Effect of giant spin splitting of bands on the optical polarization and relaxation of localized moments in semiconductors

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The effective exchange field produced by localized magnetic moments is taken into account in processes of exchange scattering of photocarriers by localized magnetic moments. The exchange field leads to giant spin splitting of the energy bands in magnetically doped semiconductors. Expressions for the probabilities of spin relaxations are obtained. The effect of exchange scattering on the spin relaxation and on the spin polarization of impurity centers under conditions of giant band splitting is studied theoretically. Situations are considered in which spin heating of the photocarriers or selective interband irradiation polarize the spins of the impurity centers irrespective of the intensity of the magnetic field. The conditions required for realization of maximum spin polarization are determined. Numerical estimates are presented for $A^{II}B^{VI}$:Mn.

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1. INTRODUCTION

From among the numerous manifestations of carrier–impurity exchange interaction in magnetically doped semiconductors, the exchange scattering of carriers by impurity centers (IC) and the spin splitting of the carrier bands by the exchange field of polarized IC are the most intensively studied. The first of these effects was revealed in experiment by the shortening of the paramagnetic-relaxation time,[1,2] and the second by the giant spin splitting of the excition optical spectra.[3–5] Even though the exchange scattering and the giant spin splitting are due respectively to the off-diagonal and diagonal parts of one and the same carrier–impurity exchange interaction, these phenomena were studied independently of each other. Such an approach was justified in the first case[1,2] because the IC density was insufficient to produce an effective exchange fields stronger than the acting magnetic field, and in the second case[3–5] it was justified by the low photocarrier density, insufficient for monitoring the IC polarization. Yet the indicated conditions are not satisfied in the case of sufficiently intense pumping of the photocarriers and sufficiently high density of the IC, when the relaxation cannot be considered independently of polarization and vice versa.

This paper deals with effects due to the joint manifestation of the diagonal at off-diagonal parts of the carrier–impurity exchange interaction, namely, the spin splitting of the bands and exchange scattering of the carriers.

2. PROBABILITY OF EXCHANGE SCATTERING OF CARRIERS BY IC

We consider a semiconductor with a cubic lattice, doped with paramagnetic centers. At sufficiently high density of the latter, the carriers behave as if their spins were acted upon by an effective magnetic field $\mathbf{G}/g\beta$ due to exchange interaction with N impurity centers[3–5] polarized by an external magnetic field $\mathbf{H}$. As a result, the spectrum of the carriers, distinguishable by their different projections p of the total angular momentum $\mathbf{J}$ ($J=\frac{1}{2}$ for the electron and hole bands Γ_6 and Γ_7, $J=\frac{3}{2}$ for the hole band Γ_8) and different wave vectors $\mathbf{k}$, takes in the important case of strong exchange fields and low temperatures,

$$g\beta H, \quad k_BT \ll G, \tag{1}$$

near the band extremum the form (S is the spin of the IC)

$$E_{pk}=\varepsilon_{pk}+(\mathbf{GJ})_{pp}, \quad \mathbf{G}=-\frac{2NI}{V_0}\langle \mathbf{S}\rangle, \tag{2}$$

where I is the exchange constant and has the sign of the ferromagnetism ($I_e>0$) for the bands Γ_6 and Γ_7 and of the antiferromagnetism ($I_h<0$) for the Γ_8 band,[6] while V_0 is the volume of the crystal. The averaging over the ensemble in (2) takes into account both the actual form of the spin Hamiltonian and the pair interactions of the IC spins with one another; ε_{pk} describes the quadratic dispersion law near the extremum of the nondegenerate bands.[1)] According to (1) and (2) the quantization axis of the spins of the electrons (holes) is $e=-\langle \mathbf{S}\rangle/|\langle S\rangle|$. It is convenient to choose this direction to be the Z axis of the coordinate frame, since $G_Z>0$ corresponds to ferromagnetic and $G_Z<0$ to antiferromagnetic carrier–impurity exchange interaction.

We assume that the energy of N spins in the external magnetic field $\mathbf{H}$, in the carrier polarization field $\mathbf{h}$ (which will be defined below), and in the crystal field exceeds the energy of the spin-spin interactions of the IC. To determine the spin density matrix of the IC it suffices then to use a single-particle kinetic equation.[8] The equations for the diagonal components of the density matrix in the case of a nondegenerate IC spectrum are equivalent to the fundamental kinetic equation and contain as the kinetic coefficients the probabilities of the relaxation transitions between the IC spin states M and M' (with energies E_M and $E_{M'}$):

$$W_{MM'}=\frac{2\pi I^2}{\hbar V_0^2}\sum_{pp'}\sum_{\mathbf{kk'}}|(2\mathbf{JS})^{pp'}_{MM'}|^2\mathcal{J}_{pk,p'k'}(\omega_{MM'}), \tag{3}$$

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$$\mathcal{J}_{pk,p'k'}(\omega)=\frac{1}{2\pi}\int_{-\infty}^{\infty}\langle a_{pk}^{+}a_{p'k'}(\tau)a_{p'k'}^{+}a_{pk}\rangle e^{i\omega\tau}d\tau,\quad \omega_{MM'}=E_M-E_{M'}.$$

We shall calculate the correlation function in (3) for the case $J=\frac{1}{2}$ by the method of equal-time temperature Green's function, using the equilibrium statistical operator of the system of carriers and of the localized moments that interact with them. For the Green's functions ($\mu=X,Y,Z$)

$$G_{pk,p'k'}=\langle\langle a_{pk}^{+}a_{p'k'};\ a_{p'k'}^{+}a_{pk}\rangle\rangle \text{ and } \langle\langle S_\mu a_{pk}^{+}a_{p'k'};\ a_{p'k'}^{+}a_{pk}\rangle\rangle$$

we shall set up the equations of motion and obtain, after the usual procedure of splitting[9] the chain of equations for the higher Green's functions, a closed equation for $G_{pk,p'k'}$. Assuming a random distribution of the N spins over the volume of the sample and the absence of carrier degeneracy, we represent the Green's function in the form ($\eta=+1$)

$$G_{pk,p'k'}(\omega+i0)=\frac{n_{pk}-n_{p'k'}}{2\pi[\tilde{\omega}+E_{pk}-E_{p'k'}+i\Gamma_{kk'}^{pp'}(\omega)]},\tag{4}$$

$$\Gamma_{kk'}^{\pm\mp}=\frac{NI^2\langle S_\mp S_\pm\rangle}{4\pi V_0}\left(\frac{2m}{\hbar^2}\right)^{1/2}[\varepsilon_k^{1/2}+\varepsilon_{k'}^{1/2}],\tag{5}$$

where m is the effective mass of the state density $n_{pk}=\langle a_{pk}^{+}a_{pk}\rangle$, ε_k and $\varepsilon_{k'}$ are the kinetic energies of the carrier before and after the exchange scattering, $\tilde{\omega}=\omega+h$ is the IC resonant frequency renormalized by the polarization field of the carriers

$$\mathrm{h}=-2eI\sum_p pn_p,\quad n_p=\sum_k n_{pk}.$$

In the case of simple bands Γ_6 and Γ_7 we have $2\sum_p pn_p=n_+-n_-$. The mean values $\langle S_\mp S_\pm\rangle$ are calculated under the same assumptions as $\langle S_\mu\rangle$ in (2). In the spin-temperature approximation for an equidistant IC spectrum, when $z\parallel\mathbf{H}$ and $\omega_0=\omega_{M,M-1}$ does not depend on M, we have

$$\langle S_\mp S_\pm\rangle=-\langle S_z\rangle[\mathrm{cth}(\beta\omega_0/2)\mp1],$$

where $\langle S_z\rangle$ is expressed in the usual manner in terms of the Brillouin function $B_S(\beta\omega_0)$. To estimate $\Gamma_{kk'}^{pp'}$ we note that $|\varepsilon_k-\varepsilon_{k'}|\geq G$ for interband transitions ($p\neq p'$). We therefore replace the sum of the radicals in the right-hand side of (5) by $G^{1/2}$ and leave out the lower pair of indices, on which $\Gamma_{kk'}^{pp'}$ no longer depends. Using this circumstance and substituting $\mathcal{J}_{pk,p'k'}(\omega)$, which is uniquely connected[9] with $G_{pk,p'k'}(\omega)$, in (3), we integrate over the final states $\mathbf{k}'$:

$$W_{M,M\pm1}=n_\pm I^2(S_\mp)_{M,M\pm1}^2(2m/\pi\hbar^2)^{1/2}(k_BT_K)^{1/2}F_{\gamma_{+-}}(g_Z),\tag{6}$$

$$F_\gamma(g)=2^{-1/2}\int_0^\infty\{x(x-g+[(x-g)^2+\gamma^2]^{1/2})\}^{1/2}e^{-x}dx,\tag{7}$$

where $g_Z=G_Z/k_BT_K$, $\gamma_{pp'}=\Gamma^{pp'}/k_BT_K$, and we used a Maxwellian distribution function (with temperature T_K) for the nondegenerate carriers.

Since the spin splittings of the bands are meaningful if

$$\Gamma_{kk'}^{pp'}\ll G,\tag{8}$$

we can simplify expression (7) by taking the limit as

$$F_\gamma(g)\xrightarrow[\gamma\to0]{}F(g),$$

where the function $F(g)$ was investigated in Ref. 10. Thus, the expressions for $W_{M,M'}$ obtained without allowance for the damping $\Gamma_{kk'}^{pp'}$ of the states $|pk\rangle$ and $|p'k'\rangle$, remain valid also at finite values of $\Gamma_{kk'}^{pp'}$, provided that Eq. (8), which is equivalent to the condition of good resolution of the carrier spin states split by the exchange field $\mathbf{G}$, is satisfied. Hereafter, on the basis of (8), we shall use (6) and (7) in the limit as $\gamma\to0$.

The generalization of (6) for arbitrary J leads to the expression

$$W_{MM'}=(S_\pm)_{MM'}\left(\frac{2}{\pi}\right)^{1/2}(I^2/\hbar^4)(k_BT_K)^{1/2}\sum_{pp'}n_pm_p^{3/2}(J_\mp)_{pp'}^2F\left(-\frac{\Delta_{pp'}}{k_BT_K}\right).\tag{9}$$

Here $\Delta_{pp'}=E_{p0}-E_{p'0}+\omega_{MM'}$ and $E_{p0}=G_Zp$; n_p and m_p are the carrier density and the effective mass of the state density in the spin subband p.

We note that (6) and (9) describe the relaxation also when the carrier spin temperature β_S-1 differs from the lattice temperature T (or, generally speaking, when the populations n_p of the spin subbands differ from the nonequilibrium values n_p^0).

3. SPIN RELAXATION OF IC

We consider now the influence of the carriers on the paramagnetic relaxation in a magnetically doped semiconductor. Under the conditions

$$n\ll N/V_0,\quad h\ll g\beta H,\tag{10}$$

which are usually realized in experiment[1-4] in the case of saturation of a small group $\Delta N\ll N$ of spins (constituting, for example, a spin packet in an inhomogeneously broadened EPR line), the relaxation is determined[2] by the exchange-scattering probability (9). In the temperature region $|\omega_{MM'}|<k_BT<G$ that is vital for the experiment, when the IC polarization is linearly connected with the reciprocal spin temperature and

$$F(G/k_BT)\approx{}^1/_2(\pi G/k_BT)^{1/2}e^{-G/k_BT},$$

the contribution of the free electrons (holes) to the relaxation of ΔN impurity centers in the case of the band Γ_6 (Γ_7) is determined by the time τ, where

$$\tau^{-1}=\frac{2}{\pi}\left[\frac{S(S+1)}{3}\right]^{1/2}[3(S_+)_{M,M-1}-1]n\left(\frac{N}{V_0}\right)^{1/2}\times\frac{|I|^{5/2}m^{1/2}\omega_0^{1/2}}{\hbar^4(k_BT)^{1/2}}\exp\left\{-\frac{2S(S+1)N|I|\omega_0}{3V_0k_B^2T_K^2}\right\}.\tag{11}$$

The expression for the rate of the relaxation of the IC in terms of the free holes in the case of the Γ_8 band ($J=\frac{3}{2}$) differs from (11) by a factor 3.

Thus, in the case of giant spin splittings of the band one can expect an anomalously strong temperature dependence of the rate of relaxation of the spin packet ($\propto T^{-1/2}\exp\{-\mathrm{const}/T^2\}$). The concentration dependence of τ also differs qualitatively from those known, leading to a prolongation of the relaxation with increasing N (so that $\tau^{-1}\propto N^{1/2}\exp\{-N\ \mathrm{const}\}$). We note that experimental observation of the temperature dependence of (11) makes it possible to determine independently the width of the exchange-interaction constant and the IC density, something usually difficult to do by spectroscopic methods.[3,4]

4. IC SPIN POLARIZATION IN EXCHANGE SCATTERING OF PHOTOCARRIERS OF THE SAME SIGN

An important distinguishing feature of optically generated carriers, compared with thermal electrons and holes, is that their spin temperature is generally speaking not equal to the lattice temperature. Let us examine the influence of this optical heating (or cooling) of the carriers on the IC polarization $\langle S_z \rangle$ in the case when the relaxation of the latter is determined by Eq. (9). This takes also into account the reaction of the polarization of the IC on the carrier spins. We start with the case when the IC interact effectively with carrier of the same sign, say with electrons ($J=\frac{1}{2}$), and assume for the sake of simplicity that the quadrupole transitions of the IC are negligible or can be taken into account together with the dipole transitions by means of a single spin-lattice relaxation time τ_{ML}. The basic kinetic equations for the IC spin-state populations N_M ($M=-S,\ldots,S$) and for the populations n_p of the conduction-electron spin subbands ($p=\pm\frac{1}{2}$) take in this case the form

$$\dot{N}_M = -N_M(W_{M,M-1}+W_{M,M+1})+N_{M-1}W_{M-1,M}+N_{M+1}W_{M+1,M}, \quad (12)$$

$$\dot{n}_p = -\sum_M [N_M W^e_{M,M+p} - N_M W^e_{M,M-p}] - n_p W_p + n_{-p} W_{-p} - n_p/\tau_r + S_p^*, \quad (13)$$

where $W_{M,M'} = W^e_{M,M'} + W^L_{M,M'}$, $W^L_{M,M'}$ describes the spin lattice relaxation of the IC, $W^e_{M,M'}$ is defined by expression (9), and $W_{\pm 1/2}$ is the probability of the relaxation transition $p=\pm\frac{1}{2} \to p'=\mp\frac{1}{2}$ and is connected with the conduction-electron spin-lattice interaction that establishes the populations

$$n_p^0 = n m_p^{3/2} \Big/ \sum_{p'} m_{p'}^{3/2} \exp\{(E_{p0}-E_{p'0})/k_BT\},$$

and S_p^*, τ_r are the rates of generation and recombination of the carriers. In (12) and (13), as well as hereafter, we put $V_0=1$. In the case $S=\frac{1}{2}$ Eq. (12) is exact.

The stationary solutions of Eqs. (12) and (13) correspond to $\dot{N}_M=\dot{n}_p=0$. If we neglect the non-equidistant IC spectrum due to the crystal field, the ratio

$$N_{M-1}/N_M = W_{M,M-1}/W_{M-1,M} \equiv e^{\varkappa} \quad (14)$$

is independent of M, and $\varkappa=\beta_S\omega_0$, where β_S is the reciprocal spin temperature of the IC. Substituting in (14) $W_{MM'}$ from (9) for $S=\frac{1}{2}$, we obtain after the necessary transformations

$$\operatorname{th}\frac{\varkappa}{2} = \frac{y-\operatorname{th}(\Delta/2k_BT_\kappa)+\operatorname{th}(\omega_0/2k_BT)\,\tau_{ML}^{-1}/\tau_{Me}^{-1}}{1-y\operatorname{th}(\Delta/2k_BT_\kappa)+\tau_{ML}^{-1}/\tau_{Me}^{-1}}, \quad (15)$$

where we put for brevity $\Delta \equiv \Delta_{+-}$; $y=(n_- - n_+)/(n_- + n_+)$ is the spin polarization of the conduction electrons

$$\tau_{Me}^{-1} = {}^1/_2 n[F(\Delta/k_BT_\kappa)+F(-\Delta/k_BT_\kappa)]U$$

is the rate of relaxation in exchange scattering of n_+ and $n_- = n_+ = (\frac{1}{2})n$ of the electrons by the IC; $U=m^2I^2\bar{v}/\pi\hbar^4$; $\bar{v}=(8k_BT/\pi m)^{1/2}$ is the average thermal velocity of the carriers in the band. Analogously, starting from (9) and (13), we obtain an equation for the polarization of the carriers

$$y = \frac{\operatorname{th}(\varkappa/2)+\operatorname{th}(\Delta/2k_BT_\kappa)+\bar{y}\tau_{erL}^{-1}/k_S(\varkappa)\tau_{eM}^{-1}}{1+\operatorname{th}(\varkappa/2)\operatorname{th}(\Delta/2k_BT_\kappa)+\tau_{erL}^{-1}/k_S(\varkappa)\tau_{eM}^{-1}}, \quad (16)$$

$$\bar{y} = (y^0\tau_{eL}^{-1}+y^*\tau_r^{-1})/\tau_{erL}^{-1}, \quad \tau_{erL}^{-1}=\tau_{eL}^{-1}+\tau_r^{-1}. \quad (17)$$

Here τ_{eL}^{-1} is the rate of the spin-lattice relaxation of the carrier; y^0 is the carrier spin polarization corresponding to the equilibrium population n_p^0 of the subbands p; $y^*=(S_-^*-S_+^*)/(S_-^*+S_+^*)$ is the polarization of the carriers at the instant of their production. Obviously $y^0<\bar{y}<y^*$ if $y^*>y^0$, or $y^*<\bar{y}<y^0$ if $y^*<y^0$. The IC spin-temperature function $k_S(\varkappa)=-2\langle S_z\rangle\coth(\varkappa/2)$ takes into account the difference between the contributions of the components of the multilevel spectrum of the IC to the carrier relaxation. At $S=\frac{1}{2}$ we have $k_S(\varkappa)=1$, and for an arbitrary S the function $k_S(\varkappa)$ describes a continuous dependence on $\varkappa$ and can be replaced in estimates by the constant $k_S=2S(2(S+1)/3)^{1/2}$. The rate of the spin relaxation of the electrons through the IC is defined in analogy with τ_{Me}^{-1}:

$$\tau_{eM}^{-1} = {}^1/_2 N[F(\Delta/k_BT_\kappa)+F(-\Delta/k_BT_\kappa)], \quad N=\sum_M N_M.$$

The system (15) and (16) of the equations linear in the spin temperatures of the IC and of the electrons is complete. Yet it can be shown that the polarizations y and $x=\tanh(\varkappa/2)$ are connected by a linear [accurate to within the replacement of $k_S(\varkappa)$ by k_S] equation ($x_0=\omega_0/2k_BT$ is the IC equilibrium polarization)

$$k_S(\varkappa)N\tau_{ML}^{-1}(x-x_0)+n\tau_{erL}^{-1}(y-\bar{y})=0, \quad (18)$$

which expresses the condition that the total angular momentum of the system is stationary. Since the exchange scattering does not change the total angular momentum of the electrons and of the IC, the expression (18) remains valid at all exchange-interaction constants. Substituting y from (18) in (15) we obtain a closed equation for the spin temperature of the IC. In the case of sufficiently strong pumping $S_-^*+S_+^*=n/\tau_r$ we have

$$\tau_{ML}^{-1} \ll \tau_{Me}^{-1}; \quad (19)$$

when the relaxation of the IC is determined by the exchange scattering of the nonequilibrium carriers, we obtain for the polarization x

$$\operatorname{th}(\operatorname{Arth} x - 2K\langle S_z\rangle) = \bar{y} - \alpha_e(x-x_0);$$

$$K = \frac{NI}{2k_BT_\kappa}, \quad \alpha_e = \frac{N\tau_{ML}^{-1}}{n\tau_{erL}^{-1}}k_S(\varkappa). \quad (20)$$

We analyze the solutions of (20) for the case where $S=\frac{1}{2}$, when the equation takes the simpler form

$$\operatorname{th}(\operatorname{Arth} x + Kx) = \bar{y} - \alpha_e(x-x_0). \quad (21)$$

The left-hand side of (21) is a single-parameter family of curves that fill densely the square $-1<x,y<1$ on the coordinate plane (Fig. 1) at $-\infty<K<\infty$. The solutions (21) are the ordinates of the points of intersection of these curves with a line drawn through the point $(x_0,\bar{y})$ with a slope $-\alpha_e$ (in this case the dependences of $\bar{y}$ and of α_e on x turn out to be inessential if $\tau_r<\tau_{eL}$ and $\alpha_e<1$).

Thus, if the exchange scattering controls the IC relaxation, and the spin relaxation of the carriers is due to recombination or interaction with phonons, then the stationary polarization x of the IC at high T coincides with the stationary polarization $\bar{y}$ of the carriers. With decreasing T, the polarization x becomes stronger if $I<0$ or weaker if $I>0$. The weakening of the polarization with increase of $K>0$ is physically due to the fact that

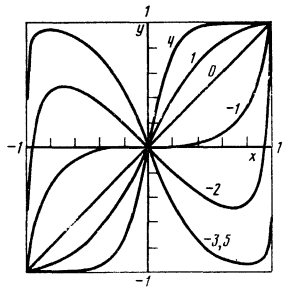

FIG. 1. Plots of the family of curves determined by the left-hand side of Eq. (21). The corresponding values of K are shown in the figure.

when the spin splitting of the bands is increased a larger role is assumed by the relaxation transitions $p=+\frac{1}{2} \to p'=-\frac{1}{2}$, which are accompanied by an IC spin flip $M \to M-1$ and decrease by the same token the polarization of the system. If $K<0$, transitions $p=-\frac{1}{2} \to p'=+\frac{1}{2}$ accompanied by enhancement of the IC polarization predominate.

In the case $-1<K<\infty$ the solution of Eq. (21) is unique; in the case $K<-1$ there appears a region of multiply valued solutions x_i $(i=1,2,3)$, and if x_1 goes over continuously into the unique solution into $K>-1$, then the signs of x_2 and x_3 coincide and are opposite to the sign of x_1.

The family of solutions $x_i=x_i(K)$ obtained with the aid of the diagrams of Fig. 1 for different values of $\bar{y}$ in the important case $\alpha_e \ll 1$ is shown in Fig. 2. It is seen that at $|K| \gg 1$ and $K<0$ it is possible to obtain almost complete polarization, both along the external field $(x_1 \to 1)$ and in a direction opposite to the field $(x_3 \to -1)$. One more solution x_2 $(x_2 \to 0$ as $K \to -\infty)$ is not stable to small fluctuations of x and can therefore not be realized in experiment. An asymptotically exact solution of Eq. (21) is obtained in the limit as $|K| \to \infty$:

$$\begin{aligned} &x=x_{1,3}=\max\{-1,\ (\bar{y}+\alpha_e x_0-1)/\alpha_e\},\quad \delta_+>0;\\ &x=x_2=(\bar{y}+\alpha_e x_0)/(K+1+\alpha_e),\quad \delta_+\delta_-<0;\\ &x=x_{3,1}=\min\{1,\ (\bar{y}+\alpha_e x_0+1)/\alpha_e\},\quad \delta_-<0;\\ &\delta_\pm=(K/|K|)\{\pm[\bar{y}-\alpha_e(\pm 1/(K+1)-x_0)]-1\},\\ &x_{1,3}=x_1,\quad \delta_+>\delta_-,\quad x_{1,3}=x_3,\quad \delta_+<\delta_-; \end{aligned} \tag{22}$$

$x_{3,1}$ differs from $x_{1,3}$ in that x_1 is replaced by x_3. In essence, expressions (22) approximate with sufficient accuracy the solutions of Eqs. (21) if $|K| \geq 4$. The multiple roots in (22) appear under the condition $K<-1-\alpha_e$ and $|\bar{y}+\alpha_e x_0|-\alpha_e/(K+1)<1$. In the other cases, expression (22) describes the only solution that coincides formally with one of the roots x_1, x_2, or x_3, depending on the signs of δ_+ and δ_-. The maximum polarization of the IC spins is reached in the limit $|K| \gg 1$ at $K<0$, if $\bar{y}+\alpha_e(1+x_0)<1$ $(x \to -1)$ or $\bar{y}=\alpha_e(1-x_0)>-1$ $(x \to 1)$. We note that these conditions can be satisfied also in a vanishingly small external field $\mathbf{H}$ $(x_0 \ll 1)$, particularly at $\mathbf{H}=0$ (when x_0 is determined by the field $\mathbf{h}$ of the polarized carriers). An interesting aspect of the theory may be the situation in which $\mathbf{H}$ is adiabatically slowly turned off after a maximum polarization of the IC is attained by unpolarized interband illumination $(y^*=0)$. The polarization of the IC preserves here its direction and is almost unchanged in magnitude. Similarly, the polarization IC is preserved in the adiabatic flow reversal $\mathbf{H} \to -\mathbf{H}$, so that the polarized spins acquire a negative temperature. The last case can be regarded as a method of obtaining a polarization corresponding to the second root of the stable equations (22).

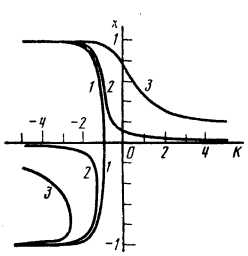

FIG. 2. Dependence of the polarization x of the IC spin system on $K=NI/2k_BT_K$ at $\alpha=0$ and different values of $\bar{y}$: 1) $\bar{y}=0$; 2) $\bar{y}=0.1$; 3) $\bar{y}=0.75$.

5. POLARIZATION OF IC SPINS IN EXCHANGE SCATTERING OF NONEQUILIBRIUM ELECTRONS AND HOLES

In magnetically doped II–VI semiconductors, the contributions of the electrons and holes to the exchange scattering have commensurate exchange constants but of opposite sign.[3-6] Since the kinetics of the spins depends substantially on the sign of I, this case calls for a special analysis. Qualitative tracking of the change of the effective polarization because of the presence of two types of carrier is convenient for simple bands, since allowance for the angular momentum $\frac{3}{2}$ of the hole complicates the situation.

We start from the kinetic equation (12) with $W_{MM'}=W^e_{MM'}+W^h_{MM'}+W^L_{MM'}$, where $W^e_{MM'}$, $W^h_{MM'}$ are determined by Eq. (9) and are proportional to the densities of the electrons and holes, respectively. In the equations for the populations n^e_p and n^h_p of the spin subbands it is necessary to take into account, generally speaking, the exchange scattering of the electrons by the holes with mutual spin flips.[11] Yet these processes are proportional to the product $n^e n^h$ and by virtue of (10) are negligibly small compared with the exchange scattering by the IC. In addition, since the mutual spin flips of the carriers are accompanied by exchange of an energy quantum equal to the sum of the exchange fields G^e and G^h, their probabilities are small if (1) is satisfied, and are proportional to $\exp[-(G^e+G^h)/k_BT_K]$. A procedure similar to that in the preceding section leads to an equation that generalizes (15) to the case of exchange scattering of two species of carrier:

$$x=\frac{\tau_{Me}^{-1}[y-\mathrm{th}(\Delta^e/2k_BT_K)]+\tau_{Mh}^{-1}[z-\mathrm{th}(\Delta^h/2k_BT_K)]+x_0\tau_{ML}^{-1}}{\tau_{Me}^{-1}[1-y\,\mathrm{th}(\Delta^e/2k_BT_K)]+\tau_{ML}^{-1}[1-z\,\mathrm{th}(\Delta^h/2k_BT_K)]+\tau_{ML}^{-1}}. \tag{23}$$

We obtain a closed system of equations for the spin polarizations x of the IC, y of the electrons, and $z=(n^h_- - n^h_+)/n^h$ of the holes, by supplementing (23) with Eq. (16) and with an analogous equation for z with the parameters of the hole band Δ^h, τ_{hM}, and τ_{hL}, whose meaning is the same as for the conduction electrons.

The requirement that the total angular momentum of the spins be stationary is derived from the aforementioned system of equations for x, y, and z and takes the

form

$$k_s(\varkappa)N(x-x_0)\tau_{ML}^{-1}+n_e(y-\bar{y})\tau_{erL}^{-1}+n_h(z-\bar{z})\tau_{hrL}^{-1}=0. \quad (24)$$

This equation, just as (18), is valid for all exchange-scattering parameters and establishes the flux balance of the spin moments connected with the spin-lattice relaxation and with the carrier recombination. Since $x-x_0$, $y=\bar{y}$, and $z-\bar{z}$ do not exceed 2 in absolute value, the condition (24) imposes the following limitations on the possible values of the polarizations:

$$|x-x_0|<(n_e\tau_{erL}^{-1}+n_h\tau_{hrL}^{-1})/NS\tau_{ML}^{-1}. \quad (25)$$

Similar estimates can be obtained for $|y=\bar{y}|$ and $|z-\bar{z}|$. It follows also from (24) that the system cannot be stationary if the signs of the deviations of the polarizations $x=x_0$, $y-\bar{y}$, and $z=\bar{z}$ are equal.

The general solution for x, y, and z depends on a large number of parameters and can be obtained by numerical methods with a computer. A qualitative analysis will be carried out for a model system that is completely symmetrical in its kinetic properties for the electrons and holes, but with opposite signs of the exchange constants: $I_e=-I_h$. Putting $n^e=n^h$, $\tau_{eM}=\tau_{hM}$, $\tau_{eL}=\tau_{hL}$, and $\alpha_e=\alpha_h$ we obtain an equation for the IC polarization with the aid of (16), (23), and (24). In the case $\tau_{erL}^{-1}\ll\tau_{eM}^{-1}$, $S=(\frac{1}{2})$, and when (19) is satisfied,

$$x\frac{1-\mathrm{th}^2 2|K|x}{1-x^2\,\mathrm{th}^2 2|K|x}=\frac{1}{2}(\bar{y}+\bar{z})-\frac{\alpha}{2}(x-x_0), \quad (26)$$

where the coefficients α and K were determined in the preceding section. The single-parameter family of curves in the left-hand side of (26) fills solidly $\frac{1}{4}$ of the area of the unit square of the coordinate plane between the abscissa axis and the line $y=x$ when $|K|$ is varied from 0 to ∞. In the case of low temperatures ($|K|>3$) the only solution of Eq. (26) is approximated by

$$x=\mathrm{sign}\left(x_0+\frac{\bar{y}+\bar{z}}{\alpha}\right)\min\left\{1,\left|x_0+\frac{\bar{y}+\bar{z}}{\alpha}\right|\right\}. \quad (27)$$

It follows from (27), in particular, that the most favorable situation for the IC polarization is realized when the signs of the polarization of the optically produced electrons and holes are identical. Conversely, at $\bar{y}+\bar{z}=0$ the polarization of the IC remains at equilibrium, in contrast to the case of exchange scattering of carriers of the same sign, when an arbitrarily large polarization of the IC might be reached at $\bar{y}=0$ (Fig. 2).

Another particular case in which analytical solutions can be obtained for x, y, and z does not require equivalence of the electron and hole bands, and is realized when the IC polarization deviates little from equilibrium: $|x-x_0|\ll 1$. The right-hand side of (16) can then be expanded in powers of $x-x_0$ and, with the first two terms retained, substituted in (24). After similar substitutions of the expansions of z, α_e, and α_h in (24) we obtain a linear algebraic equation for $x=x_0$. In the case $\alpha_e,\alpha_h\gg 1$ (which automatically leads to $|x-x_0|\ll 1$), the equation for $x-x_0$ coincides formally with (24) if the substitutions $y=y(x_0)$ and $z=z(x_0)$ are made. The contributions of the electrons and holes in this case are additive and can differ in magnitude and in sign.

6. CONCLUSION

The results of this paper are applicable not only to cubic crystals, provided that the exchange scattering remains isotropic. This pertains, for example, to the electrons in hexagonal[5] II–VI crystals. We note that the holes in uniaxial semiconductors in the Γ_9 band do not take part in the exchange scattering with spin flip if **H** is parallel to a crystal axis, and consequently can be excluded from consideration. Our results are also applicable to exchange scattering of electrons, if the exchange fields G^e and G^h exceed the intra-exciton electron-hole exchange interaction. The contribution to the IC relaxation from carriers bound into excitons is additive, and the polarization of the IC is the same as in exchange scattering of free electrons and holes in the case $n^e=n^h$.

If we use for $A^{II}B^{IV}$: Mn crystals the values $N/V_0=10^{20}$ cm^{-3}, $NI/V_0=100$ cm^{-1}, $\tau_{erL}=10^{-2}$ sec, and $\tau_{ML}\sim 10^{-6}$ sec, then the case α_e, $\alpha_h\gg 1$ is realized at helium temperatures and at photocarrier densities $n=10^{15}$ cm^{-3}, so that the change of the spin polarization for Mn^{2+} does not exceed several percent. At a lower density $N/V_0\lesssim 10^{19}$ cm^{-3} and at $T=1.5$°K ($|K|\gg 1$ as before) one can expect a lengthening to $\tau_{ML}\sim 10^{-4}$ sec, as a result of $\alpha\lesssim 0.1$, and a high degree of spin polarization of Mn^{2+} is possible. By lowering N in proportion to T (so as to preserve the condition $|K|\gg 1$), we can lengthen τ_{ML} to the spin-lattice relaxation time τ_{ML}^0 of isolated centers, at which it suffices to use the minimum pump level $S_{\min}^*=n_{\min}/\tau_r$ needed to polarize the Mn^{2+} spins. Putting $\tau_{ML}^0\approx 10^{-2}$ sec at $N/V_0=10^{18}$ cm^{-3} we get $n_{\min}=10^{11}-10^{12}$ cm^{-3}.

For more accurate estimates of the experimental conditions for the observation of polarization effects, additional measurements of τ_{ML} in $A^{II}B^{IV}$: Mn are needed in a wide interval of IC densities, $10^{18}-10^{20}$ cm^{-3}.

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[1)]The corresponding effective-mass tensor depends on the magnitude and the direction of H, and is determined by the secular matrix for the band electron in the ideal crystal,[7] supplemented by the matrix element of the operator $\mathbf{G}\cdot\mathbf{J}$, which lifts the band degeneracy completely.

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