

# Non-Markovian Effects in the Spin Transfer Dynamics in Diluted Magnetic Semiconductors due to Excitation in Proximity to the Band Edge

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**Abstract.** The non-Markovian effects in the spin dynamics in diluted magnetic semiconductors found in quantum kinetic calculations can be reproduced very well by a much simpler effective single electron theory, if a finite memory is accounted for. The resulting integro-differential equation can be solved by a differential transform method, yielding the Taylor series of the solution. From the comparison of both theories it can be concluded that the non-Markovian effects are due to the spectral proximity of the excited electrons to the band edge.

## 1. Introduction

Diluted magnetic semiconductors (DMS) are a class of workhorse materials in the field of semiconductor spintronics, since they combine the magnetic degree of freedom with the versatility and highly developed fabrication schemes of the semiconductor technology. Usually, Mn doped II-VI or III-V semiconductors are studied and a localized  $s$ - $d$  interaction between the carrier and Mn spins modelled by a Kondo-like Hamiltonian has been found to describe the magnetic properties and the spin dynamics of DMS very well.

A numerical calculation based on a quantum kinetic theory (QKT) for the spin dynamics in DMS governed by the  $s$ - $d$  interaction[1] showed that, among other phenomena, non-Markovian effects, such as overshoots or oscillations of the total spin polarization, can be found in one- and two-dimensional systems[2, 3]. The quantum kinetic theory can be presented in a more easy-to-use and intuitive way, by eliminating the correlations at the cost of a memory integral. Because it was found that, in doing so, it is crucial to account for a precession-like dynamics of the carrier-impurity correlations, the equations are referred to as *precession of electron spins and correlations* (PESC) equations[4]. In the present article, we show that the non-Markovian spin dynamics in DMS, found in the quantum kinetic theory, can be well described by an approximation of the PESC equations. The resulting integro-differential equation can be solved by a differential transform method (DTM)[5]. An analysis based on this simplified approach reveals that the non-Markovian effects are due to the proximity of the electronic excitations to the band edge.

## 2. Equation of motion

In Ref. [4] effective equations of motion for the correlation-induced spin dynamics in DMS were derived. For initially vanishing magnetization of the magnetic impurities, the time evolution of



the conduction band electron spin polarization in a DMS quantum well structure can be found from Eq. (7a) of Ref. [4]:

$$\frac{\partial}{\partial t} s_{\omega_1}(t) = -\frac{\eta}{\pi} \int_0^t dt' \int_0^{\omega_{BZ}} d\omega \cos[(\omega_1 - \omega)(t - t')] \left[ s_{\omega_1}(t') + \frac{1}{4}(s_{\omega}(t') - s_{\omega_1}(t')) \right], \quad (1)$$

where  $s_{\omega_1}$  is the mean electron spin of electrons with energy  $\hbar\omega_1$  (relative to the band minimum),  $\eta$  is the spin transfer rate in the Markov limit and  $\hbar\omega_{BZ}$  is the energy at the end of the first Brillouin zone. If we assume a parabolic band structure, we find  $\omega_1 = \frac{\hbar k_1^2}{2m^*}$  with effective mass  $m^*$  for an electron with wave vector  $\mathbf{k}_1$  and  $\eta = \frac{35}{12} \frac{J_{sd}^2 m^* n_{Mn}}{\hbar^3 d}$  with coupling constant  $J_{sd}$ , magnetic ion density  $n_{Mn}$  and quantum well width  $d$ .

It is noteworthy that in the time derivative for the total spin, where Eq. (1) is integrated over  $\omega_1$ , the term  $(s_{\omega}(t') - s_{\omega_1}(t'))$  cancels. Since this term can be expected to lead to an insignificant contribution to the total spin, we henceforth neglect this term which simplifies the analysis of the spin dynamics drastically. Despite this argument being valid only for the total spin, we shall show by numerical calculation that also the individual spin dynamics for an electron at the energy  $\omega_1$  is reasonably well described by this approximation (cf. Fig 1(c) and (d)). Thus, Eq. (1) can be reduced to

$$\frac{\partial}{\partial t} s_{\omega_1}(t) = -\frac{\eta}{\pi} \int_0^t dt' \left[ \frac{\sin((\omega_{BZ} - \omega_1)(t' - t))}{t' - t} + \frac{\sin(\omega_1(t' - t))}{t' - t} \right] s_{\omega_1}(t') \quad (2)$$

The physical meaning of Eq. (2) becomes most obvious when the Markov limit is regarded, which assumes that  $s_{\omega_1}$  changes on a much slower timescale than the oscillations of the integral kernel. Then, on the r. h. s. of Eq. (2),  $s_{\omega_1}(t')$  can be evaluated at  $t' = t$  and drawn out of the integral. Keeping in mind that  $\lim_{\omega \rightarrow \infty} \sin(\omega t)/t = \pi\delta(t)$  and that the integral ranges only over one half of the  $\sin(\omega t)/t$  peak, one finds:  $\frac{\partial}{\partial t} s_{\omega_1} = -\eta s_{\omega_1}$ , which shows a simple exponential decay of  $s_{\omega_1}$  with the rate  $\eta$ . This corresponds to a golden rule-type transfer of the electron spin to the impurity system.

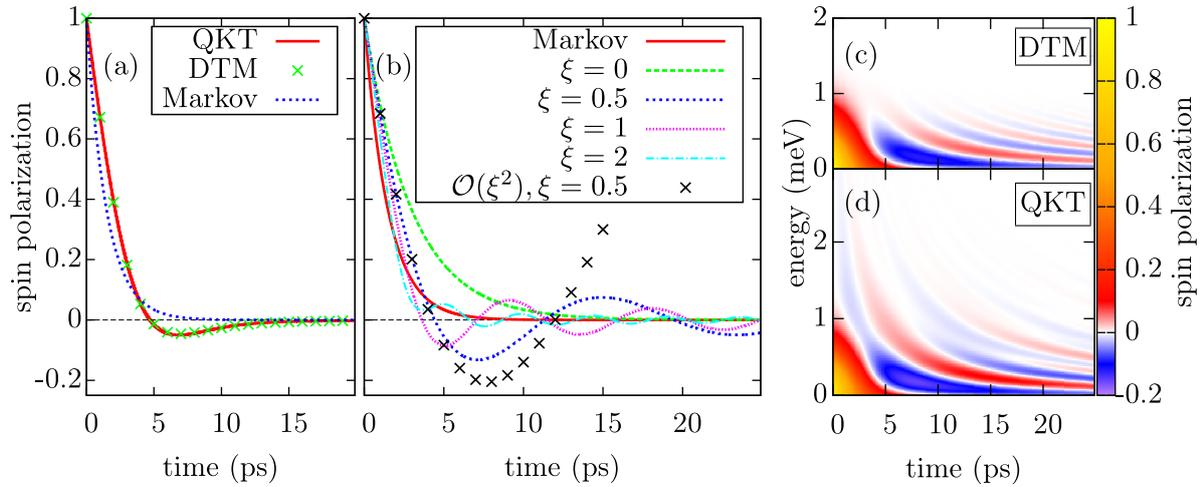
However, the condition for the applicability of the Markov limit was  $\eta \ll \omega_1$  and  $\eta \ll \omega_{BZ} - \omega_1$ . For realistic parameters (e. g., the parameters used in Ref. [2, 3] yield  $\hbar\eta \approx 0.45$  meV) and excitations far away from the end of the first Brillouin zone, only the latter condition is fulfilled, while for excitations close to the band edge,  $\omega_1$  can be of the same order of magnitude as  $\eta$ . Thus, we apply the Markov limit ( $\omega_{BZ} \rightarrow \infty$ ) only on the first term of Eq. (2). The number of parameters can be reduced by substituting  $\tau := \eta t$  and  $\xi := \omega_1/\eta$ . Then, the problem is transformed to:

$$\frac{\partial}{\partial \tau} \Phi_{\xi}(\tau) = -\frac{1}{2} \Phi_{\xi}(\tau) - \frac{1}{\pi} \int_0^{\tau} d\tau' \frac{\sin(\xi(\tau' - \tau))}{\tau' - \tau} \Phi_{\xi}(\tau'), \quad \Phi_{\xi}(0) := 1, \quad (3)$$

where  $s_{\omega_1}(t) = s_{\omega_1}(0) \Phi_{\omega_1/\eta}(\eta t)$ . Thus, the shape of the time evolution depends only on the ratio between  $\omega_1$  and  $\eta$ .

### 3. Numerical Evaluation of the non-Markovian Spin Dynamics

We solve the integro-differential equation (3) by a technique similar to Zhou's differential transform method (DTM)[5], which consists of Taylor-expanding all terms in Eq. (3) at  $\tau = 0$ . This yields



**Figure 1.** (a): Spin dynamics in a 4 nm wide  $\text{Zn}_{0.93}\text{Mn}_{0.07}\text{Se}$  quantum well with  $\eta \approx 0.67 \text{ ps}^{-1}$  with Gaussian excitaton at  $\hbar\omega = 0$  and standard deviation  $\Delta = 0.4 \text{ meV}$  (same as in Ref. [2]) according to the full quantum kinetic theory (QKT), the differential transform method (DTM, from Eq. (4)) and the Markov limit. (b): Time evolution of the spin of single electrons with fixed energies  $\hbar\omega = \hbar\eta\xi$ , compared with the Markov limit and the expression in Eq. (5) for the low- $\xi$  approximation. The spectrally resolved time evolution of the spin polarization is shown in (c) for the DTM calculation, and in (d) for the QKT (cf. Ref. [2]).

a recursion relation between the derivatives of  $\Phi_\xi$ :

$$\Phi_\xi^{(i)} = -\frac{1}{2}\Phi_\xi^{(i-1)} - \frac{1}{\pi} \sum_{0 \leq 2m \leq i-2} \frac{(-1)^m}{2m+1} \xi^{2m+1} \Phi_\xi^{(i-2-2m)}, \quad (4)$$

where  $\Phi_\xi^{(i)}$  is the  $i$ -th derivative of  $\Phi_\xi$  evaluated at  $\tau = 0$ . The numerical evaluation of Eq. (4) is very efficient and  $\Phi_\xi(\tau)$  can be calculated to high orders by substituting the derivatives into the Taylor expansion. We refer to this algorithm as the DTM calculation.

It is noteworthy that from the recursion relation (4) closed expressions can be derived for  $\Phi_\xi(\tau)$  to a certain order in the ratio  $\xi$  by combinatoric analysis of the paths from  $\Phi_\xi^{(0)} = 1$  to  $\Phi_\xi^{(n)}$  and comparing the Taylor series with that of known functions. E. g., to second order in  $\xi$ , we find:

$$\Phi_\xi(\tau) = e^{-\frac{\tau}{2}} + \frac{\xi}{\pi} [(2\tau + 4)e^{-\frac{\tau}{2}} - 4] + \left(\frac{\xi}{\pi}\right)^2 \left[ (2\tau^2 + 16\tau + 48)e^{-\frac{\tau}{2}} + 8\tau - 48 \right] + \mathcal{O}(\xi^3) \quad (5)$$

which should be valid for excitations near the band edge where  $\xi \ll 1$  can be fulfilled.

#### 4. Results

To check the validity of the approximation of neglecting the last term of Eq. (1), we compare the DTM calculation with the results of a full quantum kinetic treatment. Fig. 1(a) shows that the non-Markovian dynamics of the total spin given in Fig. 1(b) of Ref.[2] can be reproduced almost perfectly with the DTM calculation. Also, the time evolution of an individual spin of an electron with energy  $\hbar\omega_1$  is very similar in both calculations except for a high-energy tail appearing in the full quantum kinetic result, as can be seen from the spectrally resolved time evolution presented in Figs. 1(c) and (d) for DTM and QKT calculations, respectively. This finding confirms that Eq. (3) indeed captures the main non-Markovian features of the full quantum kinetic theory.

Fig. 1(b) shows the results of the DTM calculation for different values of  $\xi$ . For  $\xi = 0$ , the dynamics is given by an exponential decay with half the rate  $\eta$ , as can be seen also in the low- $\xi$  approximation in Eq. (5). For larger values of  $\xi$ , the decay rate approaches  $\eta$  and oscillations start to appear whose amplitudes eventually decrease for even larger values of  $\xi$ , where the time evolution converges to the exponential decay of the Markov limit ( $\omega_1 \rightarrow \infty$ ). Thus, the non-Markovian features are only present if the approximation  $\xi \gg \eta$  breaks down, i. e., if the excited electrons are spectrally close to the band edge, where the characteristic energy scale is given by  $\hbar\eta$ .

This can easily be understood if another derivation of the Markov limit starting from Eq. (1) is considered. If the assumption of a vanishing memory is made and on the r. h. s. the functions  $s_\omega(t')$  are evaluated at  $t' = t$ , we can first integrate over  $dt'$  and then over  $d\omega$ . Calculating the first interal gives

$$\frac{\partial}{\partial t} s_{\omega_1}(t) = -\frac{\eta}{\pi} \int_0^{\omega_{BZ}} d\omega \frac{\sin[(\omega_1 - \omega)t]}{\omega_1 - \omega} \left[ s_{\omega_1}(t) + \frac{1}{4}(s_\omega(t) - s_{\omega_1}(t)) \right]. \quad (6)$$

Using again the fact that  $\lim_{t \rightarrow \infty} \frac{\sin[\Delta\omega t]}{\Delta\omega} \rightarrow \pi\delta(\Delta\omega)$ , one again ends up with the Markov limit. For finite time  $t$ , however, the integral kernel is not yet contracted to a  $\delta$ -distribution and the finite integral limits cut off tails of the  $\frac{\sin[\Delta\omega t]}{\Delta\omega}$  function. This cut-off is particularly significant, if the peak of the integral kernel, which is given by  $\omega_1$  is close to one of the integral limits.

Furthermore, it can be seen in Fig. 1(b) that the low- $\xi$  approximation in Eq. (5) yields reasonable results for  $\xi = 0.5$  for the initial exponential decay while it fails to reproduce the long-term oscillations.

## 5. Conclusion

The non-Markovian overshoots and oscillations in the time evolution of the carrier spins in DMS found in a quantum kinetic theory can be reproduced by integro-differential equation of a much simpler form that also simplifies the interpretation considerably. A differential transform method (DTM) is employed to solve the resulting equation and allows to find closed-form expressions for low excitation energies of electrons.

It is found that a non-exponential behaviour of the time evolution of the electron spin is only present for electrons excited close to the band edge, where the decay predicted by the rate and the oscillations with frequency corresponding to the electron energies take place on the same time scale. Technically, this is due to the fact that a sinc-function that converges to a  $\delta$ -distribution in the Markov limit is cut off by the band edge. It is noteworthy that similar time evolutions have also been found in different setups, e. g., for the hole spin dynamics due to phonon scattering in a GaAs quantum well when the scattering rate is close to the phonon frequency[6].

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