Semiconductor quantum dots (QDs) are called to play a central role in the emerging field of spintronics, because their zero-dimensional confinement constitutes an optimal environment to manipulate the spin of bound electrons. This has stimulated their use as spin filters, as well as several attempts to realize solid state implementations of spin-based qubits. Understanding the spin relaxation in these structures is of utmost interest for their eventual use in practical devices. This has triggered a large number of experimental works in the last few years, where two main classes of spin transitions have been investigated, namely the spin-flip between single-electron Zeeman sublevels and the triplet-singlet (TS) transition in QDs with an even number of electrons. Remarkably, while the former systems have received much theoretical attention, the understanding of the latter is still rather limited. In particular, the TS relaxation due to spin-orbit (SO) coupling – which is often the dominant spin relaxation mechanism in semiconductor QDs – has only briefly been addressed in two-electron QDs, and many relevant features observed in experiments remain unexplained. This is the case of the role of an external magnetic field: experimental measurements away from the TS anticrossings suggest that the influence of axial fields on the spin relaxation is fairly weak. This is in strong contrast with the single-electron case, where a power-dependence of the relaxation rate on the field has been demonstrated, and it might seem surprising because the field reduces the energy splitting between the singlet and the triplet, what should enhance SO coupling. Besides, in the vicinity of the TS anticrossing, both decreased and increased relaxation rates have been reported. In this context, a unified picture describing the effect of an axial magnetic field on the TS spin relaxation rate is on demand.

In this Letter, we study the TS spin relaxation due to SO coupling in circular QDs with weak lateral confinement. Acoustic phonon emission, assisted by SO interaction, has been shown to be the dominant relaxation mechanism in this kind of QDs when cotunneling and nuclei-mediated relaxation are reduced. In fact, cotunneling is an extrinsic scattering process and can be controlled by means of reduced tunneling rates, while nuclei-mediated relaxation dramatically decreases with external magnetic fields. We show that the current experimental evidence can be reconciled within a unified picture, where the field dependence of the relaxation rate is determined by the interplay between spin-orbit coupling and phonon emission efficiency. Furthermore, we show that such interplay can be tailored in order to obtain improved spin lifetimes.

In weakly confined QDs, correlation effects may strongly influence charge and spin excitations. In order to calculate the relaxation time of excited few-electron correlated states, we need to know both ground and excited states with comparable accuracy: our method of choice is the full configuration interaction (FCI). The single-electron states are calculated within the effective mass approximation for a typical “vertical” GaAs/AlGaAs QD, with confinement potential $V(r) = V_z(z) + 1/2m^*\hbar^2(2x^2 + y^2)$, $V_z(z)$ representing the (finite) vertical confinement of a quantum well of thickness $W$, $\hbar \omega_0$ being the single-electron energy spacing of a lateral two-dimensional harmonic trap, and $m^*$ the effective mass. The lateral confinement is much weaker than the vertical one, and a magnetic field, $B$, is applied along $z$. Under these conditions, the low-lying single-electron states are well described by the Fock-Darwin spectrum and the lowest eigenstate of the quantum well. The single-electron levels can be classified by their radial quantum number $n = 0, 1, \ldots$, azimuthal angular momentum $m = 0, \pm 1, \ldots$, and spin $s_z = \uparrow, \downarrow$. In turn, the few-electron states can be labelled by the total azimuthal angular momentum $M$, total spin $S$, its $z$-projection $S_z$, and by the number $N = 0, 1, \ldots$ indexing the energy order.

We introduce the SO coupling via the linear Rashba and Dresselhaus terms, $H_R$ and $H_D$ respectively. For a quantum well grown along the [001] direction, these...
where $\pi$ and $s^z$ are the Rashba and Dresselhaus coefficients for the sample under study. $\pi^\pm$ and $s^z_\pm$ are ladder operators, which change $m$ and $s_z$ by one unit, respectively. Since the few-electron $M$ and $S_z$ quantum numbers are given by the algebraic sum of their single-electron counterparts, Rashba interaction mixes $(M, S_z)$ states with $(M \pm 1, S_z \pm 1)$ ones, and Dresselhaus interaction mixes $(M, S_z)$ states with $(M \pm 1, S_z \pm 1)$ ones.

In our calculation, the SO terms of Eq. 12 are diagonalized in a basis of few-electron states, which are computed as linear combinations of Slater determinants, $\alpha$ and $\beta$ are the Rashba and Dresselhaus coefficients, respectively. Since the few-electron $M$ and $S_z$ quantum numbers are given by the algebraic sum of their single-electron counterparts, Rashba interaction mixes $(M, S_z)$ states with $(M \pm 1, S_z \pm 1)$ ones, and Dresselhaus interaction mixes $(M, S_z)$ states with $(M \pm 1, S_z \pm 1)$ ones.

The electron-phonon interaction is taken into account as in Ref. 8. Hence, we consider not only deformation potential as in previous works (Ref. 9), but also piezoelectric field scattering. The piezoelectric field interaction is dominant when the phonon energy is small, so that it provides the main contribution to the relaxation in the interesting regions of TS anticrossings. GaAs material parameters are taken in the calculations, along with a Landé factor $g = -0.44$.

In Fig. 2 we analyze the relaxation rate from the three first excited to the ground state of two-electron QDs with different dimensions. Left (right) panels correspond to structures without (with) Rashba interaction. For the QD studied in the upper panels, one can see that the relaxation rate increases slowly with the magnetic field, and then it suddenly drops in the anticrossing region ($B \sim 2.25 - 3.25$ T). This behavior, whose physical mechanism will be explained in the following, is in qualitative agreement with various recent experiments in weakly-confined QDs. In particular the rather weak dependence with the field before the anticrossing agrees well with TS relaxation measurements. The increased relaxation rate before the anticrossing has been reported in Ref. and the reduced rate in the anticrossing region may be inferred from the long triplet lifetimes for eight-electron QDs with small ST energy splittings.

The general trends described above can be explained by the opposite effect of the magnetic field on the SO mixing and the phonon emission efficiency. On the one hand, as the singlet-triplet energy splitting decreases, SO interaction couples the states more efficiently, favouring spin relaxation. On the other hand, the phonon energy decreases, reducing the efficiency of the electron-phonon interaction. The latter effect, which follows from the different orbital quantum numbers of the initial and final electron states, occurs at a rate that is determined by the interplay between the acoustic phonon wavelength and the dimensions of the QD. For the QD of the upper panel in Fig. 2, the effect of the magnetic field on the SO interaction and phonon emission is mostly of similar magnitude, which explains the weak changes of the relaxation rate. At the anticrossing point, in spite of the fact that the SO mixing is maximum [see Fig. (b)], the phonon energy is so small (few $\mu$eV) that the spin relaxation is strongly suppressed. It is worth mentioning that this result is opposed to that predicted for the TS anticrossing in a lateral QD, where maximum relaxation rate is predicted at the anticrossing point. The origin of this difference might lie on the fact that the electron-phonon interaction matrix elements of lateral QDs with strongly asymmetric (non-parabolic) confinement potential may
be significant even for very small phonon energies.\cite{21}
As a result, SO interaction alone would dominate spin relaxation in such structures.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig2.png}
\caption{(Color online). Spin relaxation rate of the three first excited states in two-electron QDs vs. magnetic field. Top row: \( W = 10 \text{ nm}, \hbar \omega_0 = 4 \text{ meV} \); bottom row: \( W = 14 \text{ nm}, \hbar \omega_0 = 5 \text{ meV} \). The SO interaction coefficients are shown on top of each column (units of meV Å). Note that before the anticrossing dip, \( |N| = 1, 2, 3 \) correspond to the triplet sublevels \( S_z = +1, 0, -1 \), respectively. In the bottom panels, the shaded areas highlight the magnetic field window of geometrically-suppressed phonon emission (see text).}
\end{figure}

As stated before, the dimensions of the QD are known to play a critical role to determine the phonon emission efficiency.\cite{8, 17, 19} To illustrate this effect on the spin relaxation, in the bottom panels of Fig. 2 we report the rate of a QD with larger height and stronger lateral confinement than the previous one. As compared to the upper panels, one can see a visibly stronger dependence of the relaxation rate on the field. This is because for the present QD dimensions, the phonon emission efficiency turns out not to be balanced with the SO mixing effect. As a result, when the effect of SO coupling prevails the relaxation rate strongly increases with \( B \). Still, in the anticrossing region, that is now shifted towards higher magnetic fields, the suppression of phonon emission again yields a relaxation minimum. Furthermore, a new feature emerges in this case, namely a dip at about \( B \approx 0.5 \text{ T} \) (shaded area in the panels) This dip, which we had previously predicted for charge relaxation in QDs,\cite{17} comes from the geometrically-induced suppression of the phonon emission, occuring when the quantum well width is a multiple of the photon wavelength \( z \)-projection. This feature may give access to very long-lived triplet states at finite values of the magnetic field,\cite{22} where phonon-induced relaxation is usually the dominant scattering mechanism. Moreover, this dip takes place at a \( B \) value where the initial and final electron states are well-resolved energetically, which renders this minimum more useful than the one coming from the TS anticrossing. In the illustrated case, the average relaxation rate of the three Zeeman sublevels in this dip corresponds to a triplet relaxation time of tenths of seconds, two orders of magnitude over the longest triplet lifetime reported to date.\cite{6} The position and depth of this kind of relaxation minima depend on the QD height and the emitted phonon energy. Therefore, they are almost independent of the SO interaction in the structure, which in GaAs has a negligible influence on the phonon energy.

Next, we focus on the effect of the separate Rashba and Dresselhaus contributions over the spin relaxation by comparing the left and right panels of Fig. 2 in the magnetic field region before the TS anticrossing. When only Dresselhaus terms are present (left panels), the singlet mixes directly only with the higher-lying \( (S_z = -1) \) Zeeman sublevel of the triplet. As a result, relaxation from such Zeeman sublevel (dot-dashed line) is about two orders of magnitude faster than from the \( S_z = 0, +1 \) sublevels, and it exhibits a stronger dependence on the field. When a small Rashba interaction is switched on (right panels), direct mixing of the singlet with the triplet \( S_z = +1 \) sublevel is enabled. This accelerates the relaxation rate from this sublevel (dashed line) in one order of magnitude and introduces a stronger dependence on \( B \). It is worth noting that the order-of-magnitude enhancement of the relaxation rate due to the Rashba interaction is present away from the anticrossing region, where the effect of the SO interaction on \( \langle S_z \rangle \) is barely visible [see Fig. 4(b)]. From the above discussion it follows that in a magnetic field both Rashba and Dresselhaus interactions play an important role in determining the TS spin relaxation rate, as opposed to the well-known single-electron case, where the relaxation is mostly due to Rashba coupling only.\cite{5} Moreover, we see that the lifetimes of the triplet Zeeman sublevels may strongly differ depending on the relative Rashba and Dresselhaus contributions. This may be useful to selectively populate the triplet sublevels.

We now investigate the TS spin relaxation in a four-electron QD. The energy spectrum of the lowest-lying triplet and singlet states in a magnetic field, plotted in Fig. 2(a), is very different from that of the two-electron case, but it closely resembles the one found experimentally for eight-electron QDs in Ref.\cite{5} (except for the absence of eccentricity features in the zero field limit\cite{23}).

Here, we investigate the spin relaxation rate in the region \( B \approx 0.3 - 3 \text{ T} \), where the ground state is a singlet \( (M = -2) \) and the first excited state is a triplet with two possible values of the angular momentum, depending on the magnetic field: for \( B < 1 \text{ T} \) the angular momentum is \( M = 0 \), and for \( B > 1 \text{ T} \) it is \( M = -3 \).
This is because the Rashba interaction, regardless of the (here fairly strong) Rashba interaction, leads to increased lifetimes. For smaller $\Delta T$ in Fig. 3(a), which strongly enhances spin relaxation. $M$ is the anticrossing of the upper Zeeman sublevel with the dip of the singlet-triplet energy splitting. As in the two-electron cases studied above, the strong interaction between SO coupling and phonon emission efficiency can be understood in the same terms of compensating SO coupling and phonon emission efficiency. These differences arise from the different orbital initial and final states, and the (usually larger) transition energies. We predict very long triplet lifetimes using QD geometries that lead to suppressed phonon emission.

Improved lifetimes can also be obtained in four-electron QDs by selecting triplet states which do not fulfill the $\Delta M = \pm 1$ selection rule.

In summary, we have estimated the electron TS spin relaxation rate due to SO coupling in weakly-confined cylindrical GaAs/AlGaAs QDs. Experimentally observed trends of TS relaxation in magnetic fields are well understood in terms of the competing SO coupling and phonon emission efficiency. Significant differences have been found as compared to the well-known single-electron spin-flip case, including a critical role of the dot confinement to determine the phonon emission efficiency. These differences arise from the different orbital initial and final states, and the (usually larger) transition energies. We predict very long triplet lifetimes using QD geometries that lead to suppressed phonon emission. Improved lifetimes can also be obtained in four-electron QDs by selecting triplet states which do not fulfill the $\Delta M = \pm 1$ selection rule.

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These states are well separated from higher-lying states, so they might be used as a two-level system for quantum computation purposes. We compare the lifetimes of both triplet states in Fig. 3(b), where the averaged lifetimes of the three Zeeman sublevels are plotted as a function of the singlet-triplet energy splitting, $\Delta_{ST}$, with (dashed lines) and without (solid lines) Rashba interaction (Dresselhaus interaction is present in both cases). The qualitative behavior is similar for both states: the lifetime is roughly constant for large energy splittings ($\Delta_{ST} > 0.25$ meV), and it increases when the energy splitting is small ($\Delta_{ST} < 0.25$ meV). This behavior, which is in agreement with the experimental findings of Ref. 5, can be understood in the same terms of compensation between SO coupling and phonon emission efficiency as in the two-electron cases studied above. The strong dip of the $M=-3$ triplet at $\Delta_{ST} \sim 0.15$ meV is due to the anticrossing of the upper Zeeman sublevel with the $M=-4$ singlet at strong magnetic fields [at $B \approx 2.8$ T in Fig. 3(a)], which strongly enhances spin relaxation. For smaller $\Delta_{ST}$, though, the small phonon energy again leads to increased lifetimes.

An important result shown in Fig. 3(b) is that the average lifetime of the triplets differs by over one order of magnitude depending on their angular momentum, regardless of the (here fairly strong) Rashba interaction. This is because the $M=0$ triplet differs from the $M=-2$ singlet ground state in two quanta of angular momentum, and therefore direct SO mixing is not possible. In contrast, direct mixing is possible for the $M=-3$ triplet, and this makes the relaxation much faster. It then follows that, by using four-electron QDs instead of two-electron ones, one can use an external magnetic field to select excited states whose spin transition is “forbidden” even in the presence of linear SO interaction. This result is consistent with recent measurements, where different lifetimes were observed for triplet states with different orbital quantum numbers. However, in the experiment the triplet lifetimes changed by a factor of two only. The main reason for this difference is probably the ellipticity of their QDs, which mixes states with different angular momenta and hence weakens the efficiency of the $\Delta M = \pm 1$ selection rule.

FIG. 3: (Color online). (a) Energy of the four lowest-lying levels of a four-electron QD as a function of the magnetic field. The QD has $W=10$ nm and $\hbar \omega_{0}=4$ meV, $\alpha=15$ meV $\cdot \AA$ and $\beta=25$ meV $\cdot \AA$. The approximate quantum numbers $(M, S)$ are shown. Arrows are used to indicate the two different spin transitions we compare. (b) Average triplet lifetime for the two spin transitions we compare, as a function of the singlet-triplet energy splitting. $\beta = 25$ meV $\cdot \AA$, and solid (dashed) lines are used for $\alpha=0$ ($\alpha=15$) meV $\cdot \AA$. 

* Electronic address: climente@unimore.it

URL: www.nanoscience.unimore.it

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