum_{\mathbf{k}_1,\mathbf{k}_2}\left\{\left(\frac{2V_1V_2}{\Delta+\varepsilon_{2\mathbf{k}_1}} + \frac{2V_1V_2}{\Delta+U} - \mathcal{J}_2\right)^2 - \left(\frac{2V_1V_2}{\Delta+\varepsilon_{2\mathbf{k}_1}} + \frac{2V_1V_2}{U-\varepsilon_{2\mathbf{k}_1}} - \mathcal{J}_2\right)^2\right\}\frac{\exp\{i(\mathbf{k}_1-\mathbf{k}_2)\mathbf{R}_{ij}\}}{\varepsilon_{2\mathbf{k}_1}-\varepsilon_{1\mathbf{k}_2}}, \tag{8}$$

$$\Delta J_{ij}^{(3)} = -\frac{1}{N}\sum_{\mathbf{k}}\frac{2W_{ij}}{\Delta+\varepsilon_{2\mathbf{k}}}\left(\frac{V_2^2}{\Delta+\varepsilon_{2\mathbf{k}}} + \frac{2V_2^2}{\Delta+U}\right)\exp(i\mathbf{k}\mathbf{R}_{ij}) + \frac{1}{N}\sum_{\mathbf{k}}2W_{ij}\left(\frac{V_2}{U-\varepsilon_{2\mathbf{k}}} + \frac{V_2}{\Delta+\varepsilon_{2\mathbf{k}}}\right)^2\exp(i\mathbf{k}\mathbf{R}_{ij}). \tag{9}$$

The exchange integral $\Delta J^{(1)}$ represents here an intraband contribution analogous to (4) to the exchange interaction, but due to the valence-band electrons, $\Delta J^{(2)}$ corresponds to interband exchange ascribed to the lower band, and finally $\Delta J^{(3)}$ is an exchange integral analogous to (6) but with a completely filled band.

As $U \to \infty$, expressions (7)–(9) vanish, as expected, and the effective exchange is determined by the contribution of the integrals $J^{(1)}$, $J^{(2)}$, and $J^{(3)}$. It is easy to verify, by substituting the data from Table I, that for all cases of practical importance the integrals (7)–(9) at $U > \Delta$ are smaller than (4)–(6). These integrals can be grouped with the corresponding terms in (4)–(6).

It is interesting to note that by assuming $U \to \infty$ and $\mathcal{J}_{1,2,3} = 0$ we obtain, in the framework of the single-particle Hamiltonian, an effective exchange interaction that consists of ferromagnetic and antiferromagnetic parts.

3. The exchange integrals (4)–(6) can be calculated by assuming a parabolic dispersion law for both bands in the form $\epsilon_{1\mathbf{k}} = \hbar^2k^2/2m_1$ and $\epsilon_{2\mathbf{k}} = -B - \hbar^2k^2/2m_2$. By changing in the usual fashion from summation to integration over the momenta

$$\frac{1}{N}\sum_{\mathbf{k}} \to \frac{\Omega}{(2\pi)^3}\int d^3k$$

($\Omega = v/N$ is the volume per atom) we obtain an analytic expression for the integral $J^{(1)}$[16]:

$$J^{(1)} = \left(\frac{V_1^4\Omega^2 m_1^2}{2\pi^2\hbar^4 R(2m_1\Delta)^{1/2}} - \frac{\mathcal{J}_1V_1^2m_1^2\Omega^2}{4\pi^2\hbar^4R^2}\right)\exp\left(-\frac{2R(2m_1\Delta)^{1/2}}{\hbar}\right). \tag{10}$$

It is seen that, depending on the ratio of the parameters, the summary interaction (10) can be of either sign.

We obtain similarly from (5), after a first intergration over the momenta and over the angles of the vector $\mathbf{k}$, an expression for

$$J^{(2)}=-C\int_0^\infty\left(\frac{2V_1V_2}{\Delta+\varepsilon_{2k}}+\frac{2V_1V_2}{\Delta+U}-\mathcal{J}_2\right)^2$$
$$\times\exp\left\{-R\left(\frac{m_2}{m_1}\right)^{1/2}\left(\frac{2m_1B}{\hbar^2}+k^2\right)^{1/2}\right\}k\sin kR\,dk, \qquad (11)$$

where $C=\Omega^2m_2/(2\pi)^3\hbar^2R^2$.

At $V=0$ the integral (11) is equal to[20]

$$J^{(2)}=-\frac{2C\mathcal{J}_2^2Bm_1^{3/2}m_2^{1/2}}{R^4\hbar^3(m_1+m_2)}K_2\left(\frac{R[2B(m_1+m_2)]^{1/2}}{\hbar}\right), \qquad (12)$$

where $K_2(x)$ is a Bessel function of imaginary argument.

An approximate expression for the integral $J^{(2)}$ at $V\neq 0$, when the condition

$$R(2m_1\Delta)^{1/2}/\hbar<R(2m_2B)^{1/2}/\hbar\ll 1$$

is satisfied, is of the form

$$J^{(2)}=-\frac{\Omega^2m_1^{1/2}m_2}{2\pi^2\hbar^2\Delta^{1/2}R}V_2^2V_1^2+\frac{\Omega^2m_1m_2}{\pi^3\hbar^4R^3}\mathcal{J}_2V_1V_2-\frac{\Omega^2m_1^{1/2}m_2^{3/2}}{(m_1+m_2)^{1/2}\hbar^3R^4}\mathcal{J}_2^2. \qquad (13)$$

In the other limiting case

$$R(2m_1\Delta)^{1/2}/\hbar\gg\left[\frac{m_2}{m_1}+2\left(\frac{m_1}{m_2}\right)^{1/2}\left(\frac{2m_1B}{\hbar^2}\right)^{1/2}R\right]^{1/2},$$

neglecting the momentum dependence in the exchange constant in the integral (11), we get

$$J^{(2)}=-\frac{2^{1/2}m_1^{3/4}m_2^{3/4}\Omega^2B^{1/4}}{8(m_1+m_2)^{1/4}\pi^{5/2}\hbar^{7/2}R^{5/2}}\left(\frac{2V_1V_2}{\Delta}+\frac{2V_1V_2}{\Delta+U}-\mathcal{J}_2\right)^2$$
$$\times\exp\left[-\frac{R[2(m_1+m_2)B]^{1/2}}{\hbar}\right]. \qquad (14)$$

In real substances $B\approx 4-6$ eV, $\Delta\approx 1-2$ eV,[2,3], $R[2(m_1+m_2)B]^{1/2}/\hbar>5$, and $R(2m_1\Delta)^{1/2}/\hbar>2$, so that relation (14) is valid for all cases of physical interest. It is seen that the interband interaction is, as in (10), of short-range character. Another approximation can be obtained from (5) if we let the mass m_2 go to infinity after integrating over the states of the valence band and limit to range of integration over the momenta to $k_0=2\pi(4\pi\Omega/3)^{-1/3}$. As a result we obtain for $J^{(2)}$ the expression[4,5,8]

$$J^{(2)}=-\frac{\Omega^2m_2k_0^4\mathcal{J}_2^2}{\pi^3\hbar^2}F(k_0R)\exp\left\{-(2m_1B)^{1/2}\frac{R}{\hbar}\right\}. \qquad (15)$$

Here $F(x)=(x\cos x-\sin x)/x^4$ is the Ruderman-Kittel function.

With increasing lattice parameter, expression (15) can become positive. Xavier and de Graaf attempted to attribute the antiferromagnetic component of the exchange interaction to the same indirect-exchange mechanism. However, the antiferromagnetic interaction in their result[4] is due to the specifics of the assumed approximation. As seen from (12) with $V=0$, the exchange integral $J^{(2)}$ can be calculated exactly and is always negative. We note also that the previously assumed[4-8] approximation is crude, since in the compounds in questions we have $m_1\sim m_2\lesssim m_0$.[21]

The integral $J^{(3)}$ can be calculated simply, and it equals

$$J^{(3)}=-\left(\frac{m_1}{\hbar(2m_1\Delta)^{1/2}}+\frac{2}{(\Delta+U)R}\right)\frac{m_1\Omega V_1^2W}{\pi\hbar^2}\exp\left[-\frac{R(2m_1\Delta)^{1/2}}{\hbar}\right]+\frac{2W^2}{\Delta+U}. \qquad (16)$$

The decrease of the integral $J^{(3)}$ with interatomic distance is slow compared with (10) and (14), because of the absence of the coefficient 2 in the argument of the exponential of (16): at large distances, the exchange integral $J^{(3)}$ becomes dominant.

The integrals (7)–(9) are close in their structure to the corresponding expressions in (4)–(6), and they can be calculated in similar fashion. From (10)–(16) it is seen that at short distances the total exchange integral is ferromagnetic. Using the analytic expressions for the exchange integrals, we can determine the resultant exchange parameters for the interaction with the nearest neighbors I_1, taken at $R=d$ (where d is the distance between the nearest cation atoms), and with next-to-nearest neighbors, I_2, in a lattice of the NaCl type (at $R=d\sqrt{2}$). The qualitative behavior of the exchange parameters I_1 and I_2 is the same as in[16]: at short distances the resultant exchange is ferromagnetic, and at large distances it reverses sign and remains antiferromagnetic.

4. By way of example we consider a family of typical well-investigated magnetic semiconductors—the chalcogenides of europium. It is known that the large variation of the exchange constant in the chalcogenide series (by a factor 6–7 on going from EuO to EuTe) cannot be attributed to only the variation of their lattice parameter R_0,[2,16] so that to calculate the exchange integrals we must have data on the band structure. Table I lists some parameters for europium chalcogenides, obtained in experiments[2,3,14] The data on EuS and EuTe are incomplete, and we use the extrapolated values marked in the table by an asterisk(*).

In the chalcogenides of europium, the wave functions of the anion p_σ join two nearest cations, which are next-to-nearest neighbors to one another, to form an 180-degree configuration. Although the next-to-nearest cations are separated by large distances, the effective transition integral W for them turns out to be of the same order as for the nearest neighbors, owing to the additional effective overlap of the orbitals via the p-band of the chalcogen.[3]

The effective transition integral can be regarded as different from zero for only two nearest coordination spheres, and the parameter is $W\sim V_2^2/(B+U)$ and is approximately equal to $(5-12)\times 10^{-3}$ eV. The parameters $\mathcal{J}_1$ and $V_{1,2}$, as in[2,3], will be assumed to be ~0.3–0.4 eV; the intra-atomic constants $\mathcal{J}_2$ and $\mathcal{J}_3$ are small, $\mathcal{J}_2\sim\mathcal{J}_3\approx 0.1$ eV, and the effective masses are assumed to be of the order of the mass of the free electron.[21] The

TABLE II.

	$I_{1\,\text{theor}}$	$I_{1\,\text{exp}}$	$I_{2\,\text{theor}}$	$I_{2\,\text{exp}}$
EuO	0.64	0.63–0.67	−0.06	−0.07
EuS	0.20	0.20	−0.16	−0.08 (−0.06 ÷ −0.14)
EuSe	0.15	0.14	−0.17	−0.14
EuTe	0.12	0.10	−0.16	−0.21 (−0.12 ÷ −0.17)

best agreement with experiment is obtained at $W = 9.014$ eV and $V_{1,2} = 0.33$ eV.

We note that the effective constant

$$\frac{2V_1V_2}{\Delta} + \frac{2V_1V_2}{\Delta + U} - \mathcal{J}$$

in $J^{(2)}$ decreases on going from EuO to EuTe, in agreement with the available data.[5]

Table II lists the theoretically calculated values of the exchange parameters I_1 and I_2 (in °K) and their experimental values.[22] The data in the parentheses were taken from [2]. Some discrepancy in the case of Eu and EuSe for the exchange constant I_2 is apparently due to the incorrect choice of the parameters B and U (see also[23]). We note that all the mechanisms except $J^{(2)}$ make approximately the same contribution to I_1, whereas in I_2 the principal role is played by the exchange integral $J^{(3)}$. On the other hand, the term representing the Bloembergen-Rowland effective interband interaction makes in all cases a small contribution to I_1 and decreases rapidly on going from EuO to EuTe. By way of example, in EuO (EuSe) contributions to I_1 are made by $J^{(1)}/2 = 0.11$ (0.03), $J^{(2)}/2 = 0.07$, and $J^{(3)}/2 = 0.46$ (0.12), and contributions to I_2 by the values $J^{(3)}/2 = -0.06(-0.17)$.

It is clear from the results that in the general case it is impossible to separate in magnetic semiconductors any particular indirect-exchange mechanism, and it is necessary to include in the analysis of the magnetic properties of these substances the principal mechanisms that take into account the concrete singularities of the electronic structure, and to estimate their relative contributions to the effective exchange interaction.

In our analysis we made a number of simplifying assumptions (a local character of the matrix elements of the hybridization and of the intra-atomic exchange, and a simple form of the spectrum of ϵ_{1k} and ϵ_{2k} in the effective-mass approximation); in addition, in the numerical comparison with experiment it was assumed that in europium chalcogenides, on going from EuO to EuTe, one can assume that the changes of parameters such as $V_{1,2}$, $\mathcal{J}_{1,2,3}$, $m_{1,2}$, and W, are small and that it suffices to take into account the variation of only those parameters which are listed in Table I. None the less, even under these assumptions the simple model considered here gives good numerical agreement between the calculated exchange parameters and their experimental values.

In conclusions, the authors thank D. I. Khomskiĭ for useful discussions and valuable remarks.

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[1] A detailed discussion of this question is found in the book by Methfessel and Mattis, Chaps. 6 and 7 and Appendixes I and II.

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