Relaxation and coherent oscillations in the spin dynamics of II-VI diluted magnetic quantum wells

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Abstract. We study theoretically the ultrafast spin dynamics of II-VI diluted magnetic quantum wells in the presence of spin-orbit interaction. We extend a recent study where it was shown that the spin-orbit interaction and the exchange sd coupling in bulk and quantum wells can compete resulting in qualitatively new dynamics when they act simultaneously. We concentrate on Hg$_{1-x-y}$Mn$_x$Cd$_y$Te quantum wells, which have a highly tunable Rashba spin-orbit coupling. Our calculations use a recently developed formalism which incorporates electronic correlations originating from the exchange sd-coupling. We find that the dependence of electronic spin oscillations on the excess energy changes qualitatively depending on whether or not the spin-orbit interaction dominates or is of comparable strength with the sd interaction.

Ultrafast spin dynamics in semiconductors is attracting nowadays a great deal of attention. In a recent article we explored the interplay between the exchange sd interaction (EXI) and the spin-orbit interaction (SOI) in II-VI diluted magnetic semiconductors (DMS) [1]. In that study we found that the EXI and the SOI can be tuned to overrule each other or to compete on an equal level in realistic bulk and quantum well systems. Importantly, we found that the inclusion of the SOI introduces oscillations in the spin dynamics which are completely absent when only the EXI is relevant. In the present article, we characterize systematically the decay and the periods of oscillations seen in the spin dynamics in quantum wells, as functions of the excess energy of the electron population in the conduction band (mean energy of a Gaussian occupation of spin-polarized photoexcited electrons). We employ a microscopic density-matrix theory that models on a quantum-kinetic level the spin dynamics, taking into account the exchange-induced correlations and the localized character of the Mn spins [2, 3]. For the sake of brevity we shall only sketch the model here and refer the reader to Refs. [1] and [3] for a complete description.

We consider conduction band electrons in Mn-doped II-VI DMS coming from low-intensity optical excitations. We work in the regime of electron densities $n_e$ much lower than the Mn density $n_{Mn}$, in which the Mn spin variables are nearly stationary [3, 4, 5, 6]. The spin dynamics is described by $\langle s^\perp_k(t) \rangle$ and $\langle s^\parallel_k(t) \rangle$, the mean electronic spin components, perpendicular and parallel to the Mn magnetization, respectively, corresponding to the conduction-band state $k$.

In this study we concentrate on Hg$_{1-x-y}$Mn$_x$Cd$_y$Te quantum wells, since this alloy offers great flexibility in the control of the Rashba SOI. This control is achieved thanks to the strong dependence of the band gap on the doping fraction $x + y$ [7]. With this material, Rashba coefficients of the order of $\alpha_R \approx 10\text{ps}^{-1}\text{nm}$ can be obtained for realistic quantum well specifications [1]. Throughout the paper we shall assume the Mn magnetization to be perpendicular to the quantum well. Furthermore, we take a Gaussian electron occupation...
Figure 1. Electron spin dynamics in a Hg$_{1-x-y}$Mn$_x$Cd$_y$Te quantum well with Rashba interaction and no exchange sd coupling for a Gaussian electron distribution centered at $E_c$. Lines: simulations for different values of $\alpha_R$ expressed in units of ps$^{-1}$nm. Red dots: simple fits to the first oscillation of the curves with $\alpha_R = 4$ ps$^{-1}$nm (see text for details). centered at an energy $E_c$ above the conduction-band edge with a standard deviation of $E_s = 3$ meV and initial spin-polarization rotated 30° with respect to the Mn magnetization.

It is instructive to examine first the spin dynamics resulting only from the Rashba spin-orbit interaction (i.e., no exchange sd coupling is accounted for). In this case the Mn magnetization does not enter the dynamics. For later comparison we used, however, the above described initial condition where the direction of the initial electronic spin is related to direction of the Mn magnetization. Figure 1 shows the time evolution of the summed parallel spin component, $\langle s^\parallel \rangle(t) = \sum_k \langle s_k^\parallel \rangle(t)$, for three different values of the Rashba coupling constant $\alpha_R$. The cases $E_c = 0$ (Gaussian occupation centered at the band edge) and $E_c = 8$ meV are shown. We see that the Rashba interaction produces well-defined oscillations and decay. Note that without the EXI, the time evolution of the total spin is given by coherent precessions of individual electron spins around the $k$-dependent magnetic Rashba field, which are collectively responsible for the decay. Red dots in both panels of Fig. 1 are fits to the initial oscillations of the $\alpha_R = 4$ ps$^{-1}$nm evolutions, done with a function $f(t) \propto \exp\left[-(t/\tau)^2\right] \cos(2\pi t/T)$. For a Gaussian electron distribution with spins pointing in the growth direction, we find from Eq. (17) of Ref. [1]:

$$\langle s^\parallel (t) \rangle = C \int_0^\infty dk \, k \exp \left[ -\frac{(\hbar^2 k^2 - 2m^*E_c)^2}{(2m^*E_s)^2} \right] \cos(2\alpha_R kt), \tag{1}$$

where $m^*$ is the effective mass and $C$ is a constant determined by the initial value of the total spin. The integral in Eq. (1) is close to the (half-sided) Fourier transform of a function with a single central peak indicating in time regime a damped oscillation with roughly the peak frequency. An initially exponential decay of $\langle s^\parallel (t) \rangle$ would require a Lorentzian decay in the energy domain. However, the function in Eq. (1) decays much faster for large $k$ explaining why the initial behavior of $\langle s^\parallel (t) \rangle$ is much better approximated by a Gaussian than by an exponential. Indeed, Fig. 1 reveals that the Gaussian fit is almost perfect at early times but worsens somewhat later. Applying the Gaussian fit to a number of different cases we find that for given $E_c$ both $\tau \propto \alpha_R^{-1}$ and $T \propto \alpha_R^{-1}$ hold to a very good approximation. A similar behavior is observed for $|\langle s^\perp (t) \rangle| = |\sum_k \langle s_k^\perp \rangle(t)|$, whose initial evolution can be well fitted with...
Figure 2. Gaussian electron spin relaxation time, $\tau$, and period of the oscillations, $T$, in a Hg$_{1-x-y}$Mn$_x$Cd$_y$Te quantum well with Rashba interaction $\alpha_R = 4 \text{ps}^{-1}\text{nm}$ and no exchange sd coupling as a function of the excess energy $E_c$. Symbols: full calculation; lines: parabolic fit.

Figure 3. $\langle s^\parallel(t) \rangle$ with exchange sd and no Rashba interaction. $x_{\text{Mn}} = 0.3\%$, $S = 0.1$, $E_c = 0$ (red line), 4 meV (green circles), 8 meV (blue squares). Inset: $E_c = 0$, $S = 0.1$, various $x_{\text{Mn}}$: 0.1% (red), 0.2% (green), 0.3% (blue), 0.4% (magenta), 0.5% (cyan).

$$f(t) \propto \exp\left[-\left(t/\tau\right)^2\right] \cos(2\pi t/T) + 1,$$

with the same values of $\tau$ and $T$ as for $\langle s^\parallel(t) \rangle$. We found that $\tau$ and $T$ can be precisely fitted as functions of $E_c$ with parabolas, as seen in Fig. 2. The decrease of $T$ and the increase of $\tau$ with rising $E_c$ reflect the fact that the effective Rashba field is $k$ dependent becoming stronger for larger $k$.

Let us now look at the spin dynamics under the influence of the EXI only (no Rashba SOI). For the EXI coupling constant we take the value $J_{\text{sd}} = 26.8 \text{meV} \text{nm}^3$ [1]. The effects of the EXI on the carrier spins can be controlled via the Mn concentration, $x_{\text{Mn}}$, and the initial net Mn magnetization, $S = |\langle S \rangle|$ [1]. The dynamics of the electron spin component parallel to the Mn magnetization is shown in Fig. 3. As found previously, it is approximately described by an exponential decay to an in general non-zero equilibrium value with a decay time [cf. Eq.(19) of Ref. [3]] $\tau^\parallel = (J_{\text{sd}}^2 r_{\text{Mn}} m^* /\hbar^2 d) \langle S^2 - S^\parallel^2 \rangle$, where $d$ is the width of the quantum well and $\langle S^2 - S^\parallel^2 \rangle$ is a second moment of the spin-$\frac{5}{2}$ Mn system. In particular, $\tau^\parallel$ is linear in $x_{\text{Mn}}$ and independent of the excess energy $E_c$. Note that no oscillations appear in the time evolution of the parallel spin component when only the EXI is present. The perpendicular component decays to zero with a slightly different rate while it precesses around the Mn magnetization [3].

We now come to the combined effects of the Rashba SOI and EXI. Figure 4 shows the spin dynamics for $x_{\text{Mn}} = 0.3\%$, $S = 0.1$, $\alpha_R \approx 4 \text{ps}^{-1}\text{nm}$, and three different values of $E_c$. Note the semilog scale chosen to better visualize the long time behavior. This set of parameters defines a “strong sd” and “weak Rashba” situation. Accordingly, the initial decay is exponential (not Gaussian like in Fig. 1), with an $E_c$-independent decay rate, like in Fig. 3. However, at later times fairly regular oscillations appear, with essentially constant, $E_c$-dependent amplitude, due to the presence of the Rashba interaction. We note that the frequency of the oscillations depends only slightly on $E_c$, becoming higher for higher $E_c$, which indicates a slight dependence on the Rashba mechanism. The frequency of the oscillations (period of about 35 ps) is close to the precession frequency about the net Mn magnetic field, which is independent of $E_c$. The oscillations do not decay because all electrons precess with nearly the same frequency governed mainly by the Mn magnetization and thus do not dephase. The fact that the amplitude of the oscillations does not
decay with time and the near independence of the period on $E_c$ distinguish qualitatively these oscillations from the ones observed with Rashba coupling alone. Also note that again there is a saturation value different from zero as seen above in the sd-only case. However, a new feature produced by the presence of the Rashba interactions is that the perpendicular spin component does not go to zero as in the sd-only evolution (not shown for brevity).

Finally, Fig. 5 shows the time evolution of the mean perpendicular spin component under EXI and SOI for fixed Rashba constant $\alpha_R = 16\text{ps}^{-1}\text{nm}$, $E_c = 8\text{meV}$, and three different values of $x_{\text{Mn}}$. This figure shows the effect of an increasing EXI coupling in the presence of a strong Rashba coupling on the perpendicular spin component. We see that as the Mn concentration increases, starting from a Rashba-only situation in which the equilibrium value $\langle s^z_k \rangle$ is half its initial value [1], a decay to zero sets in. At the same time, the frequency of the oscillations increases and their amplitude goes down.

In conclusion, we have studied theoretically the effects of the Rashba spin-orbit interaction in II-VI diluted-magnetic-semiconductor quantum wells. We characterized the dependence of the spin dynamics on the excess energy of a Gaussian population of electrons in the conduction band. Our findings provide qualitative signatures that could aid experimentalists in distinguishing the relative importance of spin-orbit and exchange interactions in DMS quantum wells.

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