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Coherence of an electron spin in quantum dots generated by a resonant optical pulse with elliptic polarization

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ABSTRACT

We have experimentally observed the spin polarization process of single electrons in InP/InGaP quantum dots by time-resolved Kerr rotation measurements. It is found that the inversion of the spin polarization direction occurs with the variation of the intensity of the optical pulse. The spin coherence lifetime abruptly changes on the occurrence of the inversion. We have reproduced the inversion in numerical simulations using the density operator of the electron–trion four-level system, assuming a small deviation of the optical pulse from circular polarization. The change of the spin lifetime is attributed to the qualitative change of the four-level system in the electric polarization state.

1. Introduction

The two spin states of an electron with $s=\frac{1}{2}$ in a semiconductor quantum dot (QD) make up the space of the operational states of a quantum bit (qubit) [1]. This solid-state qubit has an excellence of the integratability with existing semiconductor technologies. The lifetime of the spin states in QDs is generally very long (up to microseconds) [2] owing to the suppression of the relaxation mechanisms coming from the spin-orbit coupling [3]. They can be initialized and manipulated by optical field, which is resonant or near-resonant to the electron–trion (negatively charged exciton) transitions [4]. The initialization is equivalent to the generation of coherence between the two spin states, and its fastest process is obtained by the resonant excitation by circularly polarized light. According to the optical selection rule, the $\sigma+$ excitation transfers some of the probability amplitude of the electron state $|z\rangle$ to that of the trion state $|Tz\rangle$ (here z is the growth axis of self-assembled QDs which is antiparallel to the optic axis), which results in the net electron spin polarization along $-z$ [5–7].

Here we have experimentally observed the spin polarization process of single electrons in InP/InGaP QDs by time-resolved Kerr rotation (TRKR) measurements. It is found that the inversion of the spin polarization direction occurs by the change of the pump intensity, and that the spin lifetime abruptly changes on the occurrence of the inversion. We discuss these findings by means of numerical simulations using the density operator of the electron–trion four-level system. It suggests that a small deviation of the optical pulse from circular polarization is essential for the inversion. The change of the spin lifetime is attributed to the qualitative change of the four-level system in the electric polarization state.

The outline of this article is as follows. In Section 2, we note the calculation model which is used in the discussion. The experimental observations are explained in Section 3 and discussed in Section 4 by means of the model calculation. The summary of this article is given in Section 5.

2. Calculation model

2.1. Basic Hamiltonian

In what follows, we confine ourselves to consideration of the QD electron–trion system in a magnetic field B along the x axis, which is affected by the classical field of light traveling along $-z$ (Voigt geometry). This situation is depicted in Fig. 1(a). We assume single electrons with spin $s=\frac{1}{2}$ in self-assembled QDs with the growth direction z consisting of zincblende materials. We denote the two energy eigenstates of the electron in the magnetic field $B\|x$ by $|x\rangle=|0\rangle$ and $|\bar{x}\rangle=|1\rangle$. Then the photo-excited trion is made of a hole, having a total angular momentum of $\frac{3}{2}$, and a singlet pair of electrons. We consider only the lowest states of the trion. As they are made principally of the two heavy-hole states $|m_z=\pm\frac{3}{2}\rangle$, we can define a pseudospin space to describe them ($s_T=\frac{1}{2}$), which has two energy eigenstates $|Tx\rangle=|2\rangle$ and $|T\bar{x}\rangle=|3\rangle$ in the field B. Thus we can construct the four-level system represented in Fig. 1(b). The angular frequencies ω_e and ω_h are defined so as to represent the Zeeman splittings of the electron and trion, respectively: $2\hbar\omega_e=g_e\mu_B B$

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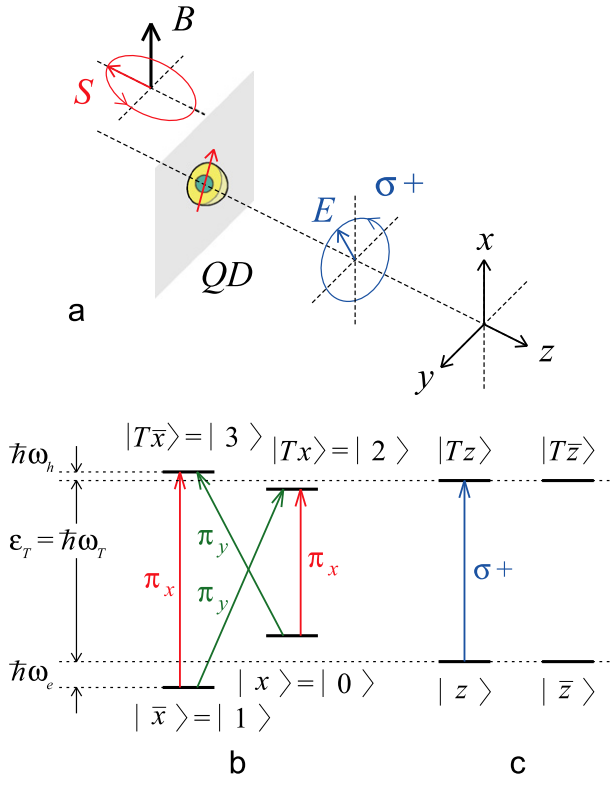

Fig. 1. (a) Schematic representation of the system under consideration. The z axis is defined to be along the growth direction of the QD, in which one conduction electron is present. The optical pulse traveling along $-z$ induces the polarization of the electron spin S, which subsequently precesses around the magnetic field $B\|x$ perpendicular to the z axis (Voigt geometry). The rotating electric field of a $\sigma+$-polarized pulse is expected to induce the spin polarization along $-z$, according to the optical selection rule. (b) Energy eigenstates of the four-level system consisting of the spin states of the electron $|x\rangle = |0\rangle$, $|\bar{x}\rangle = |1\rangle$ and those of the trion (negatively charged exciton) $|Tx\rangle = |2\rangle$, $|T\bar{x}\rangle = |3\rangle$ in the field of $B\|x$. (c) The same spin states shown in the basis of the z axis.

and $2\hbar\omega_h = g_h\mu_B B$. Here μ_B is the Bohr magneton, g_e and g_h are g factors (in the field of $B\|x$) of the electron and trion (heavy hole), respectively. We assume they have positive values in the level configuration of Fig. 1(b). The unperturbed part of the Hamiltonian of the four-level system is expressed as

$$\mathcal{H}_0 = \hbar\omega_e|x\rangle\langle x| - \hbar\omega_e|\bar{x}\rangle\langle\bar{x}| + (\varepsilon_T - \hbar\omega_h)|Tx\rangle\langle Tx| + (\varepsilon_T + \hbar\omega_h)|T\bar{x}\rangle\langle T\bar{x}|, \quad (1)$$

where $\varepsilon_T = \hbar\omega_T$ is the energy separation between the electron and trion levels at $B=0$.

In the dipole approximation, the vertical and cross transitions in the four-level system of Fig. 1(b) can be represented by the dipole moments μ and $i\mu$ which are coupled to optical electric fields of orthogonal linear polarizations π_x and π_y, respectively. As these oscillating moments are $\pi/2$ out of phase with each other (at $B=0$), we have the latter include the phase factor i. Then, in the rotating wave approximation, the interaction part of the Hamiltonian is given by

$$\begin{aligned}\mathcal{V} = &-\hbar\Omega_x(t)\{e^{-i\omega_0 t}|Tx\rangle\langle x| + e^{i\omega_0 t}|x\rangle\langle Tx|\}\\ &-i\hbar\Omega_y(t)\{-e^{-i(\omega_0 t+\delta)}|Tx\rangle\langle\bar{x}| + e^{i(\omega_0 t+\delta)}|\bar{x}\rangle\langle Tx|\}\\ &-\hbar\Omega_x(t)\{e^{-i\omega_0 t}|T\bar{x}\rangle\langle\bar{x}| + e^{i\omega_0 t}|\bar{x}\rangle\langle T\bar{x}|\}\\ &-i\hbar\Omega_y(t)\{-e^{-i(\omega_0 t+\delta)}|T\bar{x}\rangle\langle x| + e^{i(\omega_0 t+\delta)}|x\rangle\langle T\bar{x}|\}. \quad (2)\end{aligned}$$

Here ω_0 is the central frequency of the optical pulse, δ is the relative phase between the π_x- and π_y-polarized waves, $\Omega_x(t)$ and $\Omega_y(t)$ are time-dependent real optical Rabi frequencies for the respective polarizations. The basic Hamiltonian of the system under consideration is the sum of Eqs. (1) and (2),

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{V}. \quad (3)$$

We assume a hyperbolic-secant pulse envelope [4,8]

$$\Omega_0(t) = \Omega\,\mathrm{sech}(\kappa t), \quad (4)$$

where κ is the bandwidth of the pulse, and Ω is a real Rabi amplitude. Then we assume the time-dependent Rabi frequencies to be

$$\Omega_x(t) = \Omega_0(t)\cos\theta,$$

$$\Omega_y(t) = \Omega_0(t)\sin\theta, \quad (5)$$

where $\tan\theta$ determines the ratio Ω_y/Ω_x [9], which is the ratio of the amplitudes of π_x- and π_y-polarized optical electric fields. By tuning the values of δ and θ, we can obtain an arbitrary polarization state of the incident pulse. In the case of $\theta = \pi/4$ ($\Omega_x = \Omega_y$), $\delta = -\pi/2$ gives the circular polarization of $\sigma+$, which connects the states $|z\rangle = (|x\rangle + |\bar{x}\rangle)/\sqrt{2}$ and $|Tz\rangle = (|Tx\rangle + |T\bar{x}\rangle)/\sqrt{2}$ while the states $|\bar{z}\rangle = (|x\rangle - |\bar{x}\rangle)/\sqrt{2}$ and $|T\bar{z}\rangle = (|Tx\rangle - |T\bar{x}\rangle)/\sqrt{2}$ are decoupled from this pulse field.

The Hamiltonian of Eq. (3) is the variation of the existing models [8–10], which deal with coherent optical rotations of the electron spin [4,11,12].

2.2. Equation of motion

Using the Hamiltonian of Eq. (3), we compute the time evolution of the density operator $\rho(t)$ of the four-level system by solving the quantum Liouville equation

$$i\hbar\frac{d}{dt}\rho(t) = [\mathcal{H}, \rho(t)]. \quad (6)$$

For numerical calculation, it is preferable to obtain the equation in the interaction picture removing the fast-oscillating terms with ω_0 or ω_T. With this view, we define the operator $\sigma(t)$ by

$$\rho(t) = \exp\left[-\frac{i}{\hbar}\mathcal{H}_0 t\right]\sigma(t)\exp\left[\frac{i}{\hbar}\mathcal{H}_0 t\right]. \quad (7)$$

Replacing $\rho(t)$ in Eq. (6) with this expression, we obtain the equation for $\sigma(t)$

$$\frac{d}{dt}\sigma(t) = [\mathcal{V}_R, \sigma(t)], \quad (8)$$

where the operator $\mathcal{V}_R$ is defined by

$$i\hbar\mathcal{V}_R = \exp\left[\frac{i}{\hbar}\mathcal{H}_0 t\right]\mathcal{V}\exp\left[-\frac{i}{\hbar}\mathcal{H}_0 t\right]. \quad (9)$$

The specific form of $\mathcal{V}_R$ is

$$\begin{aligned}\mathcal{V}_R = &\,i\Omega_x(t)\{e^{-i\omega_1 t}|Tx\rangle\langle x| + e^{i\omega_1 t}|x\rangle\langle Tx|\}\\ &-\Omega_y(t)\{-e^{-i(\omega_3 t+\delta)}|Tx\rangle\langle\bar{x}| + e^{i(\omega_3 t+\delta)}|\bar{x}\rangle\langle Tx|\}\\ &+i\Omega_x(t)\{e^{-i\omega_4 t}|T\bar{x}\rangle\langle\bar{x}| + e^{i\omega_4 t}|\bar{x}\rangle\langle T\bar{x}|\}\\ &-\Omega_y(t)\{-e^{-i(\omega_2 t+\delta)}|T\bar{x}\rangle\langle x| + e^{i(\omega_2 t+\delta)}|x\rangle\langle T\bar{x}|\}. \quad (10)\end{aligned}$$

Here the frequencies are

$$\omega_1 = \omega_0 + \omega_e - \omega_T + \omega_h,$$

$$\omega_2 = \omega_0 + \omega_e - \omega_T - \omega_h,$$

$$\omega_3 = \omega_0 - \omega_e - \omega_T + \omega_h,$$

$$\omega_4 = \omega_0 - \omega_e - \omega_T - \omega_h. \quad (11)$$

As far as we consider resonant or near-resonant cases ($\omega_0 \simeq \omega_T$), fast-oscillating terms are absent in $\mathcal{V}_R$.