Random anisotropy of the indirect exchange and nature of the ordering of impurity spins in semiconductors

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A calculation is made of the energy of the indirect exchange interaction of impurity spins in III-V semiconductors; this energy is related to the Anderson hybridization of the impurity and valence-band *d*-electron states, and to the interference of the hybridization with the *s–d* exchange. The dependences of the exchange integrals on the distance and on the masses of the light and heavy holes are determined. The spin–orbit interaction in the valence band results in a strong anisotropy of the indirect exchange energy relative to a straight line joining a pair of interacting impurities. At large distances the exchange of the transverse components of the spin is four times stronger than the exchange of the longitudinal components. Since the impurities are distributed at random, the alloys formed by such impurities are magnetic materials with a random exchange anisotropy. Therefore, the spins should become ordered in a spin glass state irrespective of the sign of the exchange integral.

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INTRODUCTION

Semiconductors with magnetic impurities are a class of impurity magnetic materials which are just beginning to attract interest.

The indirect exchange interaction of localized spins has been investigated in the case of zero-gap semiconductors[1] and semiconductors with a finite band gap.[2,3] A theory of spin glass states in semiconductors with a many-valley electron spectrum is developed in Ref. 2. All these investigations are concerned with the indirect interaction of localized spins due to the mechanism of Bloembergen and Rowland.[4] The Bloembergen–Rowland exchange interaction involves virtual transitions of electrons from the valence to the conduction band due to the exchange interaction of the band electrons and those localized at impurities:

$$\hat{I}=-\frac{1}{2}\sum_j I(\mathbf{r}-\mathbf{R}_j)(\boldsymbol{\sigma}\mathbf{S}_j), \tag{1}$$

where $\mathbf{S}_j$ is a spin localized at a lattice site $\mathbf{R}_j$; $\frac{1}{2}\sigma$ is the electron spin operator. In the case of simple spherical energy bands of electrons and holes this mechanism leads[2,3] to the Heisenberg exchange Hamiltonian, and at large distances R the exchange interaction energy decreases on increase in R in accordance with the law

$$J(R)\sim R^{-5/2}e^{-k_0R}, \qquad k_0=[2(m_c+m_v)\varepsilon_g]^{1/2}, \tag{2}$$

where ε_g is the band gap, and m_c and m_v are the masses of electrons and holes, respectively.1)

The indirect exchange in semiconductors need not be due to the Bloembergen–Rowland mechanism, but also due to mechanisms associated with the Anderson hybridization of the d electrons of the impurities with the band electrons.[5,6] These mechanisms involve virtual transitions of electrons (holes) from a local level to a band, so that we shall call them the level–band (LB) mechanisms. These LB mechanisms have been used[5,6] to explain exchange in ordered rare-earth semiconductors. However, the results obtained apply equally to semiconductors with magnetic impurities.

According to Ref. 6, interference of the s–d interaction of Eq. (1) and of the hybridization considered in the third-order perturbation theory in the case of simple energy bands gives rise to a ferromagnetic exchange which decreases with distance in accordance with the law

$$J(R)\sim R^{-2}e^{-2k_cR}, \qquad k_c=(2m_c\Delta)^{1/2}, \tag{3}$$

where Δ is the energy of a localized level measured from the bottom of the conduction band.

In the fourth-order perturbation theory the indirect exchange is solely due to the hybridization mechanism,[5,6] which has the antiferromagnetic sign and it decreases with R somewhat more slowly than given by Eq. (3):

$$J(R)\sim -R^{-1}e^{-2k_cR}. \tag{4}$$

Magnetic impurities and their exchange interaction in III–V semiconductors are of topical interest.[7-9] However, the formulas (2)–(4) cannot be applied directly to the semiconductors because the band states in these materials cannot be described generally by plain waves. In particular, the strong spin–orbit interaction in the valence band may give rise to an anisotropic Hamiltonian of the indirect exchange via this band.

We shall find the energy of the exchange interaction of impurity spins in III-V semiconductors due to the LB mechanisms. We shall consider one of the most thoroughly investigated magnetic centers in III-V semiconductors, which are iron impurities, and we shall assume that the d shell of the impurity is half-filled (^6S state, total spin $\frac{5}{2}$).

We may expect that in the case of III-V semiconductors the contribution of the interband mechanism to the exchange is less than that of the LB mechanism, because it follows from the symmetry considerations that the matrix element of the energy $I(r)$ between the functions of the bottom of the valence band and of the conduction band vanishes.[10] Moreover, it is clear from Eqs. (2)–(4) that the interband exchange decreases more rapidly with the distance than the LB exchange.

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For these reasons the interband exchange will be ignored. We shall allow only for the hybridization of the impurity d electrons with the valence band because, firstly, the levels of magnetic centers in III-V semiconductors are usually located closer to the valence than to the conduction band and, secondly, the matrix elements of the energy of the hybridization between the d states of the impurity and the states at the bottom of the conduction band vanish.

As expected, the exchange interaction Hamiltonian is anisotropic. The exchange energy depends in a complex manner on the distance. At large distances the main contribution to the exchange comes from light holes and a reduction in the exchange on increase in R occurs in accordance with laws such as those given by Eqs. (3) and (4) with a characteristic radius $\sim(2\varepsilon_d m_l)^{-1/2}$, where m_l is the mass of the light holes and ε_d is the energy required to transfer an electron from the valence band to a d level. The sign of the exchange energy due to each of the two LB mechanisms is the same as in the isotropic case, so that the interaction is antiferromagnetic in the limit $R\to\infty$.

The exchange anisotropy depends on the distance. As a rule, the exchange of the transverse (relative to a straight line joining a pair of impurities) spin components is several times greater than the exchange of the longitudinal components. Since a straight line joining a pair of impurities is oriented at random in space, it follows that III-V semiconductors with magnetic impurities are materials with a random exchange anisotropy. Therefore, even if over the average distance the ferromagnetic exchange exceeds the antiferromagnetic mechanism, such impurity magnetic materials should not go over to a ferromagnetic state at low temperatures but to a spin glass state.

1. WAVE FUNCTIONS AND THE INTERACTION BETWEEN BAND AND LOCALIZED ELECTRONS

The valence band of III-V compounds is quadruply degenerate at the point $k=0$. The wave functions of holes $|\mathbf{k}v\gamma\rangle$ are characterized by three quantum numbers: $\mathbf{k}$ is the wave vector; v distinguishes the light and heavy holes and assumes correspondingly the values l and h; γ distinguishes the states transforming into one another as a result of time reversal, and they assume the values plus and minus (Fig. 1). It is convenient to write down the wave functions in the $|p\rangle\chi(\lambda)$ representation where $|p\rangle=X,Y,Z$ is the coordinate part of the wave function in the Kane notation[11] and $\chi(\lambda)$ is the spin part of the wave function. According to Ref. 12 (Chap. 4),

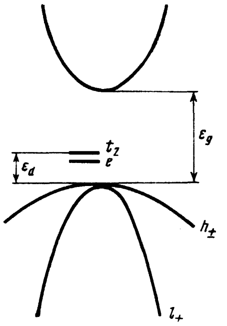

FIG. 1. Energy band model of a III-V semiconductor with a localized impurity state (d^6 configuration, ^5D state).

$$|\mathbf{k}v\gamma\rangle=e^{i\mathbf{k}\mathbf{R}}\sum_{p\lambda}\Gamma^{\lambda}_{vp\gamma}(\theta,\varphi)\,|p\rangle\chi(\lambda),\tag{5}$$

where the angles θ and φ represent the direction of the wave vector $\mathbf{k}$ and the functions $\Gamma^{\lambda}_{vp\gamma}(\theta,\varphi)$ have the following form:

$$\Gamma^{\uparrow}_{lp+}=\frac{1}{2\sqrt{6}}\sin\theta e^{-i\varphi}\begin{pmatrix}3+e^{2i\varphi}\\ 3i-ie^{2i\varphi}\\ 4\operatorname{ctg}\theta\end{pmatrix},\quad \Gamma^{\uparrow}_{lp-}=\frac{1}{\sqrt{6}}\cos\theta e^{-i\varphi}\begin{pmatrix}i\\ 1\\ -i\operatorname{tg}\theta e^{-i\varphi}\end{pmatrix},$$

$$\Gamma^{\uparrow}_{hp+}=\frac{1}{\sqrt{2}}\cos\theta e^{-3i\varphi}\begin{pmatrix}i\\ -1\\ -i\operatorname{tg}\theta e^{i\varphi}\end{pmatrix},\quad \Gamma^{\uparrow}_{hp-}=\frac{1}{2\sqrt{2}}\sin\theta e^{-i\varphi}\begin{pmatrix}1-e^{-2i\varphi}\\ -i-ie^{-2i\varphi}\\ 0\end{pmatrix},$$

$$\Gamma^{\downarrow}_{lp\mp}=\pm i\bar{\Gamma}^{\uparrow}_{lp\pm}e^{-i\varphi},\quad \Gamma^{\downarrow}_{hp\mp}=\mp i\bar{\Gamma}^{\uparrow}_{hp\pm}e^{-3i\varphi}.\tag{6}$$

The bar above the function Γ represent complex conjugacy.

We shall assume that the energy spectrum of light and heavy holes is parabolic:

$$\varepsilon_{\mathbf{k}v}=k^2/2m_v.\tag{7}$$

We shall discuss below the validity of this approximation in our case.

The results of optical experiments demonstrate that, in the zeroth approximation, the wave functions of magnetic-impurity electrons are the d atomic functions.[7] The energy ε_d required to transfer an electron from the valence band to an impurity level generally depends on the symmetry of the d states and is usually less than half the band gap. On the other hand, the experimental data on the g factor of localized electrons deduced from nuclear resonance indicate that the d states include a considerable admixture of the band states.[7] For reasons mentioned in the Introduction, we shall allow for the hybridization of the d states only with the valence electron states.

The total Hamiltonian of the system can therefore be regarded as the sum of three terms[2)]:

$$\hat{H}=\hat{H}_0+\hat{I}+\hat{V}.\tag{8}$$

Here,

$$\hat{H}_0=\sum_{\mathbf{k}v\gamma}\varepsilon_{\mathbf{k}v}a^{+}_{\mathbf{k}v\gamma}a_{\mathbf{k}v\gamma}+\sum_{im\lambda}(-\varepsilon_{dm})\,b^{+}_{im\lambda}b_{im\lambda},\tag{9}$$

where $a^{+}_{\mathbf{k}v\gamma}$ ($a_{\mathbf{k}v\gamma}$) are the creation (annihilation) operators of a hole in a valence band state $|\mathbf{k}v\gamma\rangle$; $b^{+}_{im\lambda}$ ($b_{im\lambda}$) are the creation (annihilation) operators of a hole in the d shell of an impurity which is in a state characterized by a magnetic quantum number m and a spin λ. The energy is measured from the top of the valence band in the downward direction.

The operator representing the exchange interaction between holes in the valence band and localized spins $\mathbf{S}_i$ is

$$\hat{I}=-\frac{I}{2}\sum_{i}\sum_{\substack{\mathbf{k}v\gamma\\ \mathbf{k}'v'\gamma'}}\sum_{p\lambda\lambda'}\exp\{i(\mathbf{k}'-\mathbf{k})\mathbf{R}_i\}\Gamma^{\lambda}_{vp\gamma}(\mathbf{k})\Gamma^{\lambda'}_{vp\gamma}(\mathbf{k}')\,(\boldsymbol{\sigma}_{\lambda\lambda'}\mathbf{S}_i)\,a^{+}_{\mathbf{k}v\gamma}a_{\mathbf{k}v\gamma},\tag{10}$$

where σ are the Pauli matrices, and I is the diagonal matrix element of the energy $I(r)$ between the wave functions of holes at the bottom of the band:

$$I=\langle p|I($$ [illegible]

It follows from symmetry considerations that all the diagonal elements are the same and the nondiagonal elements vanish.

Finally, the hybridization operator is

$$\hat{V}=\sum_i\sum_{\mathbf{k}v\gamma}\sum_{pm\lambda}\exp(-i\mathbf{k}\mathbf{R}_i)\Gamma^{\lambda}_{vp\gamma}(\mathbf{k})\langle p|V(r)|m\rangle a_{\mathbf{k}v\gamma}b_{im\lambda}+\text{H.c.} \tag{11}$$

Here, $|m\rangle$ is the coordinate part of the wave function of the d electrons. The expression (11) can be simplified bearing in mind that in a field of cubic symmetry the d shell splits into two subshells: t_2 and e. The wave functions of the t_2 states are transformed by the symmetry operations of the tetrahedron group in the same way as the $|p\rangle$ functions of the valence band, whereas the functions of the e states belong to a different irreducible representation. Therefore,

$$\langle p|V(r)|m\rangle=\langle p|V(r)|p\rangle\delta_{pm}=\tilde{V}\delta_{pm}, \tag{12}$$

if $m\in t_2$ and

$$\langle p|V(r)|m\rangle=0, \tag{13}$$

if $m\in e$. Substituting Eqs. (12) and (13) in Eq. (11), we obtain

$$\hat{V}=\tilde{V}\sum_i\sum_{\mathbf{k}v\gamma}\sum_{pm\lambda}\exp(-i\mathbf{k}\mathbf{R}_i)\Gamma^{\lambda}_{vp\gamma}(\mathbf{k})a_{\mathbf{k}\gamma v}b_{im\lambda}\delta_{pm}+\text{H.c.} \tag{14}$$

2. INDIRECT EXCHANGE ENERGY

The interactions (10) and (12) result in the interband exchange and in the LB exchange mechanisms. Among the latter there are also processes which in the fourth order of perturbation theory with respect to $\hat{V}$ result finally in renormalization of the energy I in the interband processes.[5] Since the electron states are of the S type and the matrix elements $\langle p|I|S\rangle$ and $\langle m|V|S\rangle$ vanish, the interband mechanism is ineffective. Therefore, we shall allow only for those processes in which the conduction band states do not participate.

In the third order of perturbation theory (in the first order with respect to $\hat{I}$ and in the second order in $\hat{V}$) the averaging over the valence band operators yields the following expression for the indirect exchange Hamiltonian:

$$H_{IV}(\mathbf{R}_{ij})=\frac{I\tilde{V}^2}{2}\sum_{\mathbf{k}\mathbf{k}_1vv_1}\sum_{\gamma\gamma_1pp_1m}\sum_{\lambda\lambda'\lambda_1\lambda_1'}\frac{\exp[i(\mathbf{k}-\mathbf{k}_1)\mathbf{R}_{ij}]}{(\varepsilon_{kv}+\varepsilon_d)(\varepsilon_{k_1v_1}+\varepsilon_d)}$$
$$\times\Gamma^{\lambda_1}_{v_1p\gamma_1}(\mathbf{k}_1)\Gamma^{\lambda}_{v_1p_1\gamma_1}(\mathbf{k}_1)\Gamma^{\lambda'}_{vp_1\gamma}(\mathbf{k})\Gamma^{\lambda_1'}_{vp\gamma}(\mathbf{k})(\sigma_{\lambda\lambda'}\cdot\mathbf{S}_i)(\sigma_{\lambda_1\lambda_1'}\cdot\mathbf{s}_{jm})\delta_{mp}. \tag{15}$$

Here, $\mathbf{s}_{jm}$ is the spin operator of a single particle (hole) at a level m related to the operators $b^{+}_{jm\lambda}$, $b_{jm\lambda}$ by the usual expressions

$$b^{+}_{jm\uparrow}b_{jm\uparrow}={}^1\!/_2+s_{jm}{}^z,\quad b^{+}_{jm\downarrow}b_{jm\downarrow}={}^1\!/_2-s_{jm}{}^z,$$
$$b^{+}_{jm\uparrow}b_{jm\downarrow}=s_{jm}{}^{+},\quad b^{+}_{jm\downarrow}b_{jm\uparrow}=s_{jm}{}^{-}.$$

In the derivation of Eq. (14) we have considered only the $d^5\rightleftarrows d^6$ transitions. We shall now allow for the fact that the exchange between the spins $\mathbf{s}_{jm}$ belonging to one impurity is much stronger than the exchange between spins belonging to different impurities. To within terms of higher orders with respect to the interimpurity exchange, we find that the intraatomic Hamiltonian (15) can only be due to processes in which the total impurity spin is conserved and only the spin projection changes. This means that the operators $\mathbf{s}_{jm}$ in Eq. (15) can be replaced with

$$(2S)^{-1}\sum_m\mathbf{s}_{jm}=(2S)^{-1}\mathbf{S}_j.$$

This last conclusion can easily be proved using the relationship[13]

$$\langle SM|\mathbf{s}m|SM'\rangle=\frac{1}{S(S+1)}\langle SM|\mathbf{S}|SM'\rangle\langle SM'|\mathbf{S}\mathbf{s}_m|SM'\rangle,$$

where M is the projection of the total spin. Consequently, the Hamiltonian (15) transforms to

$$H_{IV}(R_{ij})=-\sum_{\alpha\beta}J_1{}^{\alpha\beta}(\mathbf{R}_{ij})\mathbf{S}_i{}^{\alpha}\mathbf{S}_j{}^{\beta}. \tag{16}$$

The exchange energy is

$$J_1{}^{\alpha\beta}(\mathbf{R}_{ij})=\frac{IV^2}{2}\sum_{pp_1vv_1}\sum_{\lambda\lambda'\lambda_1\lambda_1'}\sigma^{\alpha}_{\lambda\lambda'}\sigma^{\beta}_{\lambda_1\lambda_1'}A^{\lambda_1\lambda'}_{pp_1v_1}\bar{A}^{\lambda_1'\lambda}_{pp_1v_1} \tag{17}$$

where the tensor is defined by

$$A^{\lambda\lambda_1}_{pp_1v}(\mathbf{R}_{ij})=\sum_{\mathbf{k}\gamma}\frac{\exp(i\mathbf{k}\mathbf{R}_{ij})}{\varepsilon_{kv}+\varepsilon}\Gamma^{\lambda}_{vp\gamma}(\mathbf{k})\Gamma^{\lambda_1}_{vp_1\gamma}(\mathbf{k}), \tag{18}$$

whereas the constant is $V=(2S)^{-1/2}\tilde{V};\alpha,\beta=x,y,z$.

Similarly, the indirect exchange energy $H_V(\mathbf{R}_{ij})$ originating entirely from the hybridization can be calculated in the fourth order of perturbation theory. The corresponding exchange integral $J_2^{\alpha\beta}(\mathbf{R}_{ij})$ is

$$J_2{}^{\alpha\beta}(\mathbf{R}_{ij})=-V^4\sum_{pp_1vv_1}\sum_{\lambda\lambda'\lambda_1\lambda_1'}A^{\lambda_1\lambda'}_{pp_1v_1}\bar{B}^{\lambda\lambda_1'}_{pp_1v}\sigma^{\alpha}_{\lambda\lambda'}\sigma^{\beta}_{\lambda_1\lambda_1'}, \tag{19}$$

where the tensor is

$$B^{\lambda\lambda_1}_{pp_1v}(\mathbf{R}_{ij})=\sum_{\mathbf{k}\gamma}\frac{\exp(i\mathbf{k}\mathbf{R}_{ij})}{(\varepsilon_{kv}+\varepsilon_d)^2}\bar{\Gamma}^{\lambda}_{vp\gamma}(\mathbf{k})\Gamma^{\lambda_1}_{vp_1\gamma}(\mathbf{k}). \tag{20}$$

In the adopted approximation, when the spectrum of holes is regarded as spherically symmetric, the vector $\mathbf{R}_{ij}$ is the only special direction in Eqs. (17)–(20). Therefore, in a coordinate system in which the z axis is directed along $\mathbf{R}_{ij}$, all the nondiagonal components of the tensors $J_1^{\alpha\beta}$ and $J_2^{\alpha\beta}$ vanish and we have $J_1^{xx}=J_1^{yy}$ and $J_2^{xx}=J_2^{yy}$.

Using the explicit expressions of Eq. (6) for the Γ functions, we obtain

$$J_{1\parallel}(R)\equiv J_1{}^{zz}(R)=4IV^2\left(\left|\sum_v A^{\uparrow\uparrow}_{xxv}\right|^2+\left|\sum_v A^{\uparrow\uparrow}_{xyv}\right|^2\right),$$
$$J_{1\perp}(R)\equiv J_1{}^{xx}(R)=4IV^2\left(\left|\sum_v A^{\uparrow\uparrow}_{xxv}\right|^2+\frac{1}{2}\left|\sum_v A^{\uparrow\uparrow}_{zzv}\right|^2-\left|\sum_v A^{\uparrow\uparrow}_{xyv}\right|^2\right); \tag{21}$$

$$J_{2\parallel}(R)\equiv J_2{}^{zz}(R)=4V^4\left(\sum_{vv'}A^{\uparrow\uparrow}_{xxv}B^{\uparrow\uparrow}_{xxv'}+\sum_{vv'}A^{\uparrow\uparrow}_{xyv}\bar{B}^{\uparrow\uparrow}_{xyv'}\right),$$
$$J_{2\perp}(R)\equiv J_2{}^{xx}(R)=-4V^4\left(\sum_{vv'}A^{\uparrow\uparrow}_{xxv}B^{\uparrow\uparrow}_{xxv'}+\frac{1}{2}\sum_{vv'}A^{\uparrow\uparrow}_{zzv}B^{\uparrow\uparrow}_{zzv'}-\sum_{vv'}A^{\uparrow\uparrow}_{xyv}\bar{B}^{\uparrow\uparrow}_{xyv'}\right). \tag{22}$$

Here,

$$\begin{pmatrix}A^{\uparrow\uparrow}_{xxl}\\A^{\uparrow\uparrow}_{xyl}\\A^{\uparrow\uparrow}_{zzl}\end{pmatrix}=\frac{\Omega}{(2\pi)^3}\int d^3k\ \frac{e^{ikR\cos\theta}}{\varepsilon_{kl}+\varepsilon_d}\begin{pmatrix}{}^5\!/_{12}-{}^1\!/_4\cos^2\theta\\ \dfrac{i}{3}-\dfrac{i}{2}\cos^2\theta\\ {}^1\!/_6+{}^1\!/_2\cos^2\theta\end{pmatrix}, \tag{23}$$

$$\begin{pmatrix} A^{\uparrow\uparrow}_{xxh} \\ A^{\uparrow\uparrow}_{xyh} \\ A^{\uparrow\uparrow}_{zzh} \end{pmatrix} = \frac{\Omega}{(2\pi)^3} \int d^3k \, \frac{e^{ikR\cos\theta}}{\varepsilon_{kh}+\varepsilon_d} \begin{pmatrix} {}^1/_4 + {}^1/_4 \cos^2\theta \\ \frac{i}{2} \cos^2\theta \\ {}^1/_2 - {}^1/_2 \cos^2\theta \end{pmatrix}, \tag{24}$$

and Ω is the volume of a unit cell.

The expression for the components of the tensor $B^{\downarrow\downarrow}_{\alpha\beta v}$ differ from the corresponding components of the tensor $A^{\downarrow\downarrow}_{\alpha\beta v}$ by the replacement of the denominator $\varepsilon_{kv}+\varepsilon_\alpha$ with $(\varepsilon_{kv}+\varepsilon_d)^2$ in the integrals in Eqs. (23) and (24). The integrals in Eqs. (23) and (24) and the corresponding integrals for $B^{\downarrow\downarrow}_{\alpha\beta v}$ can easily be calculated. It is then found that each component of $A^{\downarrow\downarrow}_{\alpha\beta v}$ or $B^{\downarrow\downarrow}_{\alpha\beta v}$ contains not only terms which decrease exponentially on increase in R, but also terms which decrease as a power law and are independent of the hole mass. They are due to terms containing $\cos^2\theta$ in the integrands of Eqs. (23) and (24), and since these terms appear in Eqs. (23) and (24) with identical coefficients but with different signs, then in the case of summation with respect to v in Eqs. (21) and (22) the power terms cancel out exactly, as expected.

We finally obtain

$$J_{1\parallel,\perp}(R) = \frac{\Omega^2 I V^2}{4\pi^2 R^2}[m_l \exp(-k_l R) + m_h \exp(-k_h R)]^2 \Phi_{1\parallel,\perp}(R),$$
$$J_{2\parallel,\perp}(R) = -\frac{\Omega^2 V^4}{4\pi^2 (2\varepsilon_d)^{1/2} R}[m_l^{5/4} \exp(-k_l R) + m_h^{5/4} \exp(-k_h R)]^2 \Phi_{2\parallel,\perp}(R). \tag{25}$$

Here,

$$k_l = (2m_l\varepsilon_d)^{1/2}, \quad k_h = (2m_h\varepsilon_d)^{1/2},$$

$$\Phi_{1\parallel} = [1+\tau^2 \exp(-(\tau-1)k_l R)]^{-2}[{}^2/_9 + 2\tau^4 \exp(-2(\tau-1)k_l R)$$
$$+5F^2 + F({}^2/_3 - 6\tau^2 \exp(-(\tau-1)k_l R))],$$

$$\Phi_{1\perp} = [1+\tau^2 \exp(-(\tau-1)k_l R)]^{-2}[{}^8/_9 + {}^4/_3 \tau^2 \exp(-(\tau-1)k_l R)$$
$$+2F({}^1/_3 + \tau^2 \exp(-(\tau-1)k_l R)) - F^2],$$

$$\Phi_{2\parallel} = [1+\tau^{5/2} \exp(-(\tau-1)k_l R)]^{-2}\Big\{{}^2/_9 + 2\tau^5 \exp(-2(\tau-1)k_l R)$$
$$+\frac{1}{3k_l R}[1-\tau^2 \exp(-(\tau-1)k_l R) + 2F][1-9\tau^2 \exp(-(\tau-1)k_l R) + 15F]$$
$$+{}^1/_3 F[1-9\tau^3 \exp(-(\tau-1)k_l R)]\Big\},$$

$$\Phi_{2\perp} = [1+\tau^{5/2} \exp(-(\tau-1)k_l R)]^{-2}\{{}^8/_9 + {}^2/_3(1+\tau)\tau^2 \exp(-(\tau-1)k_l R)$$
$$+{}^1/_3[1+3\tau^3 \exp(-(\tau-1)k_l R)]F$$
$$+(3k_l R)^{-1}[1-\tau^2 \exp(-(\tau-1)k_l R) + 2F][1+3\tau^2 \exp(-(\tau-1)k_l R) - 3F]\},$$

where

$$F = \frac{1}{k_l^2 R^2}[1 + k_l R - (1+\tau k_l R)\exp(-k_l R(\tau-1))], \quad \tau = \left(\frac{m_h}{m_l}\right)^{1/2}.$$

3. INDIRECT EXCHANGE ANISOTROPY

The functions $\Phi_1^{\alpha\alpha}(R)$ and $\Phi_2^{\alpha\alpha}(R)$ vary relatively slowly with the distance and are positive, i.e., exactly as in the case of simple bands the interaction $J_1(R)$ is ferromagnetic, whereas $J_2(R)$ is antiferromagnetic. At long distances such that $k_l R \gg 1$ and $(\tau-1)k_l R \gg 1$, the exchange interaction is governed by the parameters of the light holes.[3)]

It is then found that $\Phi_{1\parallel}(R) = \Phi_{2\parallel}(R) = \frac{2}{9}$ and $\Phi_{1\perp}(R) = \Phi_{2\perp}(R) = \frac{8}{9}$, and since $k_l < k_h$, it follows that the exchange integrals $J_{1\parallel,\perp}(R)$ and $J_{2\parallel,\perp}(R)$ decrease on increase in the distance as $R^{-2}\exp(-2k_l R)$ and $R^{-1} \times \exp(-2k_l R)$, respectively, and the ratios of these integrals are

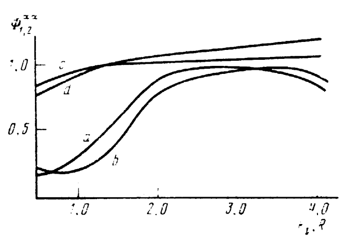

FIG. 2. Dependences of the functions $\Phi^{\alpha\alpha}_{1,2}$ on the distance: a) $\Phi_{1\parallel}$; b) $\Phi_{2\parallel}$; c) $\Phi_{1\perp}$; d) $\Phi_{2\perp}$.

$$J_{1\perp}/J_{1\parallel} = J_{2\perp}/J_{2\parallel} = 4.$$

We can thus see that in the asymptotic region the exchange interaction anisotropy is strong and spins tend to become oriented in a plane perpendicular to a straight line joining the two impurity atoms in a pair. The anisotropy disappears if $m_l = m_h$. We then have $\Phi_{1\parallel,\perp}(R) = \Phi_{2\parallel,\perp} = \frac{5}{9}$. Since in the asymptotic region the value of $J_2(R)$ decreases with distance more slowly than does $J_1(R)$, it follows that at sufficiently large distances the indirect exchange interaction is always antiferromagnetic. If the distances are not too great, the exchange anisotropy depends in a complex manner on the distance.

Figure 2 shows the dependences on R of the functions $\Phi_{1\parallel,\perp}$ and $\Phi_{2\parallel,\perp}$, which determine the anisotropy in the case of iron impurities in gallium arsenide. The curves shown in Fig. 2 were calculated assuming the following parameters: $m_l = 0.12$ and $m_h = 0.68$ in units of the free electron mass (see Ref. 12, p. 572); $\varepsilon_d = 0.5$ eV (Ref. 7); $k = 0.13\times10^8$ cm^{-1} and $k_h = 0.30\times10^8$ cm^{-1}. We can see that compared with the factors decreasing rapidly on increase in R and standing in front of $\Phi_1(R)$ and $\Phi_2(R)$ in Eq. (25), the functions mentioned above vary slowly so that in the first approximation the exchange integrals decrease as follows with the distance:

$$J_{1\parallel,\perp}(R) \sim R^{-2}[m_l \exp(-k_l R) + m_h \exp(-k_h R)]^2,$$
$$J_{2\parallel,\perp}(R) \sim -R^{-1}[m_l^{5/4} \exp(-k_l R) + m_h^{5/4} \exp(-k_h R)]^2.$$

In the case of the transverse exchange integrals these dependences are practically exact (Fig. 2).

In view of the high density of states of the heavy holes, their contribution to the exchange predominates over that of the light holes when distances are short so that $R \gtrsim k_h^{-1}$. However, if $k_h > k_l$, then the contribution of the light holes becomes more important as R increases.

The functions $\Phi_{1\parallel}$ and $\Phi_{2\parallel}$ vary with the distance R much more rapidly than do the functions $\Phi_{1\perp}$ and $\Phi_{2\perp}$ so that the exchange interaction anisotropy depends on R, but for the parameters given above we have $\Phi^{\perp}_{1,2} > \Phi^{\parallel}_{1,2}$ for all the values of R, i.e., the spins tend to become oriented in a plane perpendicular to the straight line joining a pair of impurities and this is true irrespective of the distance between them.

Since the impurities are distributed at random, it follows that the easy plane for each impurity pair is also oriented at random. Therefore, even when the ferromagnetic exchange $J_1(R)$ is stronger over the average distance than the antiferromagnetic exchange $J_2(R)$ (according to the experimental results of Ref. 8, this is true of iron dissolved in gallium arsenide), at low temperatures an alloy formed by such impurities in a III-V semiconductor should go over not to a ferromagnetic state but to a spin glass state.

The problem of the influence of the random orientation of the single-ion anisotropy axes on the nature of spin ordering is currently attracting considerable interest.[14-19] Important arguments have been put forward in support of the conclusion that in the case of a random orientation of the axes even a weak anisotropy can destroy a ferromagnetic state.

Our results show that III-V semiconductors with magnetic impurities are a new class of materials: they are magnetic substances with a random exchange anisotropy. A phase transition in such a substance is considered in Ref. 20 using a special model in which the number of the spin components goes to the limit $n\to\infty$.

It is impossible to calculate the exchange interaction accurately because we do not know the constants I and V. We shall assume that I has the value 1.5 eV obtained in Ref. 21 for the Mn impurities in HgTe. If we assume that the matrix element is $\tilde{V}\approx I$, it then follows that $V=\tilde{V}/\sqrt{2S}=0.7$ eV. At distances $R=k_l^{-1}=7.7$ Å the exchange integrals are then $J_{1\perp}\approx 20$ °K, $J_{1\parallel}\approx 7$ °K, $J_{2\perp}\approx -10$ °K, $J_{2\parallel}\approx -2$ °K.

We shall conclude by considering the validity of our approximations.

The indirect exchange interaction was calculated using perturbation theory. Since the potentials $I(r)$ and $V(r)$ are of the short-range type (their radius is of the order of the distance between the nearest neighbors a_0), it follows that the perturbation theory parameters[22] are $Ima_0^2/\hbar^2 \sim I/E \ll 1$ and $Vma_0^2/\hbar^2 \sim V/E \ll 1$, where E is of the order of the valence band width. These estimates are obtained quite easily by analyzing directly the integrals (23) and (24), for $R\sim a_0$.

At distances $R\gtrsim k_l^{-1}$ for the light holes and $R\gtrsim k_h^{-1}$ for the heavy holes the main contribution to the integrals (23) and (24) is given by the energies in the range $\varepsilon_{kv}\sim\varepsilon_d<\varepsilon_g/2$. This justifies the use of the parabolic spectrum of holes given by Eq. (7).

We assume that the spectrum of holes is spherically symmetric. This assumption is fully justified for the light holes, but the anisotropy of the heavy-hole mass is generally not weak. Therefore, at distances at which the contribution of the heavy holes to the exchange energy is still important we can expect an additional exchange anisotropy related to the crystallographic axes. Since the relative change in the heavy-hole mass with direction in III-V semiconductors is less than unity (in the case of gallium arsenide it amounts to 25–30%), it follows that the random exchange anisotropy is usually stronger than the magnetocrystalline anisotropy and allowance for the latter does not give rise to any qualitatively new phenomena.

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1) Here and later we shall assume that $\hbar=1$.

2) The strong intraatomic interaction of electrons is not used explicitly.

3) This does not mean that in the calculation of the exchange integrals we should not allow for the effect of the heavy holes at large distances. As pointed out in the preceding section, only the combined contribution of the light and heavy holes can ensure an exponential decrease in the exchange energy on increase in R.

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