

Atomistic theory of spin relaxation in self-assembled $\text{In}_{1-x}\text{Ga}_x\text{As}/\text{GaAs}$ quantum dots at zero magnetic field

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We present full atomistic calculations of the spin-flip time (T_1) of electrons and holes mediated by acoustic phonons in self-assembled $\text{In}_{1-x}\text{Ga}_x\text{As}/\text{GaAs}$ quantum dots at zero magnetic field. At low magnetic field, the first-order process is suppressed, and the second-order process becomes dominant. We find that the spin-phonon-interaction induced spin relaxation time is 40 - 80 s for electrons, and 1 - 20 ms for holes at 4.2 K. The calculated hole-spin relaxation times are in good agreement with recent experiments, which suggests that the two-phonon process is the main relaxation mechanism for hole-spin relaxation in the self-assembled quantum dots at zero field. We further clarify the structural and alloy composition effects on the spin relaxation in the quantum dots.

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The electron/hole spin in semiconductor quantum dots (QDs) is believed to be a promising candidate for solid state quantum computations [1]. Recently years, huge progress have been made experimentally in initialization, manipulation and controlling of the spins in QDs[2–5]. However, the short spin lifetime is still a major obstacle to realize the quantum computation. Until now the mechanisms for spin relaxation in QDs are still not well understood both in theory and in experiments. Spin-phonon interaction due to the spin-orbit coupling (SOC) effects is one of the main mechanisms lead to spin relaxation in the QDs [6, 7], which depends strongly on the geometries and compositions of the QDs. An accurate description of SOC is crucial to understand the spin relaxation. Unfortunately, the understanding of the spin relaxation from the atomistic level is still unavailable.

Electrons were expected to have very long spin lifetime, because they have small SOC in QDs. However, because of hyperfine interactions with nuclear spins, the electron spin coherence time is greatly reduced (~ 500 ps) [3]. The electron spin lifetime can be prolonged by applying an external magnetic field to polarize the nuclear spin [2, 3]. Unfortunately, at the same time, the electron spin lifetimes (T_1) decrease fast with the magnetic field, being proportional to B^{-5} [2]. In contrast to electrons, holes have p -like atomic wave functions. Therefore, the hyperfine interactions with the nuclear spins are small [8]. Indeed, recent experiments show that holes have very long spin lifetimes in InAs/GaAs QDs. For example, Heiss et al. found that the holes can have lifetimes of 0.27 ms at 1.5 T [4]. Only very recently, high fidelity spin states have been prepared at low magnetic field, taking the advantages of the extreme long hole spin lifetimes (~ 1 ms) [5].

At very low magnetic field, the first-order spin-phonon interaction is greatly suppressed, and the multi-phonon process becomes dominate. The $T_1 \propto B^{-5}$ law breaks down at low magnetic field [2, 9]. Most previous theoretical works focus on spin relaxation in a large magnetic field [6, 7, 10], and there are only few studies of spin relaxation in QDs at low magnetic field [9]. These studies are all based on effective mass approximations or $\mathbf{k} \cdot \mathbf{p}$ theory [6, 10, 11], in which the *effective* SOC are added in by hand in the form of Dresselhaus interactions and Rashba interactions. These continuum theories treat poorly the local strain and alloy composition effects especially for hole, which may play an extremely important role to the effective SOC.

In this letter, we present the first atomistic calculations [12] of the spin relaxation of electrons and holes at zero magnetic field in self-assembled $\text{In}_{1-x}\text{Ga}_x\text{As}/\text{GaAs}$ QDs. In this method, the Hamiltonian naturally takes the *full* SOC effects into account at the atomistic level. The method needs no fitting parameters to the QD experiments, and therefore can give quantitative predictions that can be directly compared to experiments. Our calculations show that the holes have extremely long spin lifetimes, 1–20 ms at $T=4.2$ K, which is in excellent agreement with the recent experimental data [5]. This confirms that the two-phonon scattering process with the SOC is the main mechanism for the spin relaxation of holes at zero magnetic field.

At low magnetic field, the first-order process is suppressed, because the Zeeman splitting between spin-up and spin-down states is very small. Therefore, the density of phonon states that satisfy energy conservation is extremely low. In this case, the spin relaxation via the two-phonon processes become dominant: a spin at the initial (labeled as i) state, ϵ_i , absorbs a phonon of momentum \mathbf{q} and jumps to an intermediate (s) state, ϵ_s . It emits a phonon with momentum \mathbf{k} and relaxes to the final (f) state, ϵ_f , which has an opposite spin of the initial

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state. The electron and hole spin-flip rate (τ_ν^{-1}) from the initial state to the final state is given by the second-order Fermi's Golden Rule,

$$\frac{1}{\tau_\nu} = \frac{2\pi}{\hbar} \sum_{\mathbf{q}, \mathbf{k}} \left[\sum_s' \left(\frac{M_{\mathbf{q}}^{is} M_{\mathbf{k}}^{sf}}{\epsilon_i - \epsilon_s + \hbar\omega_{\mathbf{q}}} + \frac{M_{\mathbf{k}}^{is} M_{\mathbf{q}}^{sf}}{\epsilon_i - \epsilon_s - \hbar\omega_{\mathbf{k}}} \right) \right]^2 N_q(N_k + 1) \delta(\epsilon_f - \epsilon_i - \hbar\omega_{\mathbf{q}} + \hbar\omega_{\mathbf{k}}). \quad (1)$$

where only long-wave acoustic phonons are involved in the process, $\omega_{\mathbf{q}} = c_\nu |\mathbf{q}|$, and c_ν are the sound speeds for the $\nu = \text{LA}$ and TA modes. $N_q = 1/[\exp(\hbar\omega_{\mathbf{q}}/k_B T) - 1]$ is the Bose-Einstein distribution function. The summation is over the intermediate states includes all the states except for the initial and final states. The matrix elements $M_{\mathbf{q}}^{if}$ are given by:

$$M_{\mathbf{q}}^{if} = \alpha_\nu(\mathbf{q}) \langle \psi_f | e^{-i\mathbf{q}\cdot\mathbf{r}} | \psi_i \rangle, \quad (2)$$

where $|\psi_i\rangle$ and $|\psi_f\rangle$ are the wave functions of initial and final state, respectively. $\alpha(\mathbf{q})$ is the electron-phonon coupling strength. We have considered three electron-phonon interaction mechanisms in the QDs [10]: (i) electron-acoustic-phonon interaction due to the deformation potential ($\nu = \text{LADP}$), (ii) electron-acoustic-phonon interaction due to the piezoelectric field for the longitudinal modes ($\nu = \text{LAPZ}$), and (iii) electron-acoustic-phonon interaction due to the piezoelectric field for the transverse modes ($\nu = \text{TAPZ}$). The parameters are taken from Ref. [10]. The overall spin relaxation time $T_1 = \sum_\nu 1/\tau_\nu$.

To calculate T_1 , it is crucial to obtain high-quality single-particle wave functions and energy levels [10]. In this work, we use an atomistic pseudopotential method, which has been proven to be very accurate for InAs/GaAs QDs [12–14]. We consider realistic lens-shaped $\text{In}_{1-x}\text{Ga}_x\text{As}/\text{GaAs}$ QDs embedded in a GaAs matrix containing $60 \times 60 \times 60$ 8-atom unit cells. The dots are assumed to grow along the [001] direction, on the top of the one-monolayer wetting layers.

We first relax the dot+box system by minimizing the strain energy as a function of the coordinate $\{\mathbf{R}_{n,\alpha}\}$ of the α -th atom at the site n for all atoms, using the valence force field (VFF) method [15, 16]. Once we have all the atom positions, we obtain the energy levels and wavefunctions by solving the single-particle Schrödinger equation [17],

$$\left[-\frac{\nabla^2}{2} + \sum_{n,\alpha} \hat{v}_\alpha(\mathbf{r} - \mathbf{R}_{n,\alpha}) \right] \psi_i(\mathbf{r}) = \epsilon_i \psi_i(\mathbf{r}). \quad (3)$$

$\hat{v}_\alpha(\mathbf{r})$ is the screened atomic pseudopotential for the α -th element, including a local potential and a non-local SOC term [12],

$$V_{\text{SO},\alpha} = \sum_l |l\rangle \delta V_{l,\alpha}(\mathbf{r}) \mathbf{L} \cdot \mathbf{S} \langle l|, \quad (4)$$

TABLE I: Spin relaxation times of the electrons (T_1^e) and the holes (T_1^h) in self-assembled $\text{In}_{1-x}\text{Ga}_x\text{As}/\text{GaAs}$ QDs at 4.2 K. All lengths are in nm. β^2 is the weight of the minority spins of the initial states.

height	base	x	T_1^e (s)	β^2 ($\times 10^{-4}$)	T_1^h (ms)	β^2 ($\times 10^{-4}$)
2.5	20	0	81	19	17.9	96
3.5	20	0	59	16	4.5	140
2.5	25	0	54	17	20.4	65
3.5	25	0	43	16	12.4	83
2.5	25	0.15	44	19	15.5	62
3.5	25	0.15	40	11	6.4	83
2.5	25	0.30	44	20	10.8	65
3.5	25	0.30	40	10	4.5	116
2.5	25	0.50	53	3	5.2	87
3.5	25	0.50	43	5	2.7	88

where l is the angular momentum (only $l=1$ is used). \mathbf{L} is the angular momentum operator, and \mathbf{S} is the spin operator. The atomic pseudopotentials are fitted to the bulk properties. Once the parameters are determined, there are no free parameters for modeling the QDs. The method naturally includes the Rashba and Dresselhaus SOC in a “first-principles” manner.

The single-particle Schrödinger equation is solved by the linear combination of bulk bands (LCBB) method [18]. We use eight bands for both the electrons and holes, and take the inter-band coupling (heavy-hole-light-hole coupling and the valence-conduction band coupling) into account, which is very important for hole/electron spin relaxation. A $6 \times 6 \times 16$ k-mesh converges very well the results. Due to the SOC, the wave functions mix opposite spin components, i.e., $\psi_i = \alpha |\uparrow\rangle + \beta |\downarrow\rangle$.

To calculate the matrix elements, $M_{\mathbf{q}}$, we first calculate the single-particle wave functions on a sparse k mesh following Ref. 19. We then interpolate the wave functions into a more dense $49 \times 49 \times 129$ k -mesh to get accurate results. We sum over 20 intermediate states (s in Eq. 1), which converge a spin lifetime < 1 s for electrons and < 1 ms for holes. Details on how to calculate $M_{\mathbf{q}}$ will be published elsewhere.

In Table I, we list the electron and hole spin lifetimes for some typical $\text{In}_{1-x}\text{Ga}_x\text{As}/\text{GaAs}$ QDs at $T=4.2$ K. We also list the weight of the minority spin of the initial states β^2 in the Table. From the Table, it can be seen that the electron spin lifetimes due to the spin-phonon interaction are extremely long, 40–80 s, because the spin mixture is quite small (See Table I). We find that the inter-band coupling has a large contribution to the spin relaxation for electrons. Ignoring inter-band coupling leads to a spin lifetime that is about three times longer for electrons. However, the actual electron-spin lifetimes are determined by the hyperfine interaction with nuclear spins at low magnetic field.

The hole-spin lifetimes of typical $\text{In}_{1-x}\text{Ga}_x\text{As}/\text{GaAs}$ QDs are 1 – 20 ms. For alloy dots, the typical hole-spin

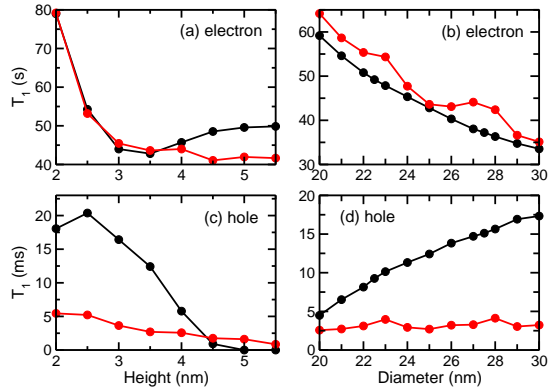


FIG. 1: (Color online) Spin relaxation times of electrons and holes in lens-shaped $\text{In}_{1-x}\text{Ga}_x\text{As}/\text{GaAs}$ QDs of different heights [(a), (c)], and base diameters [(b), (d)]. Height varies keeping diameter at 25 nm and the diameter varies keeping height at 3.5 nm. Black lines are the results for pure dots, whereas the red lines are the results for alloy dots with $x=0.5$.

lifetime is a few ms, which is in good agreement with recent experimental data (~ 1 ms) [5]. This suggests that two-phonon spin scattering is the main mechanism that leads to spin flip for holes in these self-assembled QDs at low magnetic field. This finding differs greatly from the results given in Ref. [11] where the second-order phonon process is not considered. The spin lifetimes of the holes are about three orders of magnitudes shorter than those of the electrons. This is because the single-particle energy spacing of the electrons is 3–5 times larger than that of holes, and the spin mixing due to the SOC for holes is also much stronger than that of electrons, as shown in Table I. The spin-flip times in pure QDs are significantly longer than those of alloy QDs. The main reason for the longer hole spin-flip time in pure QDs is that the energy spacing in pure QDs is larger, and the spin mixture in alloy dots tends to be slightly larger than that in the pure dots.

The atomistic theory reveals some important features that can not be observed by $k \cdot p$ method, such as geometry and alloy compositions effects to the spin relaxation. To explore the effects of geometry on the spin flip time in QDs, we calculate the spin flip time of lens-shaped QDs, as functions of dot height (diameter) while fixing the dot diameter (height). The results are presented in Fig. 1 (a) and (b) and for electrons, and Fig. 1 (c) and (d) for holes. For pure dots (black dots), the spin lifetime of the electrons decreases as dot height increases from 2 nm to 3.5 nm, and becomes flat for taller dots. The spin lifetimes of holes, also decreases with increases in the height of the dots. For extremely tall pure InAs/GaAs quantum dots, the spin flip time is extremely fast (about two orders of magnitudes faster). This is because in very tall pure InAs/GaAs QDs, holes tend to localize in the interface between the dots and matrix, resulting in a very small energy spacing, and a fast spin relaxation. The spin

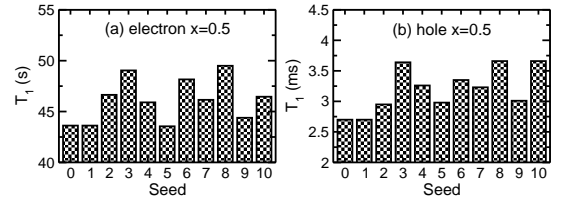


FIG. 2: Spin relaxation time of $\text{In}_{0.5}\text{Ga}_{0.5}\text{As}/\text{GaAs}$ QDs with different Ga distributions.

lifetimes decrease with increasing dot diameter for electrons [Fig. 1 (c)]. In contrast, the spin lifetimes increase with increasing dot diameter for holes [Fig. 1 (d)]. To understand the effects of geometry on the spin lifetime, we note that there are two competing factors: the energy spacing and spin mixing. For electrons, the SOC is not very sensitive to the dot geometry, and the spin lifetimes are mainly determined by the single-particle energy spacing. For holes, increasing dot height leads to a smaller energy spacing between the levels, and the spin mixing also increases with dot height because of the stronger effective SOC. Therefore the spin lifetime decreases. Increasing the dot diameter, however, leads to a smaller energy spacing between the levels, which tends to increase the spin flip time, but the spin mixing decreases with increasing dot diameter. The overall effect is that the spin lifetime increases with increasing dot diameter. Therefore, for pure InAs/GaAs QDs, a flatter QD has a much longer lifetime than a tall QD.

We also show that the geometry-dependent spin lifetime for $\text{In}_{0.5}\text{Ga}_{0.5}\text{As}/\text{GaAs}$ alloy QDs (red dots) in Fig. 1. For electrons, the spin lifetimes of alloy QDs are very similar to those of pure QDs, whereas for holes, the alloy QDs show a much weaker geometry dependence for their spin lifetime. When the dot height increases, the lifetime decreases slightly with increasing dot height. However, for most of the dots, the alloy dots have shorter lifetimes than those of the pure dots. For very tall dots, the alloy dots have longer lifetime, because the holes are not localized at the dot-matrix interface in the alloy dots. The spin lifetimes of holes are also flat with increasing of the dot diameter.

It has been shown by Mlinar and Zunger [20] that the randomness of In, Ga alloys may have large effects on their optical properties for finite sizes $\text{In}_{1-x}\text{Ga}_x\text{As}/\text{GaAs}$ QD ($< 10^5$ atoms). To investigate the alloy randomness effects on the spin relaxation time, we calculate T_1 for $\text{In}_{0.5}\text{Ga}_{0.5}\text{As}/\text{GaAs}$ QDs, with various random distributions of Ga atoms in the dots. The results are shown in Fig. 2. We find that the spin lifetimes of electrons are distributed between 44 s to 50 s, in which the lifetime fluctuation is less than 10 %. The spin lifetimes of the holes are distributed between 2.7 ms and 3.6 ms and the fluctuation of the lifetimes of holes is about 33 %. Analyses show that the spin lifetime fluctuation mainly comes

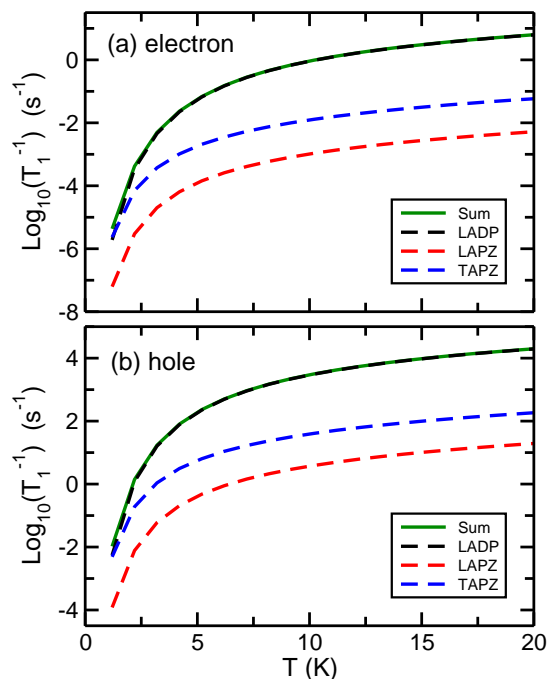


FIG. 3: (Color online) The electron- (a) and hole- (b) spin relaxation rates ($1/T_1$) of a pure InAs/GaAs QD as functions of temperature. QD base diameter=25 nm, height=3.5 nm. The black, red, and blue lines represent the spin relaxation rate due to the LADP, LAPZ, and TAPZ mechanisms, respectively. The green solid lines are the total relaxation rates.

from fluctuations in the single-particle energy spacing (0.4 meV for electrons and 1.0 meV for holes) caused by the Ga distributions.

Figure 3 depicts the spin-flip rates (green solid lines) for electrons and holes as functions temperature for a lens-shaped InAs/GaAs QD with diameter $D=25$ nm, and height, $h=3.5$ nm. The spin relaxation times for both electrons and holes increase quickly with decreasing temperature at low temperatures [9], because fewer phonons can take part in the scattering process. At lower temperatures, the holes may have extremely long lifetimes, for example, $T=2$ K, $T_1=1.5$ s, suggest that hole spins can be promising candidates for quantum computing. The spin relaxation times $T_1 \sim 1/T^2$ for temperatures >20 K, which is different from the one phonon process, where $T_1 \sim 1/T$ [9]. We also compare the contributions of the three carrier-phonon interactions to the spin flip time. Among the three mechanisms, the contributions from the LAPZ (red dashed lines) and the TAPZ (blue dashes lines) are 2 to 3 orders of magnitudes weaker than that of the LADP (black dashed lines) from 1K to room temperature (not shown). Below 1 K, the TAPZ has similar contribution to the spin flip rate to LADP.

To summarize, we presented the first atomistic calculations of the multi-phonon-induced electron/hole spin relaxation in self-assembled $\text{In}_{1-x}\text{Ga}_x\text{As}/\text{GaAs}$ quantum

dots at zero magnetic field. The calculated the spin lifetimes of the holes were in the range of a few milliseconds to 20 milliseconds at 4.2 K, which is in excellent agreement with recent experimental results. We further studied the structural and alloy composition effects on the spin lifetimes, and found that the alloy QDs have much shorter hole-spin lifetimes. Compared with previous methods, the current method has greater predictive power, and we expected it to play an important role in future studies on spin relaxations in quantum dots.

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