Optical orientation of Mn$^{2+}$ ions in GaAs

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We report on optical orientation of Mn$^{2+}$ ions in bulk GaAs under application of weak longitudinal magnetic fields ($B \leq 100$ mT). A manganese spin polarization of 25% is directly evaluated using spin-flip Raman scattering. The dynamical Mn$^{2+}$ polarization occurs due to the s-d exchange interaction with optically oriented conduction band electrons. Time-resolved photoluminescence reveals a nontrivial electron spin dynamics, where the oriented Mn$^{2+}$ ions tend to stabilize the electron spin.

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Electron spin interactions in semiconductors are in the focus of current attention as they are of interest for various applications in electronics and quantum information. A pathway to spin manipulation is to implement magnetic ions in semiconductors and exploit the strong exchange interaction between an electron (hole) and a magnetic ion (for example Mn). A spectacular example to that end is a quantum dot (QD) with a single Mn ion $^{17}$. Another model realization has been achieved for GaAs doped with Mn acceptors of low concentration, where a photoexcited electron is localized on a residual donor in vicinity of a Mn acceptor $^{11-12}$. Optical orientation of manganese acceptors and their spin control are attractive for investigation of various nontrivial scenarios of electron spin dynamics in semiconductors, e.g. multi-exponential spin decay or spin precession in effective exchange fields. Optical orientation of a single Mn$^{2+}$ ion in a II-VI QD has been recently reported $^{13,14}$. However, the question whether this is feasible in III-V materials, such as GaAs, has remained open $^{14}$. Here we report on the direct observation of optical spin orientation of Mn$^{2+}$ ions in bulk GaAs under optical illumination with circular polarized light in longitudinal magnetic fields (Faraday geometry). In spin-flip Raman scattering (SFRS) we observe spectral lines corresponding to spin-flip processes of the ionized Mn acceptor. The asymmetry of Stokes and anti-Stokes (SAS) line intensities under circular polarized excitation opens a way for direct evaluation of the manganese spin polarization, which can reach 25%. Time-resolved photoluminescence (TRPL) reveals a nontrivial electron spin dynamics resulting from the stabilizing feedback of the oriented manganese on the electron spin. The results of SFRS and TRPL agree well, supporting clearly optical manganese orientation in weak magnetic fields.

We investigate bulk GaAs doped with Mn acceptors with a concentration of $8 \times 10^{17}$ cm$^{-3}$ $^{3}$. The acceptors are partially compensated by residual shallow donors with a concentration of about $N_D \sim 10^{16}$ cm$^{-3}$ $^{3}$. This results in ionization of the acceptors located in the vicinity of donors if not illuminated by light. Detailed descriptions of the experimental techniques can be found in Ref. $^8$ for SFRS and in Ref. $^4$ for TRPL.

In order to detect manganese polarization we exploit SFRS technique, which selectively monitors the changes of Mn$^{2+}$ spin projection by $\alpha = 1, 2, 2I$ with Mn spin $I = 5/2$. It enables to measure both the Zeeman splitting $\mu_B B$ and the amplitudes of SAS components. Here $\mu_B$ is the Bohr magneton, and $g = 2$ is the Mn - Lande factor. The ratio between the Stokes and anti-Stokes line intensities, $T_S$ and $T_{AS}$, respectively is given not only by the selection rules but also by the Mn$^{2+}$ spin polarization $P_M$. It can be shown that in Faraday geometry for circular co-polarized excitation and detection with helicity $\sigma$ and for a small manganese polarization ($P_M \ll 1$) the SAS asymmetry parameter of SFRS,

$$\eta^\sigma_n = \frac{T_S^{\sigma} - T_{AS}^{\sigma}}{T_S^{\sigma} + T_{AS}^{\sigma}} = -\frac{3n}{2(I+1)}P_M(\sigma, B), \quad (1)$$

is independent on the specific Mn spin-flip mechanism. In this case $\eta^\sigma_n$ directly monitors the spin polarization of manganese. In Eq. (1) we assumed: (i) the spectral width of the spin-flip resonance profile is larger than the Zeeman spin splitting; (ii) Raman scattering without conservation of the total angular momentum of manganese and the exciton in the intermediate state is possible. Such transitions are allowed in presence of anisotropic exchange interaction between the exciton and the manganese 3d$^5$ electrons, i.e. if the symmetry of the system is reduced $^{3,10}$.

The manganese polarization $P_M(B, P_e)$ depends not only on the external magnetic field due to the thermal population of the Zeeman sublevels, but also on the non-equilibrium electron polarization $P_e$ due to the dynamical polarization of Mn spins by electrons (holes are not oriented in bulk GaAs $^{11}$). This process is similar to dynamical polarization of the nuclei in semiconductors (Overhauser effect) and has been considered for II-VI doped magnetic semiconductors $^{12}$. The dynamical polarization changes sign if the helicity of circular polarized excitation, $\sigma^+$ or $\sigma^-$, is reversed. Therefore, it is necessary to measure the two asymmetry parameters $\eta^{\sigma^+}_n$ and $\eta^{-\sigma}_n$ for the two polarizations.

Figure (1a) shows SFRS spectra for the $\sigma^+/\sigma^+$ and $\sigma^-/\sigma^-$ excitation/detection polarizations in a magnetic...
field of 2 T. Two pairs of lines corresponding to transitions where the angular momentum changes by \( n = 1, 2 \) are detected. Their magnetic field dependence follows the expected linear dependence, \( n g \mu_B B \), with \( g = 2.0 \) [see inset in Fig. 1(a)], corresponding therefore to spin-flip transitions between the Zeeman sublevels of the Mn\(^{2+}\) ions, i.e. the ionized manganese acceptors A\(^-\) [13]. The magnetic field dependence of the SAS asymmetry parameter \( \eta_n^m \) for \( \sigma^+ \) and \( \sigma^- \) excitation and \( n = 1, 2 \) is shown in Fig. 1(b). The \( \eta_n^m \) increase with \( n \) and \( B \), and they also depend on the helicity of laser excitation, from which we can conclude that optical orientation of manganese has been accomplished. Using Eq. 14 we deduce the dependence of the manganese polarization \( P_M \) on magnetic field for \( \sigma^+ \) and \( \sigma^- \) excitation. The difference between the two polarizations allows us to estimate the contribution of the optical orientation to the manganese polarization \( P_{OO} = \left[P_M(\sigma^+, B) - P_M(\sigma^-, B)\right]/2 \approx -0.25 \), independent of magnetic field for \( B > 1 \) T [see Fig. 1(c)]. The other contribution to \( P_M \), related to the thermal population of the Zeeman sublevels \( P_B = \left[P_M(\sigma^+, B) + P_M(\sigma^-, B)\right]/2 \), grows with field and is comparable with \( P_{OO} \) at \( B \approx 1 \) T. The spin-flip resonance profile has a maximum at about 1.513 eV, which corresponds to resonant excitation of the \( 1S \) sublevels (i.e. \( S = 1/2 \)) and \( g_B = 2 \). The steady state solution of Eq. (2) neglecting \( P_{eT} \) gives

\[
P_M = P_T + \frac{T_L}{T_L + T_M} \frac{I + 1}{S + 1} P_e.
\]

Here the first term corresponds to the equilibrium orientation resulting from thermalization on the Zeeman sublevels (i.e. \( P_T = P_B \)), while the second term is the dynamical polarization \( P_{OO} \) by electrons. The electron polarization is determined by the laser helicity. A polarization \( |P_e| = 0.36 \) was directly measured by optical orientation at the band edge under the same experimental conditions. Therefore, Eq. (3) can be used for evaluating the time ratio \( T_M/T_L \) and the actual temperature \( T \) in the region of illumination. Using the experimental value \( P_{OO} = -0.25 \) we obtain \( T_M/T_L = 2 \) and \( g_B g B/T = 0.35 \) (for \( B = 1 \) T). This corresponds to \( T = 4 \) K, i.e. the real temperature under illumination is increased by 2 K. The same value for \( T \) follows from the magnetic field dependence of \( P_B \) when fitted with a Brillouin function, see Fig. 1(c).

Electron spin-flip scattering with Mn\(^{2+}\) ions should also manifest itself in the dynamics of the average electron spin. Indeed, since the Mn polarization will be transferred back into the electron spin-system, the average electron spin should contain information about the Mn polarization. In absence of other relaxation channels the dynamics of electron polarization is given by an expression similar to the \( T_M \)-term in Eq. (2)

\[
\frac{dP_e}{dt} = -\frac{1}{\tau_S} \left[ P_e - P_{eT} \left( S + 1 \right) \left( P_M - P_T \right) / T + 1 \right],
\]

since it reflects the conservation of total spin in the flip-flop process. The electron spin relaxation time \( \tau_S \) depends on the Mn\(^{2+}\) concentration \( N_M \) and time \( T_M \) through \( \tau_S = S(S+1) N_e \left( T + 1 \right) / N_M T_M \). For small photoelectron concentrations \( N_e \ll N_M \) the electron spin relaxation time is much shorter than that of manganese, i.e. \( \tau_S \ll
$T_M$ (a similar relation holds for the electron-nuclear system). From TRPL $\tau_S \approx 10$ ns and, therefore, one can estimate $T_M \approx 10$ μs for $N_e = 10^{14}$ cm$^{-3}$, and $N_M \sim N_D \sim 10^{16}$ cm$^{-3}$. Under pulsed photoexcitation with repetition period $t_i$ (which satisfies the condition $T_M > t_i > \tau_S$), the manganese spin polarization is gradually accumulated and shows no change within time $t_i$. Therefore, in weak magnetic fields ($|P_T| \ll 1$) the solution of Eq. (4) in a time domain can be written as

$$P_e(t) = \frac{I+1}{S+1}P_M + \left[ P_i - \frac{I+1}{S+1}P_M \right] \exp \left(-\frac{t}{\tau_S}\right). \quad (5)$$

At the moment of pulsed excitation $t = 0$ the maximum possible electron polarization $P_i = -0.5$ is generated [11]. Subsequently it decays with a characteristic time $\tau_S$ until it reaches the plateau, which corresponds to the steady state non-equilibrium manganese polarization. The plateau results from the spin back flow from manganese to the electrons, providing a long-lived electron spin memory.

In previous TRPL measurements in weak longitudinal magnetic fields we already observed a slow spin relaxation dynamics (up to 1 μs) of electrons localized on shallow donors in GaAs:Mn [4]. However, the small signal-to-noise ratio in these studies did not allow us to draw an unambiguous conclusion on the non-exponential spin evolution. Large power densities induce undesired local heating. Therefore, we increased the setup sensitivity by enlarging the illumination area by a factor of 400, while keeping the pulse energy density low at $P_{exc} \approx 10$ nJ/cm$^2$. The intensity transient of the donor-acceptor ($D^0 - A^0$) PL line is shown in Fig. 2(a), showing a decay with 100 ns lifetime.

The decay of circular polarization degree $\rho_e(B,t) = -P_e(B,t)/2$ gives direct access to the spin dynamics of the oriented electrons. Figure 2(b) shows a nontrivial electron spin dynamics [10]. We find an initial electron spin $|P_e(0)| = 0.4$, which is close to the maximum value of 0.5. After decay within several tens of ns the electron spin polarization reaches a plateau, whose level increases with magnetic field and reaches 0.35 for $B = 156$ mT. Using Eq. (5) we determine the magnetic field dependencies of $\tau_S$ and $P_M$, as presented in Fig. 2(c). The electron spin relaxation time increases from 20 to 100 ns in a magnetic field of 150 mT. The plateau values corresponding to $P_M$ are symmetric with respect to magnetic field inversion and they change sign if the excitation helicity sign is reversed (i.e., they follow the electron spin polarization). This corroborates our conclusion about optical orientation of manganese. The manganese orientation is absent in zero magnetic field, in accord with [1], however, it appears in weak magnetic fields and saturates for $B > 150$ mT in line with the $P_{OO}$ behavior from SFRS, see Fig. 1(c).

We can exclude any influence of dynamical polarization of lattice nuclei since nuclear polarization should appear in much smaller magnetic fields $B \geq B_L \approx 0.3$ mT, where $B_L$ is the local field onto a nucleus by the neighboring nuclei [11]. Moreover, we do not observe any plateau in p-GaAs samples doped with non-magnetic Ge acceptors [4]. Thus, the plateau evidences manganese dynamic polarization in weak longitudinal magnetic fields, which suppress Mn$^{2+}$ spin relaxation.

Above we discussed the spin transfer between the localized electrons and the Mn$^{2+}$ ions. This transfer occurs due to fluctuations of the exchange interaction given by the Hamiltonian $H_{sd} = -b\hat{S}\cdot\hat{I}$. Here $\hat{S}$ and $\hat{I}$ are the electron and manganese spin operators, respectively, and $b$ is a constant depending on the electron probability at the Mn$^{2+}$ site. Apart from the fluctuation term the exchange interaction in mean-field approximation contains an expression like $-\langle b \rangle P_M \hat{S} - \langle b \rangle f \hat{S} \hat{P}_e \hat{I}$, where the exchange constant $\langle b \rangle$ is the average electron probability at the Mn$^{2+}$ site and $f$ is the donor filling factor. The first term describes the interaction of the electron spin with the effective exchange field of manganese $B_M = -\langle b \rangle P_M/\mu_B g_e$ (analogue of the Overhauser field). The second term corresponds to interaction of manganese spins with the effective field of the electrons $B_e = -f\langle b \rangle \hat{S} \hat{P}_e/\mu_B g$ (analogue of the Knight field). For small excitation densities $f \ll 1$, and as a result $B_M \gg B_e$.

The presence of the exchange manganese field can be detected via changes in the electron Larmor precession frequency in an external magnetic field. Since manganese polarization is absent in Voigt geometry [4] it is necessary to perform this experiment in oblique magnetic field for $\sigma^+$ and $\sigma^-$ polarized excitation. These data are summarized in Fig. 3. Figure 3(a) shows the oscillations of the circular polarization degree of the PL in time for the two opposite helicities ($B = 55$ mT applied at an angle $\alpha \approx 70^\circ$ with respect to the light propagation direction). Indeed we see that the Larmor frequencies $\Omega_{\pm}$ corresponding to electron spin precession with $g_e = -0.42$ are different ($\Omega_- - \Omega_+ = 0.11$ ns$^{-1}$). Based on this differ-
We can plot the dependence of the effective magnetic field \( B_M = \hbar (\Omega_+ - \Omega_-)/2 \mu_B g_e \) on external magnetic field \( B \), as shown in Fig. 3(b). The clear correlation between the magnetic field dependencies of \( B_M \) and the plateau value \( P^{pl}_M \) indicates their common origin: both are proportional to the non-equilibrium manganese polarization \( P \).

One can also estimate the exchange constant \( \langle b \rangle \). We note that in oblique magnetic fields no oscillations with the Mn\(^{2+}\) \( g \) factor are observed. This indicates that the manganese orientation is parallel to the direction of the external magnetic field \( B \), i.e., effective optical pumping occurs only for the spin component along \( B \). Therefore, the polarization \( P_M(\alpha) = P_M(\alpha = 0) \cos(\alpha) \). Then in the oblique geometry the field \( B_M = -\langle b \rangle |P_M(\alpha = 0) \cos(\alpha)/\mu_B g_e| \). Simultaneously from Eq. \[ \] we have for the plateau \( P^{pl}_M = \frac{1}{2+1} P_M(\alpha = 0) \). The ratio of \( B_M/P^{pl}_M \) from Fig. 3(b) allows one to derive \( \langle b \rangle = 0.5 \mu eV \). This is in agreement with the value of \( b_0 = 2.4 \mu eV \) for Mn\(^{2+}\) in the center of the donor, giving an upper limit for \( \langle b \rangle \).

In conclusion, we have demonstrated optical orientation of Mn\(^{2+}\) acceptors in GaAs using two optical techniques. Dynamic manganese polarization is established in weak longitudinal magnetic fields \((B \leq 100 \text{ mT})\), which are required to suppress the Mn\(^{2+}\) spin relaxation. The optically oriented Mn\(^{2+}\) ions maintain the spin and return part of the polarization back to the electron spin system providing a long-lived electron spin memory.

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**Appendix A: Derivation of Equation (1)**

Raman scattering of circular polarized light accompanied by spin flip of Mn\(^{2+}\) in longitudinal magnetic field (Faraday geometry) is shown in Fig. 4. In the initial state \( |\sigma_1 \omega_1 \rangle \) the photon with circular polarization \( |\sigma_1 \rangle \) and energy quant \( \hbar \omega_1 \) propagates along the magnetic field \( B \). There is Mn\(^{2+}\) ion in a quantum state \( |M \rangle \) with a given spin projection \( M = -5/2 \ldots + 5/2 \) on \( B \) direction and Zeeman energy \( E_M = \pm \mu_B B M \) (Bohr magneton \( \mu_B > 0 \), \( g \)-factor \( g = 2.0 \)). In the intermediate state there is an exciton \( |X \rangle \), which due to exchange interaction transfers the manganese from state \( |M \rangle \) into \( |M' = M \pm n \rangle \), while changing its state into \( |X' \rangle \). In the final state \( |\sigma_2 \omega_2 \rangle \) the photon with circular polarization \( |\sigma_2 \rangle \) and energy quant \( \hbar \omega_2 = \hbar \omega_1 + \mu_B B (M - M') \) propagating along the direction \( B \) is registered. In case if the manganese projection is increased (decreased) by \( n \) the photon energy decreases (increases) by \( n \cdot \mu_B B \) with respect to initial energy value, leading to the frequency shift of the scattered light in Stokes (anti-Stokes) region.

The number of light scattering events (in time per unit volume) in the direction parallel to the magnetic field from the initial into the final state is given by the probability of scattering \( W_{\sigma_1 \omega_1 M \rightarrow \sigma_2 \omega_2 M'} \) from \( |\sigma_1 \omega_1 M \rangle \) into \( |\sigma_2 \omega_2 M' \rangle \) and the initial manganese states \( |M \rangle \) distribution function \( N_M(B) \) in magnetic field under excitation.
with $|\sigma_1\rangle$ polarized light.

$$\mathcal{I} = W_{\sigma_1 \omega_1 M \rightarrow \sigma_2 \omega_2 M'} N_M^\sigma (B).$$  \hspace{1cm} (A1)$$

The dependence of $N_M^\sigma (B)$ from excitation light helicity $|\sigma_1\rangle$ is especially important. It takes into account possible optical orientation of manganese ions. Since the spin-flip is detected at the energy $\mu_B g B \cdot |M' - M|$ in the final state of the crystal there is only manganese spin, which is flipped, while all other lattice variables are fixed. Therefore the probabilities for back and forward transitions are equal to each other

$$W_{\sigma_1 \omega_1 M \rightarrow \sigma_2 \omega_2 M'} = W_{\sigma_2 \omega_2 M' \rightarrow \sigma_1 \omega_1 M}. \hspace{1cm} (A2)$$

If the spectral width of spin-flip resonance profile is larger than the Zeeman splitting of manganese spin sublevels, then the transition probability weakly depends on the light frequency $\omega_1 (\omega_2)$. Such kind of situation is realized in GaAs:Mn with $N_{Mn} \sim 10^{17} \text{cm}^{-3}$. Therefore we assume that $\omega_1 = \omega_2 = \omega$ in Eq. (A2) and leave index $\omega$.

Let us show that for identical polarizer and analyzer positions ($\sigma_1 = \sigma_2 \equiv \sigma$) the asymmetry parameter $n_\sigma^\mu$ does not depend on exact form of probabilities $W_{\sigma M \rightarrow \sigma M'} \equiv W_M, M' = W_{M', M}$ for small manganese polarization $P_M (\sigma, B) = \frac{1}{2} \sum_{M = -5/2}^{5/2} M \cdot N_M^\sigma (B)$. In this case the detected signal corresponds to the emission of excitons, which do not change their spin polarization in the scattering process. We consider the spin-flip process of nth order when $M' = M \pm n$, where $1 \leq n \leq 2I = 5$. Then the intensity of transitions in Stokes range ($M' = M + n$, photon energy shift to lower energies by $n \cdot \mu_B g B$) for polarizer and analyzer with polarization $\sigma$

$$I_n^S = \sum_{M = -5/2}^{5/2} W_{M, M + n} N_M^\sigma (B). \hspace{1cm} (A3)$$

In the same way the intensity of transitions in anti-Stokes range ($M' = M - n$, photon energy shift to higher energies by $n \cdot \mu_B g B$)

$$I_n^{AS} = \sum_{M = -5/2}^{5/2} W_{M, M - n} N_M^\sigma (B). \hspace{1cm} (A4)$$

We assume that the manganese spin sublevels distribution function

$$N_M^\sigma (B) = \frac{1}{2I + 1} \left[ 1 + \frac{3M}{I + 1} P_M (\sigma, B) \right] \hspace{1cm} (A5)$$

is defined only by polarization. It may take place when the manganese spin system is mainly disordered, i.e. $P_M \ll 1$, while the alignment parameters of higher order are negligible. The equation (1) follows from Eqs. (A2-A5) and the following relations

$$\sum_{M = -5/2}^{5/2} W_{M, M - n} = \sum_{M = -5/2}^{5/2} W_{M - n, M} = \sum_{M = -5/2}^{5/2} W_{M', M + n} = \sum_{M = -5/2}^{5/2} W_{M + n, M} =$$

and

$$\sum_{M = -5/2}^{5/2} M \cdot W_{M, M - n} = \sum_{M = -5/2}^{5/2} M \cdot W_{M - n, M} = \sum_{M = -5/2}^{5/2} (M' + n) \cdot W_{M', M + n} = \sum_{M = -5/2}^{5/2} (M + n) \cdot W_{M, M + n}.$$}

Here we take into account that $W_{M, M'} = 0$ if one of the indexes $M (M')$ is out of the $[-5/2, +5/2]$ range and $W_{M, n + M} = W_{n + M, M}, W_{M, n - M} = W_{M - n, M}$.

It follows that the Eq. (1) does not contain probability $W_{M, M'}$. It is also seen that the asymmetry parameter is proportional to the scattering order $n$. The Eq. (1) is deduced for the identical positions of polarizer and analyzer. In case if they are crossed the probabilities of the transitions become important and the difference in intensities of Stokes and anti-Stokes components may take place even without manganese polarization. For example, $\sigma^+$-photon creates the exciton with momentum projection $m = +1$ and after interaction with manganese it transforms into the state with $m = -1$ and subsequently emits a $\sigma^-$-photon. Simultaneously the manganese momentum projection increases by 2 so that photon frequency is shifted into the Stokes region. The anti-Stokes component for such scattering process does not exist at all. However this is related to selection rules and not to manganese polarization. In order to find the manganese polarization in case of crossed polarizer and analyzer it is necessary to compare the intensity of Stokes component in $\sigma/\sigma$ configuration with the intensity of anti-Stokes component in the opposite $\sigma/\sigma$ configuration, which is not always convenient from experimental point of view.

We emphasize that for $n > 1$ the Eq. (1) is valid only for a spin-flip of single manganese. It is not valid if there are two ions in the exciton localization region and each of these ions flips its spin resulting in the total projection change of $n$. Therefore the sample should be doped in a such way that the average number of Mn$^{2+}$ ($I = 5/2$) ions in the exciton localization volume is below 1. This corresponds to the case of GaAs doped with Mn.
main part of manganese are bound with the holes forming the neutral magnetic acceptor with the total spin $F = 1$. Due to the partial compensation of the acceptors with shallow donors ($N_D \sim 10^{16} \text{ cm}^{-3}$) the number of ionized acceptors (without the hole) is comparable with $N_D$. In this case the number of ions in the region of exciton localization (Bohr radius $a_X = 12 \text{ nm}$) is not larger than 1 because the parameter $\frac{4}{3}N_D a_X^3 \leq 0.1$. Note that the mentioned condition can be violated for the spin flip of magnetic acceptors ($F = 1$), which have significantly larger concentration of $N_A^0 = 8 \times 10^{17} \text{ cm}^{-3}$ in the studied samples. In exciton localization volume there are 2-3 of such acceptors, which allows the combined spin-flip processes.

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[16] SFRS is related to excitation of a $D^+X$ located closely to an ionized acceptor $A^-$. TRPL shows a long lifetime of the $D^0 - A^0$ transition, during which the electron undergoes several hundreds of jumps between the donors. On its way the electron can approach an ionized acceptor and stimulate the spin-flip transition. Therefore, spin polarization of $A^-$ ions will occur for both SFRS and TRPL. Note, that similar flip-flop process will take place at the neutral acceptors, whose number is approximately 60 times larger, although the exchange interaction with them is smaller due to screening effects originating from the anti-ferromagnetic interaction between a hole and a manganese spin. However SFRS signal related to $A^0$ is not observed in the studied sample.