

Spin-flip relaxation time of conduction electrons in $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ quantum wells

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We present theoretical calculations of the spin-flip relaxation time of conduction electrons in $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ quantum wells. The spin-flip scattering arises from the s - d exchange interaction between the conduction and localized electrons. The scattering efficiency is larger with the localized spins in the well than in the barrier, typical time scales being tens of picoseconds and nanoseconds, respectively. A biased double-quantum-well structure should provide an ideal means for controllable change of the spin-flip scattering time.

Recent developments in the physics of semiconductor heterolayers have included the successful growth and preliminary studies of $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ quantum wells and superlattices.¹ In these materials there exists, apart from the regular quantum size effects (size-induced blue shift of the band gap), a whole class of new effects brought about by the presence of localized magnetic moments ($S = \frac{5}{2}$) arising from the half-filled d shell of the Mn^{2+} ions. The interplay of the magnetic properties and of the size quantization has only begun to be explored. Recent time-resolved measurements of the Mn^{2+} magnetization² due to the creation of spin-polarized photoelectrons, or conversely of the time-resolved decay of the photoluminescence polarization,³ point out the existence of a very fast decay of the spin alignment of the conducting electrons. Band-structure-induced spin-flip scattering mechanisms (Elliott-Yaffet,⁴ D'yakonov-Perel⁵) appear unlikely to be the cause of such a fast decay, basically because $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ alloys are wide-gap materials with little valence-band admixture in the conduction-band eigenstates away from the zone center. It is the purpose of the present work to report on calculations of the spin-flip scattering time of conduction electrons in $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ - $\text{Cd}_{1-y}\text{Mn}_y\text{Te}$ quantum wells due to the s - d exchange interaction. To our knowledge, calculations of such interactions have only been undertaken in the case of bulk $\text{Hg}_{1-x}\text{Mn}_x\text{Te}$ or $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ alloys.⁶ Our goals are to obtain a theoretical estimate of the influence of the size quantization on the spin-flip processes to assess the order of magnitude of the spin-flip scattering time, and in particular to establish its functional dependence on parameters such as the well thickness and the Mn mole fraction under specific intrasubband and intersubband transitions.

The Hamiltonian of a conduction electron in a $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ - $\text{Cd}_{1-y}\text{Mn}_y\text{Te}$ quantum well at zero magnetic field is written as

$$H = H_{\text{QW}} - \sum_{\mathbf{R}_i} J(\mathbf{r}_i - \mathbf{R}_i) (S_i^z \sigma^z + \frac{1}{2} S_i^+ \sigma^- + \frac{1}{2} S_i^- \sigma^+). \quad (1)$$

$$W_{n \rightarrow n'} = \sum_{\mathbf{R}_i} W_{n \rightarrow n'}(\mathbf{R}_i), \quad (4)$$

$$W_{n \rightarrow n'}(\mathbf{R}_i) = \frac{m^* J^2}{2\hbar^3 A} \chi_n^2(z_i) \sum_{n'} \chi_{n'}^2(z_i) \left\{ \left(\frac{3}{4} - \sigma_z^2 \right) \left(\frac{35}{4} - \sigma_z^2 \right) \left[\frac{35}{4} - (S_i^z)^2 \right] - \sigma_z S_i^z \right\} Y \left(E_n + \frac{\hbar^2 k_{\perp}^2}{2m^*} - E_{n'} \right), \quad (5)$$

In Eq. (1), H_{QW} is the quantum-well effective Hamiltonian acting on the envelope function whose eigenstates are of the form

$$\Psi_{n\mathbf{k}_{\perp}} = \frac{1}{\sqrt{A}} \exp(i\mathbf{k}_{\perp} \cdot \mathbf{r}_{\perp}) \chi_n(z), \quad (2)$$

where A is the sample area, $\mathbf{k}_{\perp} = (k_x, k_y)$, and $\mathbf{r}_{\perp} = (x, y)$. $\chi_n(z)$ is an eigenfunction of

$$\left[\frac{P_z^2}{2m^*} + V_b(z) \right] \chi_n(z) = E_n \chi_n(z). \quad (3)$$

In Eq. (3), $V_b(z)$ is the confining potential energy which is zero in the wells ($\text{Cd}_{1-x}\text{Mn}_x\text{Te}$) and equal to V_b in the barriers ($\text{Cd}_{1-y}\text{Mn}_y\text{Te}$). The conduction-band discontinuity is not very well known in this material system. It is however, believed to be large and in the following we will take V_b as equal to $0.9(y-x)\Delta E_g$, where ΔE_g is the band-gap energy difference between CdTe and the hypothetical cubic MnTe ($\Delta E_g \sim 1.6$ eV).

The exchange integral $J(\mathbf{r} - \mathbf{R}_i)$, appearing in the Heisenberg-like term in Eq. (1), is nearly localized in the unit cell which contains the localized spin S_i . It therefore acts as a contactlike interaction with a strength J when acting on the envelope function $\Psi_{n\mathbf{k}_{\perp}}$. We are interested in evaluating the spin-flip relaxation time $\tau_{n\mathbf{k}_{\perp}\sigma_z}$ of a conduction electron whose spatial eigenstate is $\Psi_{n\mathbf{k}_{\perp}}$ and whose spin component along the growth axis is σ_z . To do so, we first compute the transition rate $W_{n \rightarrow n'}$ from an initial state $|n, k_{\perp}, \sigma_z, S_i^z, S_i^z, \dots, S_i^z\rangle$ to all the final states $|n', k'_{\perp}, \sigma'_z, S_i^z, S_i^z, \dots, S_i^z\rangle$ of the (electron + spins) global system due to the spin-flip part of the Heisenberg-like term at the Born approximation. This transition rate is then averaged over the random positions of the Mn^{2+} ions as well as over the initial states of the localized spins. Since there is no applied field, the average of S_i^z is zero and that of $(S_i^z)^2$ is $\frac{1}{3} S(S+1) (= \frac{35}{12})$. More specifically we have obtained

where $Y(x)$ is the step function [$Y(x) = 1$ if $x > 0$, $Y(x) = 0$ elsewhere], and

$$\frac{1}{\tau_{nk_{\perp}\sigma_z}} = \sum_{n'} \frac{1}{\tau_{n \rightarrow n'}}, \quad (6)$$

$$\frac{1}{\tau_{n \rightarrow n'}} = \frac{35}{24} \frac{m^*}{\hbar^3} J^2 Y \left(E_n + \frac{\hbar^2 k_{\perp}^2}{2m^*} - E_{n'} \right) \sum_j C_j \int_{\Omega_j} dz_j \chi_n^2(z_j) \chi_{n'}^2(z_j). \quad (7)$$

$\tau_{nk_{\perp}\sigma_z}$ is σ_z independent as expected since $B=0$. In Eq. (7), C_j is the volume concentration of Mn^+ ions in the j th layer (well, left barrier, right barrier). One recognizes in Eq. (7) an expression which is very similar to that obtained for alloy scattering.⁷ This is to be expected since in both cases the scatterers are of very short ranges. Therefore, the functional dependence of the scattering time upon the quantum-well thickness L or barrier height V_b should be very similar. We also expect a significant difference in the magnitude of the spin-flip relaxation times, depending on whether the scattering takes place in the well or in the barrier.

Let us first analyze the dependence of the spin-flip scattering time upon the quantum-well thickness of the intrasubband transition in the ground subband [$n=n'=1$ in Eq. (7)] in the case when scattering takes place in the quantum well. In the limit of very small L , $\chi_1(z)$ leaks heavily into the barrier and the short-range scatterers, all localized in the well, become less efficient: $\tau_{1 \rightarrow 1}$ diverges when L goes to zero. Similarly, in the limit of large L , we expect a divergency of $\tau_{1 \rightarrow 1}$. This is because the scattering matrix element for a given z_i becomes proportional to L^{-1} which, when squared and averaged over all the quantum-well sites z_i , leads to the $\tau_{1 \rightarrow 1}$ proportionality with L . Notice that the discreteness of the z eigenstates $|E_n\rangle$ is an important factor in that a similar calculation for the bulk material would result in a \sum_j in Eq. (7) which is L independent, due to the degeneracy ($L/2\pi$) of the final states.

A reasoning similar to that given above leads to the conclusion that any intrasubband term $\tau_{n \rightarrow n}$ (intersubband term $\tau_{i \rightarrow j}$ with $j > i$) increases linearly with L at large L and diverges when L approaches L_n (L_j) from above, where L_n (L_j) is the critical thickness at which E_n (E_j) is no longer confined in the well. In fact, in the limit of large L , both $\tau_{n \rightarrow n}$ and $\tau_{i \rightarrow j}$ approach their asymptotic values, which can be evaluated by using for χ 's in Eq. (7) the eigenfunctions of a quantum well with infinite barrier:

$$\frac{1}{\tau_{n \rightarrow n}} = \frac{35}{16L} \frac{m^* J^2}{\hbar^3} C_{\text{well}}, \quad (8)$$

$$\frac{1}{\tau_{i \rightarrow j}} = \frac{35}{24L} \frac{m^* J^2}{\hbar^3} C_{\text{well}} Y \left(E_i + \frac{\hbar^2 k_{\perp}^2}{2m^*} - E_j \right), \quad (9)$$

where C_{well} is the volume concentration of localized spins in the well material.

In order to numerically evaluate the various scattering times we have used the following parameters: $m^* = 0.096m_0$ and $J = (0.22 \text{ eV}) \times a_0^3/4$ (Ref. 8), where $a_0 = 6.487 \text{ \AA}$. The volume concentration of spins in the well should, in a strict Born approximation, be taken to be equal to $4x_{\text{well}}/a_0^3$, where x_{well} is the Mn mole fraction in

the well. However, it is known that the Mn spins couple antiferromagnetically with a nearest-neighbor coupling constant of a few degrees. Thus, at low temperatures, the nearest-neighbor spins are locked into a singlet ground state of pairs and are less liable to flip the spin of a conduction electron. Because of this pair formation and as a conservative approximation, we have used in the calculations a C_{well} equal to $4x_{\text{well}}(1-x_{\text{well}})^{1/2}/a_0^3$. This warrants that only localized spins which have no nearest neighbors may flip the conduction-electron spin. In any event, one can still use the following calculated curves with another C_{well} by simply scaling the results of τ . Figure 1 shows the results of $\tau_{i \rightarrow j}$ and $\tau_{n \rightarrow n}$ for spin-flip scattering in the well in the case of $\text{Cd}_{0.935}\text{Mn}_{0.065}\text{Te}-\text{Cd}_{0.62}\text{Mn}_{0.38}\text{Te}$. The minimum scattering time for the intrasubband contribution is ~ 14 ps, which would make the spin magnetization of the conduction electrons $\frac{1}{2} \mu_B g^* (n_{1/2} - n_{-1/2})$ to decay twice as fast. ($n_{\pm 1/2}$ is the areal concentration of conduction electrons with $\sigma_z = \pm \frac{1}{2}$ and g^* is their effective Landé g factor.) The spin-flip scattering can therefore be quite fast; at low temperatures it is faster than the acoustic phonon emission and impurity scattering time. It remains, however, slower than the optic-phonon emission (~ 0.2 ps in GaAs quantum wells and certainly shorter in

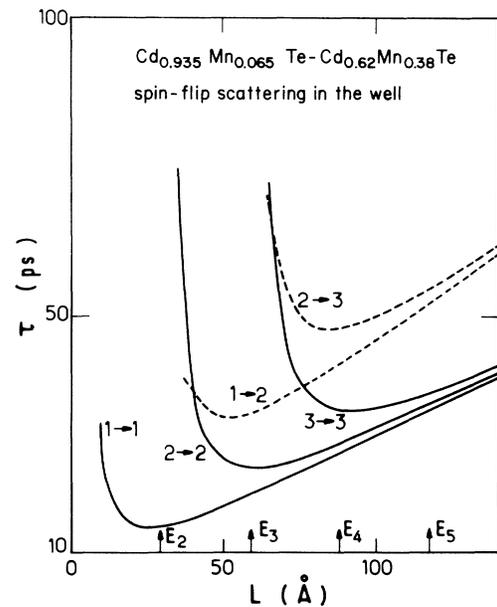


FIG. 1. Intrasubband (solid lines) and intersubband (dashed lines) spin-flip relaxation times of conduction electrons vs the quantum-well thickness L in the case of $\text{Cd}_{0.935}\text{Mn}_{0.065}\text{Te}-\text{Cd}_{0.62}\text{Mn}_{0.38}\text{Te}$ quantum wells. The spin-flip scattering occurs in the well. The arrows labeled E_2, E_3, \dots on the abscissa correspond to the thickness beyond which E_2, E_3, \dots become bound.

CdTe-based heterolayers which are more polar).

The spin-flip scattering in the barrier is much less efficient than that in the well to the extent that the χ_n envelope functions are usually less localized in the barrier. In this case Eq. (7) can be rewritten as

$$\frac{1}{\tau_{n \rightarrow n'}} = \frac{35m^*}{24} C_{\text{barrier}} J^2 P_n P_{n'} \times \frac{\kappa_n \kappa_{n'}}{\kappa_n + \kappa_{n'}} Y \left(E_n + \frac{\hbar^2 k_{\perp}^2}{2m^*} - E_{n'} \right), \quad (10)$$

where $\kappa_n, \kappa_{n'}$ are the electron evanescent wave vectors in the barrier and $P_n, P_{n'}$ are the integrated probabilities of finding the electron in either barriers for the n th and n' th bound states, respectively. From Eq. (10) it is clear that for a given L the $\tau_{n \rightarrow n'}$ will become shorter with increasing n , opposite to the previous case of spin-flip scattering in the well. This complementary behavior results from an increasing leak of χ_n into the barrier to shorten the scattering time therein. The $\tau_{n \rightarrow n'}$ expression at large L can be analytically derived if one remarks that E_n and $E_{n'}$ are $\ll V_b$ in this limit and that P_n varies as $(2/L)(E_n/V_b \kappa_n)$:

$$\frac{1}{\tau_{n \rightarrow n'}} \sim \frac{35}{24} m^* J^2 C_{\text{barrier}} \left(\frac{nn' \hbar^2 \pi^2}{m^* L^3 V_b} \right)^2 \frac{1}{\kappa_n + \kappa_{n'}}, \quad (11)$$

i.e., $\tau_{n \rightarrow n'}$ increases as L^6 at large L . For the opposite limit of vanishing L , $\tau_{1 \rightarrow 1}$ also diverges. This situation actually mirrors that discussed earlier for scattering in the well in the asymptotic regime of large L ; here $\tau_{1 \rightarrow 1}$ diverges as L^{-1} at small values of L . Finally, when L approaches L_n from above, again $\tau_{n \rightarrow n}$ and $\tau_{i \rightarrow j}$ diverge.

An example of the thickness dependence of the spin-flip scattering time in CdTe-Cd_{0.81}Mn_{0.19}Te is presented in Fig. 2. We notice that for very narrow wells $\tau_{1 \rightarrow 1}$ can be small (less than 100 ps) but it rapidly increases with increasing L to exceed 1 ns for a 50-Å well. The large difference between the in-well and in-barrier spin-flip relaxation times is primarily caused by the pronounced localization of the quantum-well eigenstates in the well. However, our choice of the effective number of spins also enhances this difference. For instance, the effective Mn mole fraction for spin-flip scattering is 1.5% (2.9%) for an actual Mn fraction of 19% (6.5%). Thus the number of available scatterers is strongly reduced by the requirement of being without nearest neighbors. Still, the wavefunction localization argument prevails even if one assumes that all the Mn ions are available for spin-flip scattering: As can be seen from Figs. 1 and 2, the scattering in the well will be more efficient than that in the barrier, except for very narrow wells.⁹

This difference between the two types of scattering can be explored in a single heterostructure: a biased double quantum well of CdTe and Cd_{1-x}Mn_xTe wells sandwiched between Cd_{1-y}Mn_yTe barriers. In general, the two lower eigenstates of this double well will, under flat-band condition, correspond to eigenfunctions which are well localized either in the CdTe or Cd_{1-x}Mn_xTe well. The localization will become more pronounced for increasing thickness of the midbarrier. Assuming that the ground state is at zero electric field localized in the

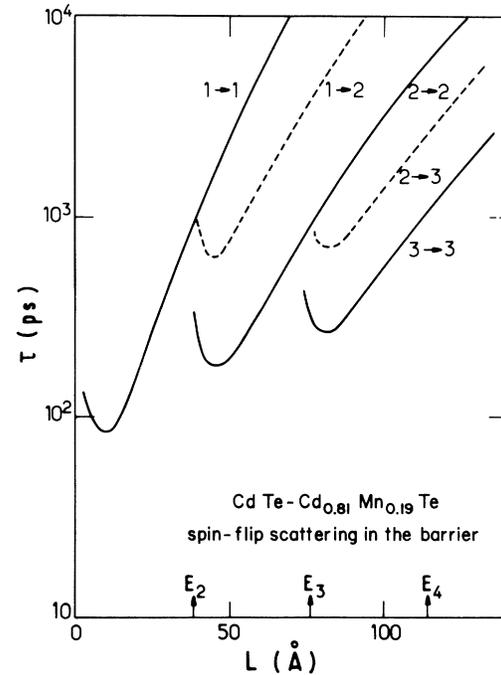


FIG. 2. Intrasubband (solid lines) and intersubband (dashed lines) spin-flip relaxation times of conduction electrons vs the quantum-well thickness L in the case of CdTe-Cd_{0.81}Mn_{0.19}Te quantum wells. The spin-flip scattering occurs in the barrier. The arrows labeled E_2, E_3, \dots on the abscissa correspond to the thickness beyond which E_2, E_3, \dots become bound.

Cd_{1-x}Mn_xTe well, an electric field with proper polarity will lift the Cd_{1-x}Mn_xTe well upward in energy with respect to the CdTe well. Beyond a threshold field ($F \sim \Delta E_1 / ed$, where ΔE_1 is the difference of the ground states in the two wells and d is the sum of the midbarrier thickness and half of the CdTe and Cd_{1-x}Mn_xTe layer thicknesses), the ground state will be localized in the CdTe well. Thus, one will have a structure where the spin-flip scattering time can be controlled by an external parameter to oscillate between short (~ 30 ps) and long (~ 1 ns) values.

Systematic experiments have been lacking in the literature about the spin-flip scattering time in Cd_{1-x}Mn_xTe quantum wells. The data which are the closest to our calculations are provided by the study of the decay of the polarization of the photoluminescence when the sample has been excited with circularly polarized light.³ The excitation creates spin-polarized photoelectrons. If one assumes that the photogenerated holes immediately lose their orientations, the decay of the photoluminescence polarization should reflect that of the electron spin orientation. In an 86-Å Cd_{0.935}Mn_{0.065}Te-Cd_{0.62}Mn_{0.38}Te well, the photoluminescence polarization decays with a time constant of 3-4 ps.³ Our calculations (Fig. 1) lead to a spin-flip time of ~ 26 ps, corresponding to an orientation decay time of ~ 13 ps. This is within an order of magnitude agreement. However, it is not clear whether the holes can indeed lose all their orientation on such a short time scale or whether some defects cannot accelerate the decay of the photoluminescence polarization. We believe that our calcula-

tions are of relevance to the fundamental understanding of the process of exchange-induced spin-flip scattering. Their applicability in realistic situations in the presence of other possible processes and complications must await further, systematic experimental results.

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⁹Many experiments actually involve superlattices rather than single quantum wells. The predicted dissymmetry between the in-well and in-barrier scatterings requires thick enough superlattice barriers to prevent coupling between neighboring wells. In the opposite limit of strong coupling, a bulklike description as outlined in Ref. 6 should be used.