Carrier spin relaxation in diluted magnetic quantum wells: Effect of Mn spin correlations

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We demonstrate theoretically that the presence of holes, either resident or photocreated, in diluted magnetic quantum wells accelerates the spin relaxation of electrons via a mechanism which has been previously overlooked. This effect is due to the spin correlations, which establish between magnetic ions coupled via hole-mediated Ruderman-Kittel-Kasuya-Yoshida interactions in the paramagnetic phase. As a consequence, the electron spin relaxation becomes temperature and hole density dependent, in contrast to existing theories. Our theory qualitatively reproduces the increase of the electron spin relaxation rate with pump power observed in n-doped CdMnTe magnetic quantum wells [Ben Cheikh *et al.*, Phys. Rev. B **88**, 201306 (2013)]. It also predicts a decrease of the spin relaxation rate with temperature, as observed, although not in the same temperature range.

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I. INTRODUCTION

The physics of the nonequilibrium spin in semiconductors and semiconductor nanostructures has been extensively studied during the past decades, both experimentally and theoretically, boosted by the ideas of potential applications [1,2]. The density and temperature dependence of the electron spin relaxation time in nonmagnetic n-doped bulk semiconductors and quantum wells (QWs) is relatively well understood [3,4]. Depending on the density of electrons, either hyperfine (low density) or spin-orbit (high-density) interaction dominates the relaxation process [3]. Spin relaxation times of order of ∼100 ns have been achieved in bulk semiconductors with intermediate electron density close to metal-to-insulator transition, where a compromise between hyperfine and spin-orbit spin relaxation is achieved [5]. The spin relaxation in two-dimensional nanostructures is usually faster, and for the same reason as in bulk semiconductors the longest spin coherence times are obtained for weakly localized spins [6]. In the samples with the optimized electron density, the increase of the lattice temperature leads to additional delocalization of electrons, so that spin-orbit relaxation is enhanced via Dyakonov-Perel mechanism [7]. The increased optical pumping power has the same effect on the electron spin; the reason for this behavior is assumed to be the heating of the resident electrons by the photoexcitation [8].

Diluted magnetic semiconductors (DMSs), and their nanostructures eventually hosting a two-dimensional electron or hole gas, do not fit the scheme described above. In these materials the carrier spin relaxation is much faster than in their nonmagnetic counterparts. Such fast relaxation is a consequence of the exchange scattering with the magnetic dopants [9–13]. The role of the exchange scattering is also evidenced by the magnetic field dependence of the transverse electron spin relaxation time T_{2e}^*, measured by Kerr rotation experiments in Voigt geometry [10,13–16]. Indeed, T_{2e}^* exhibits a very characteristic nonmonotonous magnetic field dependence (at least at relatively low Mn concentrations and at liquid helium temperatures): an initial decrease at low fields up to 1–2 T, followed by an increase at higher fields. Recalling that inhomogeneous broadening mechanisms contribute to T_{2e}^*, this behavior can be consistently understood if the inhomogeneous heating of the Mn spin system induced by the laser pulses is taken into account [13,15]. As the temperature is increased the inhomogeneous heating is strongly suppressed, and one observes a nearly field-independent T_{2e}^* [13].

Studies of the temperature dependence of the electron spin relaxation time in magnetic QWs are scarce but indicate either almost constant spin relaxation time [10,11], or even weakly increasing relaxation time with temperature in a regime of low manganese concentration [13]. The zero-field power dependence is also puzzling as the spin relaxation time may either decrease [13] or increase [10] with optical pump power.

Calculations of the electron spin relaxation time based on the Fermi golden rule fail to reproduce the experimental results, even in the simplest case of conduction band electrons confined in a CdMnTe quantum well. Indeed, calculated spin relaxation times are systematically about five time longer than the experimental values [12,13]. More elaborate theories based on quantum kinetic equations have been developed [17,18]. In the theory presented in Ref. [17] the electron spins were considered as a subsystem interacting with a bath of magnetic impurities. In Ref. [18] the authors established the quantum kinetic equations for the whole electron-impurity system, thereby including the electron-impurity spin correlations [18]. These theories allow calculating the magnetic field dependence of both the longitudinal and the transverse electron spin relaxation times. However, the calculated values do not differ at zero field from those obtained within the Fermi golden-rule approach. Shmakov *et al.* considered another electron spin relaxation process, which appears in the presence of a nonzero magnetization and of quantum well width fluctuations [16]. In such circumstances, in addition to the magnetic fluctuations, the electron spin experiences long-range fluctuations of the exchange field related to the quantum well width fluctuations. Being long range, these fluctuations can be more efficient for electron spin relaxation than the magnetic fluctuations. However, this mechanism is not effective in zero magnetic field and cannot fill the gap between the existing experimental data

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and theory. Recent quantum kinetic theory points towards the importance of non-Markovian effects, particularly in the case of excitons [19], which may explain the difference between theory and experiment at least in the case of nondegenerate QWs.

Concerning the hole spin relaxation, it has been clearly shown that the quantum confinement stabilizes the heavy-hole spin [10] and that the spin relaxation time decreases at elevated temperatures due to heavy-hole–light-hole mixing away from the Γ point [11]. Indeed, at the Γ point in absence of mixing the hole spin flip by *p-d* exchange scattering is strictly forbidden.

In this paper we point out the effect of manganese spin-spin correlations on carrier spin relaxation [20,21]. These correlations induced by holes via Ruderman-Kittel-Kasuya-Yoshida (RKKY) interactions lead to a novel contribution to the carrier spin relaxation, which is not captured by the Fermi golden rule. RKKY interactions mediated by holes are responsible for the carrier-induced ferromagnetism in DMSs and are known to be strongly temperature dependent [22]. Decreasing temperature leads to the buildup of correlations even well above the ferromagnetic transition. We show that these correlations are responsible for temperature and power dependence of the electron spin relaxation.

Electrons could also induce spin correlations between magnetic atoms. However, we note that ferromagnetic ordering has never been observed in *n*-doped QWs, in agreement with the Mermin-Wagner theorem, which forbids a ferromagnetic ordering at finite temperature in case of Heisenberg-like interaction [23]. Even if one argues that a ferromagnetic transition can exist in *n*-doped QWs due to spin-orbit interaction [24], the Curie temperature will be lower than the mean-field approximation value, itself being already quite low due to the small values of the *s-d* exchange constant α compared to the *p-d* exchange constant β (in CdMnTe $\alpha = 0.25\beta$) and of the electron effective mass. Hence, electron-induced Mn-Mn spin correlations are expected to be quite small and can be safely neglected.

Let us outline that the enhancement of the carrier spin scattering due to hole-induced magnetic correlations in the Mn spin system has a well-known analogy in the physics of phase transitions. Indeed, the enhancement of the light scattering in the vicinity of the gas-to-liquid transition takes place due to the enhancement of the density fluctuations in this critical regime. This phenomenon is called critical opalescence. In the DMS spin system, the role of the light is played by electron or hole spin, and the enhanced spin scattering is due to the critical magnetic spin fluctuations in the vicinity of the paramagnetic-to-ferromagnetic transition, instead of the density fluctuations.

In the following we will focus on spin relaxation of electrons. We will not consider hole spin relaxation in order to avoid complications due to the valence band mixing, but in principle, the theory can be extended to this case. For simplicity, the theory is developed for strictly two-dimensional systems.

The paper is organized as follows. The theoretical description of electron spin relaxation in the presence of Mn spin-spin correlation is given in Sec. II. Then in Sec. III we discuss how this theory may explain the existing experimental results, especially the observed power and temperature dependence of electron spin relaxation time in *n*-doped DMS QWs. Finally, our results are summarized in Sec. IV.

II. THEORY

A. Electron spin relaxation in the presence of Mn spin correlations

The Hamiltonian describing the exchange interaction between electron and manganese has the form

$$\hat{V} = \alpha \hat{\mathbf{S}} \cdot \hat{\mathbf{M}}(\mathbf{r}),\ \hat{\mathbf{M}}(\mathbf{r}) = \sum_i \delta(\mathbf{r}_i - \mathbf{r})\hat{\mathbf{J}}_i, \tag{1}$$

where $\hat{\mathbf{S}}$ and $\hat{\mathbf{J}}_i$ are electron and manganese spin operators, respectively, and $\mathbf{r}_i$ are manganese 2D coordinates. The von Neumann equation describing the time evolution of the electron density matrix can be written in the basis of plane waves as

$$\frac{\partial \hat{\rho}_{kk'}}{\partial t} = -\frac{i}{\hbar}(E_k - E_{k'})\hat{\rho}_{kk'} - \frac{i}{\hbar}\sum_p (\hat{V}_{kp}\hat{\rho}_{pk'} - \hat{\rho}_{kp}\hat{V}_{pk'}). \tag{2}$$

We consider typical DMSs where the manganese concentration n_{Mn} is much larger than the electron concentration n_e. Therefore, one can assume the Mn spins to be static. In the case of small perturbation, the off-diagonal matrix elements of the density matrix are much smaller than the diagonal matrix elements. Hence, one can solve Eq. (2) for the off-diagonal elements omitting terms $V_{kp}\rho_{pk'}$ and then reintroduce the calculated off-diagonal elements in the equation for the diagonal elements. After Fourier transform we get

$$-i\omega\hat{\rho}_{kk}(\omega) + \frac{i}{\hbar}[\hat{V}_{kk}, \hat{\rho}_{kk}] = -\frac{2\pi}{\hbar}\int d\mathbf{p}\,\delta(E_p - E_k) \times \left[\frac{\hat{V}_{kp}\hat{V}_{pk}\hat{\rho}_{kk}(\omega) + \hat{\rho}_{kk}(\omega)\hat{V}_{kp}\hat{V}_{pk}}{2} - \hat{V}_{kp}\hat{\rho}_{pp}(\omega)\hat{V}_{pk}\right]. \tag{3}$$

In Eq. (3) one can average over the manganese spatial and spin distribution assuming that average value of manganese spin is zero. Under this assumption, the linear α term disappears and the correlator of the exchange interaction reads

$$\langle \hat{V}_{kp}\hat{V}_{pk}\rangle = \alpha^2 \hat{S}_i \hat{S}_j \int d\mathbf{r} e^{i(\mathbf{p}-\mathbf{k})\mathbf{r}} \langle \hat{M}_i(0)\hat{M}_j(\mathbf{r})\rangle. \tag{4}$$

The Mn spin-spin correlator $G_{ij}(\mathbf{r}) \equiv \langle \hat{M}_i(0)\hat{M}_j(\mathbf{r})\rangle$ in this equation is assumed to be translation invariant; i.e., it depends only on the differences between space coordinates. For electron spin $\mathbf{S_k} = \mathrm{Tr}(\hat{\mathbf{S}}\rho_{kk})$ one can write the dynamic equation

$$\frac{\partial \mathbf{S_k}}{\partial t} = -\hat{\Gamma}(\mathbf{k})\mathbf{S_k}, \tag{5}$$

$$\hat{\Gamma}(\mathbf{k}) = \frac{\alpha^2 m_e}{2\hbar^3}\int d\mathbf{r} J_0(kr) e^{-i\mathbf{kr}} \times \left[\hat{\mathrm{I}}\,\mathrm{Tr}\{\hat{G}(\mathbf{r})\} - \frac{\hat{G}(\mathbf{r}) + \hat{G}^{\mathrm{T}}(\mathbf{r})}{2}\right], \tag{6}$$

where the superscript T stands for the transposition, m_e is the electron effective mass, and J_0 is the Bessel function of the first kind.

In the limit of uncorrelated manganese spins, the correlator in (4) becomes

$$G_0^{ij}(\mathbf{r}) = \left\langle \sum_{nm} \hat{J}_n^i \delta(\mathbf{r}_n) \hat{J}_m^j \delta(\mathbf{r}_m - \mathbf{r}) \right\rangle = \delta_{ij}\delta(\mathbf{r}) \frac{J(J+1)n_{\mathrm{Mn}}}{3}. \quad (7)$$

Inserting this correlator in Eq. (6) one obtains the well-known result for the electron spin relaxation rate due to the exchange interaction with magnetic impurities [17,25],

$$\gamma_0 = \frac{\alpha^2 J(J+1) n_{\mathrm{Mn}} m_e}{3\hbar^3}, \quad (8)$$

which is independent of both electron momentum and electron concentration.

Let us take into account the correlations between manganese spins. The interaction between manganese spins can be written as

$$\hat{V}_{JJ} = \sum_{n \neq m} \hat{J}_n^i B_{ij}(\mathbf{r}_n - \mathbf{r}_m) \hat{J}_m^j, \quad (9)$$

where $B_{ij}(r_n - r_m)$ represents one-half of the Mn-Mn interaction in order to take into account the double counting in the sum. The manganese concentration being much larger than the electron concentration, there is a large number of magnetic centers within an area with characteristic size of the order of the electron wavelength. One can thus replace the discrete Mn distribution by a continuous Mn spin distribution and consider $M_i(\mathbf{r})$ as a classical field,

$$V_{JJ}(\{\mathbf{M}\}) = \int d\mathbf{r} d\mathbf{r}' M_i(\mathbf{r}) B_{ij}(\mathbf{r} - \mathbf{r}') M_j(\mathbf{r}'). \quad (10)$$

Furthermore, we assume a Gaussian Mn spin distribution. The resulting correlator reads

$$\hat{G}(\mathbf{r}) = \frac{\int D[\mathbf{M}]\, \mathbf{M}(0)\mathbf{M}^{\mathrm{T}}(\mathbf{r}) e^{-A\int \mathbf{M}^2 dS} e^{-\frac{V_{JJ}(\{\mathbf{M}\})}{T}}}{\int D[\mathbf{M}]\, e^{-A\int \mathbf{M}^2 dS} e^{-\frac{V_{JJ}(\{\mathbf{M}\})}{T}}} = \hat{\mathrm{I}}\frac{\delta(\mathbf{r})}{A} - \frac{1}{A}\int \frac{d\mathbf{k}}{(2\pi)^2} e^{-i\mathbf{k}\mathbf{r}} \hat{B}(\mathbf{k}) [\hat{\mathrm{I}} A T + \hat{B}(\mathbf{k})]^{-1} \equiv \hat{G}_0 + \hat{G}_c, \quad (11)$$

where T is the lattice temperature, and the functional integral $\int D[M]$ is carried out over all possible magnetic configurations M. Hereafter we omit the argument $(\mathbf{r})$ of the correlation function. The normalization constant A is determined from the requirement that one must recover the correlator in Eq. (7) in the absence of interaction

$$\hat{G} \xrightarrow[B(k)=0]{} \hat{G}_0 = \hat{\mathrm{I}}\frac{\delta(\mathbf{r})}{A}, \quad A = \frac{3}{J(J+1)n_{\mathrm{Mn}}}. \quad (12)$$

Substituting (11) and (12) in (6) one obtains the electron spin relaxation rate caused by spin-flip scattering on correlated manganese spins,

$$\hat{\Gamma}(\mathbf{k}) = \gamma_0 [\hat{\mathrm{I}} + \hat{\mathcal{G}}(\mathbf{k})], \quad (13)$$

$$\hat{\mathcal{G}}(\mathbf{k}) = \frac{A}{2}\int d\mathbf{r} J_0(kr) e^{-i\mathbf{k}\mathbf{r}} [\hat{\mathrm{I}}\,\mathrm{Tr}\{\hat{G}_c\} - \hat{G}_c]. \quad (14)$$

Note that this spin relaxation rate depends on the electron wave vector, in contrast with the prediction of the Fermi golden rule.

B. Hole-mediated RKKY correlations

Let us apply the results of Sec. II A to the hole-mediated correlations of magnetic impurities or so-called RKKY correlations [20,21]. We consider that the interaction between magnetic ions is isotropic in space and has a uniaxial spin-spin anisotropy. The corresponding Hamiltonian reads

$$\hat{V}_{JJ} = \sum_{n \neq m} B_\perp(r_{nm})\left(\hat{J}_x^n \hat{J}_x^m + \hat{J}_y^n \hat{J}_y^m\right) + B_\parallel(r_{nm}) \hat{J}_z^n \hat{J}_z^m. \quad (15)$$

We focus on the z component of electron spin relaxation, which depends only on $B_\perp(r)$; thus, we keep only the first term of Eq. (15).

This interaction is characterized by its strength B^* and by its spatial range r_i,

$$B^* = -\int d\mathbf{r} B_\perp(r), \quad (16)$$

$$r_i^2 = \frac{1}{2}\frac{\partial^2}{\partial k^2}\left[\frac{B_\perp(k=0)}{B_\perp(k)}\right]\Bigg|_{k=0}. \quad (17)$$

Here we defined B^* such that $B^* > 0$ to account for the ferromagnetic character of the RKKY interaction at short distance, and r_i can be identified with the hole coherence length and is given by $\frac{1}{r_i} \sim \frac{1}{\lambda_T} + \frac{1}{l_p}$, where l_p is the hole mean free path and $\lambda_T = h/\sqrt{2m_h k_B T}$ is the thermal length. The general expression for B^* is given by

$$B^*(n,T) = \frac{\beta^2 m_h}{4\pi\hbar^2} F\left(\frac{T}{T_F}\right), \quad (18)$$

where $T_{\mathrm{F}} = \pi\hbar^2 n_h/m_h$ is the Fermi temperature, and F is a function defined in the Appendix.

In the limiting case of degenerate (nondegenerate) hole gas $F(t) \to 1$ ($F(t) \to 1/t$), one gets

$$\text{degenerate:} \quad B^*(n,T) \sim \frac{\beta^2 m_h}{4\pi\hbar^2}, \quad (19)$$

$$\text{nondegenerate:} \quad B^*(n,T) \sim \frac{\beta^2 n_h}{4T}, \quad (20)$$

where m_h is the heavy-hole mass and n_h is the hole concentration.

In order to obtain a simple analytical expression for the electron spin relaxation rate, we use the approximation

$$B_\perp(r) = -\frac{B^*}{2\pi r_i^2} K_0\left(\frac{r}{r_i}\right), \quad (21)$$

where K_0 is the modified Bessel function of the second kind. This function has the expected asymptotic behavior for the RKKY interaction as $B_\perp \sim \ln(r)$, $r \to 0$ and $B_\perp \sim e^{-r/r_i}$, $r \to \infty$. Inserting Eq. (21) into Eqs. (11) and (14) one obtains the in-plane correlation function

$$G_c = \frac{B^*}{2\pi r_i^2 A^2 T} K_0\left(\frac{r}{r_c}\right), \quad (22)$$

where

$$r_c = r_i \sqrt{\frac{T}{T - B^*/A}}, \tag{23}$$

and the electron spin relaxation rate

$$\Gamma_z(k) = \gamma_0 \left[1 + \frac{B^*/A}{T - B^*/A} \frac{1}{\sqrt{1 + (2kr_c)^2}} \right]. \tag{24}$$

We can express this equation in terms of Curie temperature T_C, that is, the temperature at which the correlation length r_c in Eq. (23) diverges:

$$T_C - \frac{B^*(n, T_C)}{A} = 0. \tag{25}$$

For $T \ll T_F$ the hole gas is degenerate. We get

$$\Gamma_z(k) = \gamma_0 \left[1 + \frac{T_C}{(T - T_C)\sqrt{1 + (2kr_c)^2}} \right], \tag{26}$$

$$T_C^{FD} \sim \frac{\beta^2 m_h J(J+1) n_{\mathrm{Mn}}}{12\pi\hbar^2}, \tag{27}$$

$$r_c \sim (1/\lambda_T + 1/l_p)^{-1} \sqrt{\frac{T}{T - T_C}}, \tag{28}$$

while for $T \gg T_F$ the holes obey a Boltzmann statistics. We get

$$\Gamma_z(k) = \gamma_0 \left[1 + \frac{T_C^2}{(T^2 - T_C^2)\sqrt{1 + (2kr_c)^2}} \right], \tag{29}$$

$$T_C^B \sim \sqrt{\frac{\beta^2 n_h J(J+1) n_{\mathrm{Mn}}}{12}}, \tag{30}$$

$$r_c \sim (1/\lambda_T + 1/l_p)^{-1} \sqrt{\frac{T^2}{T^2 - T_C^2}}. \tag{31}$$

The relaxation rate given by Eqs. (26) and (29) includes two terms: a spin-flip term γ_0, which is the Fermi golden rule result, and a second term γ_c corresponding to the relaxation on the correlated Mn spin field. The latter can be understood as the result of the spin diffusion on correlated areas with characteristic size r_c. Indeed, an electron passes through such areas during an average time $\tau_{fl} = r_c/v_e$, where v_e is the electron velocity (ballistic regime, i.e., $r_c \ll l_p^e$, where l_p^e is the electron mean free path). In these correlated areas the average value of a Mn spin field is nonzero. As an example, in the case of Boltzmann distribution, its order of magnitude is given by the square root of the prefactor in Eq. (22): $M \sim \sqrt{J^2 n_{\mathrm{Mn}} T_C^2/(\lambda_T^h T)^2}$. So, electron spin rotates by the angle $\delta\varphi = \omega_{fl}\tau_{fl} \ll 1$ while passing this area. Using $\omega_{fl} \sim \frac{\alpha}{\hbar} M$ we obtain $\delta\varphi = \sqrt{\hbar\gamma_0/E_F^e T_C}/\sqrt{T - T_C}$. For temperatures not too close to T_C, $\delta\varphi \ll 1$, so that the electron spin diffusion coefficient reads $D_\varphi \sim \omega_{fl}^2 \tau_{fl}$. This contribution to the electron spin relaxation rate $1/\tau_s'$ can therefore be obtained from the condition $D_\varphi \tau_s' = 1$,

$$\frac{1}{\tau_s'} \approx \omega_{fl}^2 \tau_{fl}, \tag{32}$$

which identifies with the second term in (29).

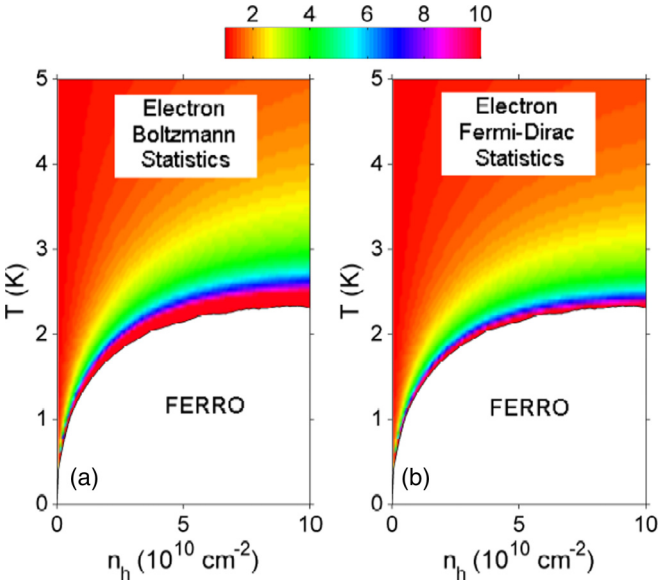

FIG. 1. 2D color maps of the enhancement factor $\Gamma_z(k)/\gamma_0$ ($m_h = 0.25m_0$, $m_e = 0.11m_0$, $\beta = 0.012$ eV nm^2, $n_{\mathrm{Mn}} = 1.8 \times 10^{14}$ cm^{-2}, and m_0 is the free electron mass). The white area corresponds to the ferromagnetic phase where the theory is not valid. (a) Boltzmann distribution of electrons; (b) Fermi-Dirac distribution ($n_e = 2 \times 10^{11}$ cm^{-2}).

C. Results

The above theory applies equally to two different experimental situations. First, it applies to n-doped QWs submitted to continuous-wave optical excitation, where Mn spin correlations are induced by the photoexcited holes. These correlations then impact the resident electrons spin relaxation. Second, it applies to p-doped QWs, where the resident holes imprint Mn spin correlations. These correlations then impact the spin relaxation of photoexcited electrons. These two cases are quite generally described by the same set of equations (23)–(25), which reduce either to Eqs. (29) and (30) when $T \gg T_F$ or to Eqs. (26) and (27) when $T \ll T_F$. In addition, the electron spin relaxation rate must be evaluated either at the thermal wave vector (electron Boltzmann distribution) or at the Fermi wave vector (electron Fermi-Dirac distribution).

The main results of the theory are gathered in Fig. 1, which shows color maps of the enhancement factor $\Gamma_z(k)/\gamma_0$ calculated in the plane (n_h, T) for $T > T_C$, and for two different electron distributions: first, in the case of a nondegenerate electron gas [Fig. 1(a)], $\Gamma_z(k)$ being evaluated at the thermal wave vector $k = k_T$; second, in the case of a degenerate electrons gas [Fig. 1(b)], $\Gamma_z(k)$ being evaluated at the Fermi wave vector $k = k_F = \sqrt{2\pi n_e}$ [26]. For the calculation we assume a short-range scattering mechanism in which the scattering time is constant ($\tau_p = 140$ fs, corresponding to a hole mobility $\mu_h = 1000$ V cm^{-2} s^{-1}).

The analogy with critical opalescence mentioned in the Introduction is clearly seen in Fig. 1. $\Gamma_z(k)$ increases when temperature approaches T_C and finally diverges at T_C, due to the critical magnetic fluctuations.

III. DISCUSSION AND COMPARISON WITH EXPERIMENT

The above theory applies quite naturally to the spin relaxation of photoexcited electrons in p-type diluted magnetic QWs [27]. Unfortunately, the experimental data are very scarce [12] and do not allow a test of the theory. On the other hand, there are more systematic measurements of electron spin relaxation in undoped or n-doped QWs [10,13]. In this case, one may expect to witness the influence of photoexcited holes on the electron spin relaxation. We focus on the spin relaxation in absence of external magnetic field to avoid the spin dephasing introduced by magnetic inhomogeneities [13]. In this case the calculated electron spin relaxation time $\tau_s^e = \Gamma_z(k)^{-1}$ can be identified with the measured T_{2e}^*.

The electron spin relaxation times have been measured by pump-probe time-resolved Kerr rotation experiments. Thus, whether the Mn spin correlations will settle or not depends on the relative values of the Mn spin relaxation time $T_1^{\rm Mn}$ and of the pulse repetition period T_p. If $T_1^{\rm Mn} \gg T_p$, spin correlations will establish at the same value as in a continuous-wave excitation of the same average power. On the contrary, if $T_1^{\rm Mn} \ll T_p$, spin correlations will decay between optical pulses and are not expected to be important.

Both of these situations have been encountered experimentally [10,13]. In Ref. [10] the sample consisted in ZnSe QWs containing several quarter-monolayer MnSe planes, resulting in relatively large average Mn concentrations, short $T_1^{\rm Mn} \leqslant 1$ ns, and very short electron spin relaxation time $\sim$1 ps. In addition, $T_p = 0.5$ μs is much larger than $T_1^{\rm Mn}$ and there is no cumulative effect. In these conditions the electron looses its spin before Mn spin correlations settle. It is unlikely that Mn correlations affect the electron spin relaxation in this case. In Ref. [13] the samples consisted in n-type modulation-doped CdMnTe QWs with much lower Mn concentration. The laser repetition period was $T_p = 12$ ns, and $T_1^{\rm Mn} \sim 10$ ns [28], so that the correlations will be somewhat weaker than in the continuous-wave regime. Nevertheless, we will still use cw excitation to handle this situation. In addition, in these samples the doping level was high enough to suppress excitonic effects by screening, thereby making it possible to study the electron spin relaxation not influenced by the binding into an exciton. This is in contrast with pump-probe experiments on undoped bulk CdMnTe, which revealed the exciton spin relaxation [14]. The measured electron spin lifetime is also weakly influenced by the optical pumping because, the density of carriers photoexcited by a pump pulse is much less than n_e, and second the recombination time $\tau_r \gtrsim \tau_s^e$. In these conditions the calculated τ_s^e can be directly compared to the experimental value [29].

Figure 2(a) shows how the calculated power dependence compares with the data for samples A and B taken from Ref. [13] (Sample A, $n_e = 3.4 \times 10^{11}$ cm^{-2}, atomic fraction of manganese $x = 0.0021$, QW width $L = 30$ nm; Sample B, $n_e = 1.9 \times 10^{11}$ cm^{-2}, $x = 0.0007$, $L = 30$ nm). The Fermi temperatures are, respectively 85 K and 47 K for these two samples. Hence, at the temperatures $T < 15$ K explored in Ref. [13] the electron gas is degenerate. The Mn-Mn correlations are induced by the photoexcited holes in these n-doped samples. We calculate $\Gamma_z(k_F)$ using Eq. (24) and

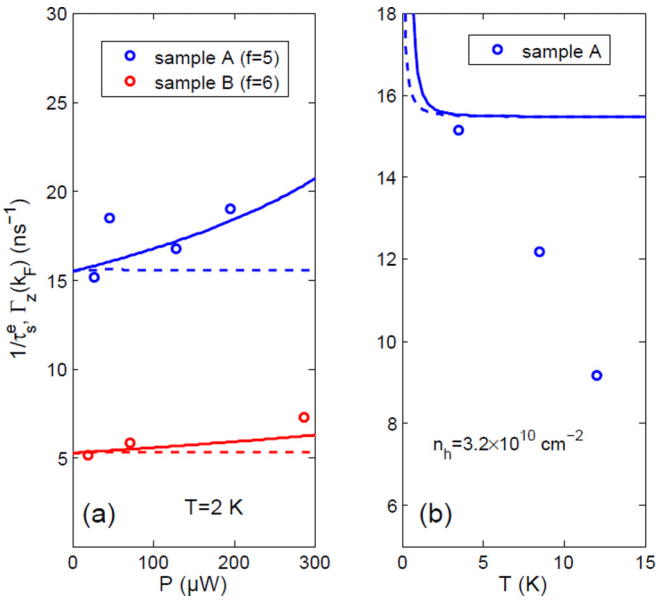

FIG. 2. Comparison between calculated (lines) and measured (circles) electron spin relaxation rates. Solid lines are calculated by taking into account the Mn-Mn correlations induced by photo-excited holes assuming either a Boltzmann statistics of holes [Eq. (20), solid lines], or a Fermi-Dirac statistics [Eq. (18), dashed lines]. In order to fit the experimental values (open circles from Ref. [13]) the calculated γ_0 [Eq. (8)] had to be multiplied by a correction factor f. (a): Power dependence of the electron spin relaxation rate for samples A and B. (b): Temperature dependence for sample A. The highest plausible hole density in the experimental conditions of Ref. [13] has been assumed in this calculation (all data from Ref. [13]).

introduce the correlations induced by a nondegenerate hole gas [Eq. (20), or using the general expression given by Eq. (18) (solid and dashed lines, respectively]. Note that because the holes are photoexcited they may have a temperature higher than the lattice. In absence of experimental data on hole mobility in CdMnTe QWs we treated τ_p as a fitting parameter. The increase of the observed spin relaxation rate is well reproduced by the calculation provided one assumes a nondegenerate hole gas and $\tau_p = 0.5$ ps (0.1 ps) for sample A (sample B). However, for the hole densities estimated from experimental conditions the general expression Eq. (18) should be used, because the hole gas becomes degenerate as the power increases. In these conditions the variation of the relaxation rate becomes very weak, because the critical temperature is almost independent of hole density. Note, however, that experimentally the effect is not very pronounced either, so that agreement can be considered as reasonable (note that the absolute value of τ_s^e was scaled by a factor $f \sim 5$ in order to account for the fact that the measured relaxation time is systematically shorter than the calculated one [12]).

Figure 2(b) shows the calculated and measured temperature dependencies of the spin relaxation rate. Experimental data correspond to sample A from Ref. [13]. The calculation does predict a decrease of relaxation rate with increasing temperature, as observed, albeit at a lower temperature. Clearly, the theory is not able to reproduce quantitatively the observed temperature dependence. One must assume that

another mechanism is responsible for the observed decrease of the relaxation rate. A more convincing test of the theory would be a comparison with temperature dependence of electron or hole spin relaxation in *p*-doped QWs, but systematic measurements are missing in this case [12].

IV. CONCLUSIONS

In conclusion, we have considered theoretically the influence of Mn spin correlations on the electron spin relaxation time in zero magnetic field in CdMnTe quantum wells. Such spin correlations may be induced by holes via RKKY interaction. At low temperatures an additional contribution to the spin relaxation rate comes from the fluctuating exchange field created by correlated Mn spins. As temperature increases, spin correlations weaken, and electron spin relaxation slows down to the value given by the temperature-independent single spin-flip processes. Increasing the optical pumping power, and thus the hole density, favors the buildup of spin correlations and thereby accelerates the electron spin relaxation.

We compared the predictions of the theory with available experimental data on *n*-doped CdMnTe QWs. We found some qualitative agreement but clearly the observed and predicted power dependence of spin relaxation rates are too weak to allow for a very reliable test of the theory. The observed temperature dependence is more pronounced but cannot be reproduced by the theory. We are led to the conclusion that the observed decrease of electron spin relaxation rate has a physical origin different from the mechanism considered in this paper. In addition, the quantitative mismatch, as large as a factor of five, between experimental and theoretical spin relaxation times is not solved by the proposed theory. Finally, a more relevant test of the theory would require systematic measurements of electron or hole spin relaxation in *p*-doped samples having higher Mn concentrations. This would permit measurements closer to the ferromagnetic transition, where a steeper acceleration of spin relaxation due to critical magnetic fluctuations is expected.

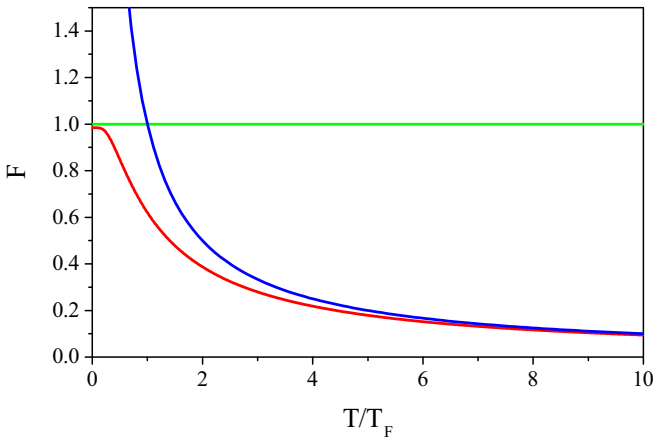

FIG. 3. Dependence of F on the reduced temperature T/T_F (red line). The green line corresponds to the Boltzmann limit $F \to T_F/T$ (for $T \gg T_F$); the blue line corresponds to the Fermi-Dirac limit $F \to 1$ (for $T \ll T_F$).

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APPENDIX

The RKKY interaction between magnetic ions in two dimensions has been discussed in Ref. [21] for degenerate carriers. In order to take into account the temperature dependence, one must use the more general formula [30,31]

$$B^*(r) = \frac{\beta^2 m_h}{4\pi\hbar^2}\int_0^\infty dk\, k J_0(kr) N_0(kr) \frac{1}{1+e^{\frac{E_k-\mu(T)}{T}}}, \quad \text{(A1)}$$

$$\mu(T) = T\ln\left(e^{\frac{T_F}{T}}-1\right), \quad \text{(A2)}$$

where J_0 and N_0 are, respectively, Bessel and Neumann functions of first and second kind and $\mu(T)$ is the chemical potential of carriers responsible for the RKKY interaction. In order to obtain the interaction strength B^*, one needs to integrate this equation on space coordinates

$$B^*(n,T) = \frac{\beta^2 m_h}{4\pi\hbar^2} F\left(\frac{T}{T_F}\right), \quad \text{(A3)}$$

$$F\left(\frac{T}{T_F}\right) = -2\pi\int_0^\infty dq\, q\int_0^\infty \frac{dx\, x J_0(qx) N_0(qx)}{1+e^{\frac{q^2 T_F}{2\pi T}-\frac{\mu(T)}{T}}}. \quad \text{(A4)}$$

The temperature dependence of function F is presented in Fig. 3.

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