Spin relaxation in diluted magnetic semiconductors: GaMnAs as example

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Abstract
The paper deals with a study of the magnetic impurities spin relaxation in the diluted magnetic semiconductors above the Curie temperature. Systems with a high concentration of magnetic impurities where magnetic correlations take place were studied. The proposed theory assumes the main channel of the spin relaxation being the mobile carriers, which provide the indirect interactions of the magnetic impurities. This theoretical model is supported by the experimental measurements of the manganese spin relaxation time in the GaMnAs by means of spin-flip Raman scattering. As has been found with a temperature increase the spin relaxation rate of the ferromagnetic samples grows, tending to that measured in a paramagnetic sample.

Keywords: spin relaxation, diluted magnetic semiconductors, GaMnAs

(Some figures may appear in colour only in the online journal)

1. Introduction
The diluted magnetic semiconductors (DMS) are a challenging system, since these functional materials combine the semiconductors and the magnetic properties [1, 2]. In order to construct, manipulate, and incorporate these materials in the conventional electronics, the mechanisms of a formation and a destruction of the magnetic order should be comprehended. By now, the magnetic ordering mechanism being well studied [3], the processes of the spin relaxation in the correlated systems are yet to be fully researched. The present paper will addresses this issue.

The nature of the ferromagnetism in the materials in question relates to the exchange interaction of the localized spins and the carriers [4–6]. The ferromagnetic interaction of the magnetic moments of impurities can be mediated either by the band electrons or by the holes as in CdMnTe [7], else by the localized carriers forming an impurity band as the GaMnAs. In the GaAs doped with the Mn, the $S = 5/2$ local moments of the Mn ions are exchange coupled with the holes provided by a substitution of the Ga with the Mn [8–12]. At the low doping regime, with the average distance between the ions exceeding a Mn acceptor radius, the exchange interaction of the manganese d shell electrons with a total spin $S = 5/2$ and the hole with an angular momentum $J = 3/2$ causes the splitting of the ground state into four levels with a total angular momentum $F = 1 \div 4$ [13, 14].

An increase in the Mn concentration leads to the overlapping of acceptor bound holes [15], causing a diffusion of the spins and the charges. Since it has happened, the indirect exchange interaction of the manganese by the holes determines the main magnetic properties of the system. Depending on the ratio of the average value of indirect exchange and its dispersion, the spin glass or a ferromagnetic ordering at low temperatures occurs [16]. There is no magnetic order at high temperatures ($T > T_C$) and subsystem of the magnetic ions can be considered as many body interacting system with the correlations. Another system with the similar properties is an ideal crystal with the free band carriers doped with the magnetic impurities. For a high concentration of the magnetic impurities the spin glass or the ferromagnetic ordering occurs, if the RKKY (Ruderman–Kittel–Kasuya–Yosida) temperature is higher than the Kondo temperature. The spin system of the magnetic centers and the delocalized carriers has been correlated in a paramagnetic phase as well. Therefore, the correlation of the magnetic ions is mediated by the mobile carriers in both systems. In addition, these carriers provide the main channel of the magnetic impurities spin relaxation. This paper...
analyzes the spin relaxation of the magnetic localized spins and describes its different regimes in the paramagnetic phase. The theory is supported by the experimental measurements of the manganese transversal spin relaxation rate in the GaMnAs with the Mn concentration up to a few percent including both the paramagnetic samples (with the Curie temperature lower than that of a liquid helium) and the ferromagnetic ones with the Curie temperatures of the dozens of kelvins. The spin flip Raman scattering (SFRS) technique as has been recently shown (see reference [17]) allows a direct measuring of a manganese spin relaxation time. The present paper elaborates the theory of the manganese spin relaxation for the ferromagnetic GaMnAs, taking into account the ferromagnetic and the paramagnetic phases. The main part of the manganese spin relaxation in the ferromagnetic phase results from the holes spin relaxation and the collective spin precession of the Mn d shell spins and the holes spins. With a temperature increasing, the linear increase of the Mn spin relaxation rate occurs in the paramagnetic phase, this being predicted theoretically and observed experimentally. However, no analysis of the paramagnetic samples with zero Curie temperature (lower than that of a liquid helium) has been made. As is shown in this paper, these types of samples with a similar Mn concentration demonstrate another behavior of the spin relaxation. It is also observed experimentally that the linear increase of the Mn spin relaxation rate is the first part of the spin relaxation behavior for the ferromagnetic samples in the paramagnetic region. The theory proposed describes these features of the spin relaxation in the paramagnetic region, linking the ferromagnetic samples with the pronounced Curie temperature and the paramagnetic samples with zero Curie temperature as well.

2. Samples and experimental setup

The samples, which are 500 nm-thick (Ga, Mn)As films, have been grown for the present study by the molecular-beam epitaxy on the GaAs (001) semi-insulating substrates covered with the 100 nm GaAs buffer layers. All the studied samples have not been subjected to annealing. The studied paramagnetic (PM) and four ferromagnetic (FM) samples have a Mn content of $x = 0.008$ (PM), and $x = 0.01, 0.014, 0.025, 0.043$ (FM), respectively. The Mn concentration was obtained from the lattice constant of the (Ga, Mn)As layer determined by high resolution x-ray diffraction [18]. To derive the Mn concentration, Vegard's law was applied, assuming the value of 0.598 nm for the lattice constant of 'zinc-blende-like MnAs, and neglecting the lattice-constant change due to the possible presence of AsGa point defects. Superconducting-quantum-interference-device (SQUID) measurements have been carried out with a temperature ranging from 5 to 400 K to confirm the absence of the MnAs nanoclusters in the FM samples. The Curie temperatures of the FM samples measured by the SQUID are summarized in Table 1 (for details also see reference [17]).

The SFRS spectra have been analyzed by the Jobin-Yvon U1000 double monochromator, equipped with a cooled GaAs photomultiplier and conventional photon counting electronics. The lines of He–Ne, Kr-ion and tunable Ti-sapphire lasers have been used to excite the SFRS. The laser power densities focused on the sample was about 50 W cm$^{-2}$. The experiments have been carried out at the temperature range of 1.6–200 K and the magnetic fields up to 5 T in the backscattering Voigt geometry. The Raman spectra have been recorded in the Voigt geometry in the $x(\sigma, \pi)\bar{x}$ polarization configuration, with $\bar{x}$ and $x$ perpendicular to the sample plane and the magnetic field $B$ directed along $\bar{z}$ and $\sigma, \pi$, denoting the linear polarizations of the exciting ($\sigma$) and the scattered ($\pi$) light with the electric field vector of the light being perpendicular (for $\sigma$) or parallel (for $\pi$) to $B$.

3. Experiment

The Raman spectra (see Figure 1) of two diluted (Ga, Mn)As samples: the paramagnetic PM ($x = 0.8\%$, solid magenta line) and FM1 ($x = 1\%$, solid blue line) recorded in a magnetic field $B = 5$ T at the temperatures $T = 2$ K show strongly polarized Mn-SFRS line with a magnetic field dependent Raman shift. These SFRS lines are active only in $x(\sigma, \pi)\bar{x}$ or $x(\pi, \sigma)\bar{x}$ configuration (with $\sigma||[001]$) and are forbidden in $x(\sigma, \pi)\bar{x}$ and $x(\pi, \sigma)\bar{x}$. The magnetic field dependence of the Mn-SFRS line energy shift in the PM sample can be represented as $\Delta = g_0^{Mn} \mu_B B$ with $g_0^{Mn} = 2.01$, which does not depend on the temperature. On the contrary the magnetic field dependence of the Raman shift for the SFRS line in the FM samples can be represented as $\Delta = g_0^{Mn} \mu_B B$ with $g_0^{Mn} = 2.01$ at $T$ above

| Sample number | Mn content (%) | $T_C$ |
|---------------|----------------|-------|
| PM            | 0.8            | —     |
| FM1           | 1              | 40 K  |
| FM2           | 1.4            | 48 K  |
| FM3           | 2.5            | 54 K  |
| FM4           | 4.3            | 55 K  |
the Curie temperature $T_C$ only. Below the $T_C$ $g$-factor of the FM sample depends on the temperature $g_{\text{Mn}}^0 = g(T)$ [17]. The SFRS linewidth is usually determined by the homogeneous and the inhomogeneous broadening. The inhomogeneous broadening of the SFRS line reflects the spread of the Mn ion $g$ factor, and therefore should depend on a magnetic field. Since the SFRS linewidth in the PM and FM samples does not depend on the magnetic field, see right panel of figure 2, it is concluded that the measured Mn SFRS linewidth is determined exclusively by the transversal (homogeneous) lifetime of the Mn spin subsystem. Therefore, the Mn spin subsystem lifetime can be determined by the SFRS linewidth. In the PM sample the SFRS linewidth is not sensitive to the wide range of temperatures as it is shown by deep blue squares in figure 1. To the contrary, the SFRS line width of the FM sample strongly depends on the temperature as is obvious by the comparison of the Raman spectra measured at $T = 2$ K (solid blue line in figure 1) and $T = 170$ K (solid red line). The temperature dependence of the SFRS linewidth measured at $B = 5$ T for a few FM samples is shown in the left panel of figure 2. In the FM1 sample the SFRS linewidth is insensitive to the temperature below $T_C$ and strongly increases at $T > T_C$. A further increase of the temperature ($T > 130$ K) leads to the linewidth saturation at a value close to the PM sample line width. The linewidth of FM samples with a higher Mn content demonstrates a similar to the FM1 sample behavior. However the Mn content increase reveals itself in decrease of the slope of the temperature dependence of the line width at $T > T_C$ and absence of the saturation at the high temperature range. Note that the SFRS signal strongly decreases in all samples at $T > 150$–$180$ K that does not allow to reach the saturation in the samples with the Mn content higher than 1.4%.

4. Theory

The structure in question is a bulk semiconductor doped with the magnetic impurities. The indirect exchange (RKKY) of the magnetic impurities occurs, provided there are delocalized carriers [4, 6]. The delocalized carriers may appear by the impurity band formation (for example if the magnetic impurities are donors or acceptors, as the GaMnAs) or there could be band electrons/holes. In general, the proposed theory can also be valid for the localized carriers, provided that their wave function overlaps with many magnetic impurities. With the increasing concentration of the magnetic impurities the indirect exchange interaction at low temperatures leads to the ferromagnetic transition [3].

The magnetic properties should be described by two magnetic subsystems with their own magnetization [17, 19]. One of them consists of the magnetic impurities spins $M_S(x, t)$ while the other consists of the delocalized carriers spins $M_J(x, t)$. These two subsystems interact by the exchange interaction with the constant $\lambda$. This method has been applied successfully to a study of the ferromagnetic resonance in the rare Earth garnets [19]. The magnetization dynamics of these two subsystems are analyzed to obtain a magnetic impurity transversal spin relaxation time [17]. For GaMnAs these subsystems are the Mn spins and the holes spins.

The research is focused on the paramagnetic region ($T > T_C$, $T_C$ is the Curie temperature), where the homogeneous magnetic field-induced magnetization is low. To determine the transversal spin relaxation, the homogeneous modes frequency of the magnetization precession in the magnetic field should be found. The theory in question should be phenomenological, only the main ingredients being taken into account. Thus, the simplest equations of the magnetization motion in the external magnetic field is given:

$$\frac{dM_S}{dt} = \frac{\mu g_S}{\hbar} B \times M_S - \frac{\lambda g_S}{\hbar} M_J \times M_S, \quad (1)$$

$$\frac{dM_J}{dt} = \frac{\mu g_J}{\hbar} B \times M_J - \frac{\lambda g_J}{\hbar} M_S \times M_J + D \Delta M_J - \gamma_J (M_J - M_J^0), \quad (2)$$

where $g_S$, $g_J$—are $g$-values of the magnetic ions and the delocalized carriers, $D$, $\gamma_J$—are the spin diffusion coefficient and the relaxation rate of the delocalized carriers, $M_J^0$—is the homogeneous magnetic field induced magnetization, $B$ is the external magnetic field applied along the $z$ axis. These equations may be supplemented with the magnetic anisotropy.

Figure 2. (Left panel) The temperature dependence of SFRS line width (proportional to the manganese spin relaxation rate) for the samples with a different Mn concentration, from $x = 0.8\%$ up to $x = 4.3\%$ measured in the external magnetic field $B = 5$ T. (Right panel) The magnetic field dependence of the SFRS line width for the samples with Mn concentration $x = 0.8\%$ and $x = 4.3\%$. 

field [20], the shape field, however for a large applied field they could be neglected. It is assumed that the magnetic impurities mainly interact with the delocalized carriers, other contributions like the spin-phonon, the spin–nuclear interactions etc being neglected. This assumption is justified as only the mobile carriers can mediate an interaction with the distant localized spins, simultaneously serving as a spin relaxation channel. The spin diffusion is governed by the carrier diffusion and the carrier spin–spin interaction. The spin relaxation is caused by the spin–orbit and the spin–spin interactions. Both of these parameters may depend on the temperature, the carriers, and the magnetic impurities concentrations. The magnetization of the magnetic impurities subsystem is presented as follows \( M_S = M_S^0 + M_S(t) \), where \( M_S^0 \) is the homogeneous magnetic field induced magnetization and \( M_S(t) \) is the small homogeneous deviation of magnetization precessing in the external field. For the delocalized carriers, the subsystem magnetization has an additional fluctuating term \( M_J = M_J^0 + M_J(t) + \delta M_J(x, t) \) since they are effectively coupled with the phonons.

The magnetic field induced magnetization in the paramagnetic phase is low compared to the external magnetic field \( \lambda M_J^0 \ll B \). Else the \( M_J^0 \) leads to a renormalization of the g-factor [19], as observed in the GaMnAs in the ferromagnetic phase [17]. The frequency of the homogeneous mode may be determined, as summed that its amplitude is infinitely small, i.e. less than the fluctuation \( \delta M_J(x, t) \gg M_J(t) \).

The following variables are introduced to simplify the equations of the motion of the magnetization: \( B_J = B_J/\lambda \), \( B_S = B_S/\lambda \), \( \lambda_S = \lambda_{S0}/\lambda \), \( \lambda_J = \lambda_{J0}/\lambda \), \( M^0 = M^0 + iM^f \). With these assumptions, the equation for the fluctuating part of the hole magnetization is:

\[
\frac{d\delta M_J}{dt} = B_J \times \delta M_J + D \nabla \delta M_J - \gamma_J \delta \dot{M}_J, \quad (3)
\]

and for the homogeneous parts of magnetization:

\[
\frac{dM_J^0}{dt} = -iB_J M_J^0 + i\lambda_J (M_J^0 + \delta M_J^0 )^* M_S^0 - M_J^0 M_S^0 \}
\]

\[
\frac{dM_J^0}{dt} = -iB_J M_J^0 + i\lambda_J (M_J^0 + \delta M_J^0 )^* M_S^0 - M_J^0 M_S^0 \}
\]

\[
\frac{dM_J^0}{dt} = -iB_J M_J^0 + i\lambda_J (M_J^0 + \delta M_J^0 )^* M_S^0 - M_J^0 M_S^0 \}
\]

There are two scenarios for the spin relaxation of the magnetic impurity depending on the rate of its rotation in the characteristic fluctuation field \( \lambda_J \delta \dot{M}_J \) and the time of the fluctuations dissipation \( \tau_{\text{diss}} \). For the strong fluctuations or with the slow dynamics \( \lambda_J \delta \dot{M}_J / \tau_{\text{diss}} \gg 1 \) the spin relaxation time of the magnetic impurities is determined by the dissipation time of \( \gamma_S \approx 1 / \tau_{\text{diss}} \). This equation is to be calculated.

Otherwise \( \lambda_J \delta \dot{M}_J / \tau_{\text{diss}} \ll 1 \) the dynamics of the fluctuations should be considered by integrating the equations (7), (4), then substituting them into the equation (6), and averaging them over the fluctuations. As a result:

\[
\frac{d\delta M^0_J}{dr} = -iB_J M^0_J (t) - \frac{\lambda_J^2}{2} \int d\tau \langle \delta M^+_J (x, t) \delta M^-_J (x, \tau) \rangle M^0_J (\tau). \quad (8)
\]

The equation for the frequency \( \omega_S = B_S - i\omega_0 \) may be obtained, where \( J(\omega) \) is the Fourier transform of the last part of (8). Assuming the spin are precess \( B_S > H_B \), the equation for the spin relaxation rate is

\[
\gamma_S \approx \Re \left\{ \frac{\lambda_J^2}{\pi^2} \int \frac{dk}{k} G(k, B_S) K(k) \right\}, \quad (9)
\]

where \( K(x) = \langle \delta M^+_J (x) \delta M^-_J (0) \rangle \) is the static fluctuation correlator, \( G(k, \omega) = [\omega^2 + \gamma_S - i(\omega - B_J)]^{-1} \) is the Green function of the transverse components of the equation (3), \( \Re \)—being the real part.

The correlator of the static fluctuations may be obtained by the phenomenological Hamiltonian by expanding it in a power series in \( \delta M_J \) to the quadratic terms:

\[
H_\text{eff}(\delta M_J) = C \frac{\delta M^\dagger_J \delta M_J}{\delta \chi_J \delta \chi_J} + \alpha \delta M^\dagger_J \delta M^0_J, \quad (10)
\]

where \( C \) is the square of the characteristic scale of the magnetic correlations; as it is known, this constant being proportional to the Curie temperature \( (C \sim T_C) \). Constant \( \alpha \) defines the magnetic susceptibility. For a correlator there is:

\[
K(x) = \int D[\delta M_J] e^{-\int dx H_\text{eff}(\delta M_J)/T} \delta M^\dagger_J(x) \delta M^0_J(0), \quad (11)
\]

\[
K(k) = \frac{T}{\pi(Ck^2 + \alpha)}. \quad (12)
\]

So, the magnetic impurities spin relaxation rate may be obtained:

\[
\gamma_S = \Re \left\{ \frac{\lambda_J^2}{8\pi D \sqrt{C \alpha} + C \sqrt{D \gamma_S - i(B_S - B_J)}} \right\}. \quad (13)
\]

The asymptotic for the high magnetic field \( |B_S - B_J| \gg D\alpha/C \) is \( \gamma_S \approx \lambda_J^2 T / 8 \pi C \sqrt{2D(B_S - B_J)} \). Since coefficients of this equation are almost independent of the temperature, it predicts a linear increase in the spin relaxation rate with the temperature. This tendency agrees with a linear increase in the relaxation rate in the paramagnetic phase with the temperature observed in the GaMnAs. Such a linear growth seems unrealistic though, especially at the high temperatures. It contradicts the observed saturation of the relaxation time of the Mn spin versus temperature for the GaMnAs samples as well, see the right panel of figure 1. The fluctuation correlator (12) has radius \( r_c = \sqrt{C/\alpha} \) and fluctuation power \( \int dx K(x) = T / 2 \alpha \) which grows infinitely with temperature. To obtain it, the increasing of number of delocalized carriers in the region with the scale \( r_c \) are needed. However, the process stops having reached a certain maximum value of the fluctuation; it is limited by the Coulomb interaction of the carriers. The maximum
fluctuation is a full spin of the delocalized carrier at its Fermi wavelength. For example, for the GaMnAs, it is a total spin of a hole on a manganese. To consider it, the fluctuation Hamiltonian (10) for the high values of $\delta M_1$ in a nonlinear regime should be found.

This is too complex a task, yet one may find the parameter describing the transition to the high temperature regime for the fluctuations in general. It is proposed to modify the equation for the correlator (11), taking into account the maximum fluctuations by replacing $\delta M_1 = M_0Y$, where the constant $M_0$ value limiting fluctuations is introduced. The maximum fluctuation is determined by a spin of the delocalized carrier located at the Fermi wavelength $\lambda = a/\sqrt{3} \sim M_0a^{-3/2}$. The correlator in $k$ space is

$$K(k) = M_0^2 \left[ \frac{D[Y]}{Y^2} \right] e^{-\int d^2k \delta(k)(M_0^2/T-G(Y))} = M_0^2 \left[ \frac{T}{M_0\epsilon_f}\right],$$

where $\epsilon_f(k)$ is a dimensionless energy of fluctuations. The limit for the integration is infinites and function $G(Y)$ play role of the limitation of the fluctuations. When $Ya^{-3/2} > 1$, $G \gg 1$.

The function $g$ defines the correlator and has the following asymptotic:

$$K(k) = \left[ \frac{T}{\epsilon_f(k)} \right] \rightarrow T \frac{e^{-C\gamma/a}}{r}, \quad M_0\epsilon_f(a^{-1}) \gg T,$$

$$K(k) = M_0^2 \rightarrow M_0^2 \delta(r), \quad M_0\epsilon_f(a^{-1}) \ll T.$$ (15) (16)

The example of the correlation calculations of equation (14) one can find in the appendix for a model cutoff. Thus, the general equation of the magnetic impurities spin relaxation rate is obtained:

$$\gamma_S \approx \frac{\lambda^2}{4\pi} Re \left\{ \int_{0}^{1/a} d\kappa k^2 \frac{1}{\left[Dk^2 + \frac{\gamma_j}{B_S - B_j} \right]} M_0^2 \right\}$$

$$\times g \left[ \frac{T}{M_0\epsilon_f}\right],$$

the integral being limited by the Fermi wavelength of the delocalized carriers $a$.

This equation describes various scenarios of the spin relaxation of the magnetic impurities in the DMS for the paramagnetic phase. For the ferromagnetic samples (with the Curie temperature higher than that of the liquid helium), there is a linear increase in the temperature (provided that all of the coefficients are close to the constant in the temperature), it gets a saturation then due to the attainment of the maximum fluctuation. The saturation temperature defined by $T_{sat} \approx M_0^2\epsilon_f(a^{-1})$ is proportional to the fluctuations energy band. The fluctuations energy band $\sim T_C$ is related to the Curie temperature. Thus, for the systems with a low Curie temperature, the spin relaxation rate of the magnetic impurities is in the saturation.

This system which are paramagnetic at all temperatures (with the Curie temperature being lower than that of the liquid helium) or the ferromagnetic samples in the saturation region (equation (17) for the high temperatures) have the strong fluctuations, since they reach the maximum value:

$$\gamma_S \approx \frac{\lambda^2}{4\pi D a^2} \gamma_j, \quad \delta M_1 \gg |B_S - B_j|,$$

$$\gamma_S \approx \frac{\lambda^2}{12\pi D a^3(\gamma_j^2 + |B_S - B_j|^2)}.$$

This equation is natural, it describes the fluctuation field of the spin $J$ acting on the spin of the magnetic impurities, the fluctuations dissipation time being the diffusion spreading of the $a^3$ region or the spin relaxation. In case of strong fluctuations or the slow dynamics $\delta M_1 \tau_{dis} \gg 1$, as mentioned earlier, the spin relaxation time of the magnetic impurities is determined by the fluctuations dissipation time $\gamma_S \approx 1/\tau_{dis}$. This time is the minimum of the relaxation time of the spin of the delocalized carriers or a diffusion propagation of fluctuations $\gamma_S \approx \min[\gamma_j, D/\max(C, a^2)]$. Note that in case of the slow dynamics, some other spin relaxation processes such as a hyperfine interaction may be important.

5. Discussion and comparison with the experiment

For the GaMnAs with the concentrations about atomic percent estimates show that $\delta M_1 \tau_{dis} \ll 1$ and the equation (17) for the manganese spin relaxation rate is to be used. The cutoff length $a$ in this DMS is determined by the localized hole radius. The maximum fluctuation is a hole spin on a manganese ion. For the paramagnetic samples with $x < 1\%$, the spin relaxation rate is independent of the temperature (left panel of figure 2), which is predicted by the equations (18) and (19). This saturation is related to the low Curie temperature in these samples. For the samples with $x > 1\%$, which have a Curie temperature of some tens of kelvin, the spin relaxation rate increases linearly with the temperature in the paramagnetic region and then tending to the saturation.

The differences and the similarities of the paramagnetic samples (with $T_C \rightarrow 0$) and the ferromagnetic ones in the paramagnetic phase need being stress. The spin relaxation rate at the saturation (see equations (18) and (19)), is determined by the parameters $M_0, D, \gamma_j$ which vary monotonically with a change of the concentration of the delocalized carriers and of the magnetic centers. Whereas the saturation temperature $T_{sat}$ proportional to the fluctuations energy band is defined by the collective effects, which are critically sensitive to a change in the concentration of the delocalized carriers and of the magnetic centers, leading to the large Curie temperature increase. This feature is clearly seen in the GaMnAs. Thus, for a sample with the manganese concentration $x = 0.8\%$ demonstrating the paramagnetic behavior at the temperatures above the liquid helium, the spin relaxation saturation regime is already achieved at the very low temperatures (see blue squares in left panel of figure 2). Whereas, for a sample with $x = 1\%$, which has an insignificant difference in the concentration with the
paramagnetic one, but has a pronounced ferromagnetic transition at $T_C = 35$ K, a linear increase in the spin relaxation rate with the subsequent saturation is observed (see red circles in left panel of figure 2). At the same time, the saturation values for these samples with similar concentrations differ insignificantly.

In figure 3 comparison of the experimental data from figure 2 (dots) and the model simulation (solid curves) of the spin relaxation rate in the paramagnetic phase for two samples with the different Curie temperatures and similar $l_D/a = 3, [B_S - B_J]/\gamma_J = 0.1$ is shown. These values are similar to those used for the GaMnAs as in reference [17]. The saturation temperatures are $T_{\text{sat}} = 3$ K for the PM sample and $T_{\text{sat}} = 130$ K for the FM1 sample. For the FM sample saturation temperature has same order as Curie temperature. For the paramagnetic sample there is no observed Curie temperature but the fluctuations energy band is estimated as the spin glass transition temperature observed for the samples with a similar concentration $x = 0.66$ as 7 K in [21]. A similar behavior for the Curie temperature has been observed as well, the sample with $x = 0.66$ having zero Curie temperature and the sample with $x = 0.87$ having the Curie temperature of 17 K, but close Mn concentration. The dependence of the function $g(T/T_{\text{sat}})$ used to fit the experimental data is a linear function as equation (15) up to $T_{\text{sat}}$, then a constant value as equation (16) with the sharp change from one dependency to another on scale $T_{\text{sat}}$.

### 6. Conclusion

To conclude with, the theory of the magnetic impurity spin relaxation in the DMS with the carriers mediating the ferromagnetism, while simultaneously providing their spin relaxation, has been elaborated. It presumes the magnetic correlations; therefore, it can be applied to the materials with the high concentration of the magnetic impurities. The theoretical model has been compared with the data of the manganese transversal spin relaxation time measured by the Raman spin-flip scattering in the GaMnAs. It is discovered that for the ferromagnetic sample in the paramagnetic phase the spin relaxation rate saturates with the temperature increase, tending to the temperature independent value measured in the paramagnetic sample with a slightly lower manganese concentration. This feature relates to the sharp changes in the spin fluctuations energy band and as a consequence to the Curie temperature.

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### Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

### Appendix A.

Here in the model calculation of the delocalized carriers magnetic fluctuations is demonstrated. The saturation effect may be taken into account phenomenologically by introducing a cutoff into the integration over the fluctuating field.

$$
\exp \left( - \int \frac{dx}{T} \frac{H_{\text{eff}}(\delta M_j)}{T} \right) \rightarrow \exp \left( - \int \frac{dx}{T} \frac{H_{\text{eff}}(\delta M_j)}{T} \right)
$$

$$
\int \frac{dx}{T} \frac{dM_j^2}{M_0^2}.
$$

(20)

The value $M_0$ limits the fluctuations, being the maximum fluctuation of the spin of the delocalized carriers, which can be found on any magnetic impurity. The model cutoff correctly takes into account general features of the correlator (15) and (16):

$$
K(r) = \frac{1}{8\pi C} \frac{T e^{-\sqrt{r/C}}}{r}, \quad M_0 \rightarrow \infty,
$$

$$
K(r) = M_0^2 \delta(r), \quad M_0 \rightarrow 0.
$$

For any temperature the correlator is:

$$
K(k) = \frac{T}{2(Ck^2 + \alpha + T/M_0^2)}.
$$

(21)

The equation of the magnetic impurities spin relaxation rate can be obtained by this model correlator:
\[
\gamma_S = \frac{\lambda^2}{4\pi^2} \Re \left\{ \int_0^{1/a} \frac{\text{d}k^2}{Dk^2 + \gamma J - i\gamma_B - B_J} Ck^2 + \alpha + T/M_0^2 \right\} = \frac{\lambda^2}{4\pi^2} \Re \left\{ \frac{T}{\gamma J} \frac{\alpha M_0^2 + T}{CM_0^2 l_D^2} \arctan \left( \frac{CM_0^2 l_D^2}{\alpha M_0^2 + T} \right) - \frac{1 - i\gamma_B - B_J/\gamma J}{\sqrt{1 - i\gamma_B - B_J/\gamma J}} \arctan \left( \frac{1 - i\gamma_B - B_J/\gamma J}{\alpha M_0^2 + T} \right) \right\},
\]

(22)

where \( l_D = \sqrt{D/\gamma_J} \) is the spin diffusion length. Note that this model correlator (21) may not accurately describe a transition from the linear temperature regime to the saturation value as the interactions of the fluctuations, which may be important to fit the experiment, has not been taken into account.

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