Hole spin initialization in Quantum Dots by a periodic train of short pulses

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Abstract. We model pump-probe experiments leading to the all-optical initialization of the hole spin of a p-doped InAs/GaAs quantum dots ensemble. We consider selection rules of mixed hole states and include periodic excitation conditions. Hyperfine interaction is taken into account as the common decoherence mechanism for the spins of electrons and holes. We show that the degree of hole spin polarization can be maximized by quenching the action of the hole hyperfine interaction with a small applied magnetic field. However additional hole spin relaxation mechanisms, in the microsecond time range, determine the absolute value of this maximum.

1. Introduction

Spin of an individual carrier localized on a nanometer scale is a promising candidate for implementation of spin-based devices or qubits, since its localization implies the suppression of spin relaxation mechanisms related to its movement or diffusion. Future applications in spintronics and quantum information fields require fundamental studies on initialization, manipulation and readout of the carrier spin polarization. In this direction, a special attention is devoted to the all-optical approach which allows, in addition to spin polarization and readout, a very fast speed of spin manipulation. In last years, electron spin initialization has been demonstrated under resonant and pulsed excitation for electrons confined in quantum dots[1]. Recent experiments also show that it is possible to initialize a single hole spin confined in a QD by using a cw optical excitation[2], or by immersing it in a diode structure and using a resonant optical excitation followed by a fast electron tunneling controlled by an applied voltage[3].

Here, we show that it is possible to initialize the spin of an ensemble of single holes confined in QDs by using a train of short pulses. We model recent pump-probe experiments[4] and calculate the degree of hole spin polarization.

2. Experiments, model and discussion

We measured the photo-induced circular dichroism (PCD) of a sample containing p-doped self-assembled InAs QDs. More technical details about this sample were given elsewhere[4]. A picosecond Ti:sapphire laser is split into pump and probe beams (the repetition frequency is 76 MHz). The beams propagate along the growth axis z, and are tuned to the energy of the lowest-allowed optical transition of InAs QDs containing a single resident hole (1.35 eV). The pump beam is $\sigma^+/\sigma^-$ modulated at 42 kHz with a photo-elastic modulator, and creates a positive trion in its ground state. The probe beam is linearly polarized. After transmission through the sample, it is decomposed into its two circular components, and the difference of their intensities is measured with a balanced optical bridge.
Figure 1 (a) shows the temporal behaviour of the low-temperature PCD signal obtained in absence and in presence of an external magnetic field applied along z direction, i.e., in Faraday configuration. We observe at least two components: a short-living component associated to the polarization of the photo-created species, and a nonzero PCD signal at negative pump-probe delays. This long-living component indicates that the spin polarization is not fully relaxed within the $T_L = 13$ ns repetition period of the laser pulses and is unambiguously associated to the net spin polarization of the resident holes, the only species present in the sample after the radiative recombination of trions ($T_R \approx 800$ps$^4$). The density of excitation is chosen in such a way that the value of the PCD signal obtained for negative delays reaches a maximum. That corresponds to a mean pump power density $\approx 60$ Wcm$^{-2}$. Figure 1 (b) shows the variation of the PCD signal at negative delays as a function of the applied magnetic field in Faraday configuration. The signal presents a minimum at zero field and a plateau of saturation at a value $B_s = 10$ mT with a quasi-Lorentzian behaviour between these two extrema. In previous works, it has been demonstrated that this experimental curve is related to the mixed character of the hole states and that the study of this Lorentzian dependence gives information about the strength of the hole-hyperfine interaction$^4,5$.

Figure 1(a) 2K Temporal behaviour of PCD signal for two values of the applied magnetic field in Faraday configuration. The zero level is common at both curves. Dashed arrows at negative pump probe delay indicate the signal associated to hole-spin polarization. (b) Magnetic field dependence of the PCD signal at negative delay. The solid line is the calculated degree of hole spin polarization defined as $J_z(0^-)/J_m$.

The temporal behaviour of the PCD signal is given by the difference of the photo-induced absorption for the $\sigma^+$ and $\sigma^-$ components of a probe beam:

$$PCD(t) = \alpha^p_{\sigma^+}(t) - \alpha^p_{\sigma^-}(t) \propto [\tilde{\rho}_{s_{3/2}}(t) - \tilde{\rho}_{s_{1/2}}(t)] - [\rho^s_{\uparrow}(t) - \rho^s_{\downarrow}(t)] ,$$  

where $\alpha^p_{\sigma^+}(t)$ ($\alpha^p_{\sigma^-}(t)$) is the absorption of a $\sigma^+$ ($\sigma^-$) probe photo-induced by the $\sigma^+$ pump pulse, $\tilde{\rho}_{s_{3/2}}$ ($\tilde{\rho}_{s_{1/2}}$) represents the population of the $\uparrow \downarrow = |\tilde{\psi}_{s_{3/2}}\rangle (\downarrow \uparrow = |\tilde{\psi}_{s_{1/2}}\rangle)$ mixed hole state, and $\rho^s_{\uparrow}(t)$ ($\rho^s_{\downarrow}(t)$) represents the population of the trion state $\uparrow \downarrow \uparrow (\downarrow \uparrow \downarrow)$ containing a spin-up (down)electron.

The mixed hole states can be written as:

$$|\tilde{\psi}_{s_{3/2}}\rangle = \frac{1}{\sqrt{1 + |\beta|^2}} \left( |\psi_{s_{3/2}}\rangle + \beta^* |\psi_{s_{1/2}}\rangle \right)$$  

where $\beta = \frac{\langle \psi_{s_{3/2}} | \tilde{\psi}_{s_{1/2}} \rangle}{\sqrt{1 + |\beta|^2}}$.
with $|\varphi_{\pm1/2}\rangle$ the pure light-hole (lh) state, where $\beta$ is a function of the strain tensor components. The PCD signal is written as a function of the projections along $z$ of the hole spin $J_z(t)$ and the photo-created electron spin $S_z(t)$:

$$P_{\text{CD}}(t) \propto \frac{2(1+|\beta|^2)}{3-|\beta|^2} J_z(t) - 2S_z(t) = \frac{J_z(t)}{J_m} - 2S_z(t)$$  \hspace{1cm} (3)

For mixed hole states, a circularly polarized pump operates a Rabi oscillation with a transfer factor $\sin^2(\theta/2)$ between the mixed hole state $\downarrow$ and the trion state $\uparrow\downarrow\downarrow$, while a transfer factor $\sin^2(\beta|\Theta/2\sqrt{3})$ occurs between the hole state $\uparrow$ and the trion state $\uparrow\downarrow\uparrow$ (see figure 2). The pulse area is defined by the expression $\Theta = \int dE(t) dt / h$, where $d$ is the dipole transition matrix element and $E(t)$ is the envelope of the pump optical field. Hole mixing also affects deeply the optical selection rules for spontaneous emission. Figure 2 shows two linearly polarized lights associated to the crossed transitions, which appear in addition to the circularly polarized ones. All emission rates are given in the caption of figure 2.

Figure 2. Schematic representation of the interconnected dynamics of the hole and the electron spins. At $t=0$ pump beam creates trions. During the lifetime of trions, $T_R$, the photo-created electron spins (the resident hole spins) precess in the effective nuclear field $\vec{B}_N(e)$, while they recombine. After recombination, hole nuclei interaction continues to be responsible of the hole spin dephasing observed in the quantum dots ensemble. $T_{\Lambda}^h$ is the associated dephasing time of the mixed hole with $T_{\Lambda}^h$ the dephasing time of pure hh.

Once the electron and hole spins have been initialized by a short pump pulse, we assume that the main decoherence mechanism for the spins of electrons and holes is the hyperfine interaction, which has a different nature for electrons [6,7] and holes [5,8] but is modelled in both cases by a frozen effective nuclear field denoted $\vec{B}_N(e)$ for electrons, and $\vec{B}_N^h$ for holes. Then, in a single QD, the hole spin $J(t)$ (resp. the photocrated electron spin $S(t)$) will be submitted to a frozen nuclear field $\vec{B}_N^h$ (resp. $\vec{B}_N^e$) and a magnetic field $\vec{B}$ applied along the $z$-axis (Faraday configuration). For simplicity, we have considered a constant envelope wave function for hole and electron in the QD. The correlation between $\vec{B}_N^h$ and $\vec{B}_N^e$ has been included by fixing the following relations between the in-plane and out-of-plane components :

$$\frac{B_{N_x}^h}{B_{N_y}^h} = \frac{(2|\beta|/\sqrt{3}) B_{N_x}^e}{B_{N_y}^e} = \frac{(2|\beta|/\sqrt{3}) g_e T_e^c}{\sqrt{1+|\beta|^2} g_h T_{\Lambda}^c} \quad (4)$$
where \( g_e \) and \( g_h \) are the Landé factor for electrons and holes, \( T_{\alpha}^x \) and \( T_{\alpha}^h \) are the ensemble dephasing times due to hyperfine interaction for electron and pure hh respectively[5]. These relations are exact in presence of a single nuclear species. In presence of two different species, a study of the correlation between both nuclear fields should be needed. However both fields being mainly due to the In nuclei (because of the larger value of \( I_{p0} \)), we kept the relations (4) between the nuclear fields.

As the hole-spin polarization is not relaxed within the time interval between two pump pulses, we have considered the coupled spin dynamics of a resident hole-spin and of the corresponding photo-generated electron spin under periodic excitation and sought for a stationary solution where the hole polarization just before the arrival of a pump pulse equals the one at delay \( T_L \). The experimental measurements being performed on an ensemble of QDs, one gets the total electron spin and hole spin by averaging over an anisotropic Gaussian distribution of \( B_h^0 \)[5].

The fit of the data (solid lines of figure 1) are quite good. In order to fit the temporal behaviour of PCD at \( B_s \) in particular to reproduce the short-living component and the difference of PCD signal between \( t=2ns \) and \( t=13ns \), we need an additional hole-spin relaxation process characterized by a time \( T_1=0.3\mu s \). Several values of the used parameters were determined experimentally in our sample: \( g_e=-0.4, T_R=700ps, T_{\alpha}^h=550ps, \) or found in the literature: \( g_h=1.5, T_{\alpha}^h=5\) ns [5]. We have also to note that for pure hh states the excited state population shows a periodic behaviour as a function of the pump pulse area \( \Theta \); the hole-spin polarization reaches a maximum at \( \Theta=\pi \) and a zero at \( \Theta=2\pi \). In our case, for mixed hole states, the zero of the hole spin polarization, is reached for a value \( \Theta<2\pi \). Indeed, \( S_0(t=0^-) \) is equal to zero for \( \sin^2(\Theta/2) = \sin^2(|\beta|/2\sqrt{3}) \) (see also figure 2), i.e. \( \Theta = 2\pi/|1+|\beta|/\sqrt{3}| \), and then a broad maximum has to be reached at \( \Theta_{\text{max}} \approx \pi \), as confirmed by the obtained fitting value \( \Theta=1.5 \).

Bottom panel of figure 3 shows the calculated ratio \( \kappa \) (defined in the caption of figure 3) at negative delay (0°) as a function of of \( |\beta| \), for \( T_1=0.3 \) \( \mu s \) and \( T_1 \) infinite. We remark that this calculated ratio is independent of the value of \( T_1 \), but increases with increasing \( |\beta| \). \( \kappa \) can be compared directly to the experimental ratio \( \frac{\text{PCD\textsubscript{\text{(-130ps,B\textsubscript{\text{average}})}}}}{\text{PCD\textsubscript{\text{(-130ps,B=0)}}}} \) when the pump power has been chosen to maximize the PCD \( (t=130ps) \) signal at \( B=0 \). From this comparison it is possible to determine the \( |\beta| \) value, in our case \( |\beta|=0.45 \). Top panel of figure 3 shows the calculated degree of hole-spin polarization at \( B_s \), as a function of \( |\beta| \). We observe that the value of the maximum degree of hole spin polarization is fixed not only by \( |\beta| \) but also by the value of \( T_1 \). This maximum degree of hole spin polarization is equal to unity for pure hh states and \( T_1 \) infinite, but is clearly reduced for mixed hole states or \( T_1=0.3\mu s \). In conclusion, time dynamics study of PCD signal confirms a long-range hole spin relaxation time (\( \mu s \) range)[9]. Further experimental investigation on ultra-slow hole-spin dynamics is required.

**Figure 3** Top panel shows the maximum value of hole-spin polarization at negative pump-probe delay time as a function of mixed hole parameter \( |\beta| \). This maximum is obtained for a couple of values \( (\Theta=\Theta_{\text{max}}, B=B_s) \). Solid line gives these values for \( T_1 \) infinite and dotted line for \( T_1=0.3 \)\( \mu s \). Bottom panel shows the dependence on \( |\beta| \) of the \( \kappa \) ratio for negative delays. \( \kappa \) is defined for \( \Theta_{\text{max}} \) as the ratio of the hole spin polarization at \( B_s \) to the hole spin polarization at \( B=0 \). We underline the fact that the value of \( \Theta_{\text{max}} \) is different for different values of \( |\beta| \). Black circles represent \( \kappa \) for both \( T_1 \) values.
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