Mechanism of ultrafast spin-polarization switching in nanostructures

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We consider time-dependent processes in the optically excited hybrid system formed by a quantum well (QW) coupled to a remote spin-split correlated bound state. The spin-dependent tunneling from the QW to the bound state results in the nonequilibrium electron spin polarization in the QW. The Coulomb correlations at the bound state enhance the spin polarization in the QW. We propose a mechanism for ultrafast switching of the spin polarization in the QW by tuning the laser pulse frequency between the bound state spin sublevels. Mn-doped core/multishell nanoplatelets and hybrid bound state-semiconductor heterostructures are suggested as promising candidates to prove the predicted effect experimentally. The obtained results open a possibility for spin polarization control in nanoscale systems.

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I. INTRODUCTION

As power consumption in modern electronics becomes one of the central problems, utilization of electron spin is very promising for spintronic devices and information processing [1–3]. It requires precise manipulation/switching of spin polarization [4,5]. Generation and detection of spin-polarized currents is the key problem in the spintronic devices [6–13]. There is a growing interest in semiconductor spin lasers, in which the spin polarized carriers are injected by circularly polarized light or by electrical injection [14–16]. The spin lasers demonstrate threshold reduction [17,18] and gain in polarization degree for spin to optical polarization conversion [19,20]. Perhaps the greatest potential of spin lasers is ultrafast spin and polarization dynamics [15]. Spin-polarized light-emitting diodes also proved promising for the spin injection, a pure circular polarization of the electroluminescence at room temperature with no external magnetic field has been demonstrated [21]. A great progress has been made in stationary spin transport in magnetic [22–25] and nonmagnetic tunnel junctions in the presence of spin-orbit and exchange interactions [26,27] and in quantum dot (QD) systems [28–30] in magnetic field.

For charge and spin control in small devices time-dependent effects and transient processes are essential [9,31–36]. Thus, time evolution of spin and charge configurations in correlated low-dimensional systems is of great interest both from fundamental and technological points of view. Time-dependent characteristics also provide an important information about the properties of nanoscale systems. Nowadays, there are various experimental methods (polarization photoluminescence (PL), magneto-optical Faraday/Kerr effect, etc.) for time-resolved detection of the spin polarization [1,37].

One of the most perspective ideas of controlling spin polarization is based on the carriers spatial separation. Hybrid bound state-semiconductor heterostructures formed by a QW and bound states (such as magnetic impurity ions) separated by a thin spacer seems to be good candidates to realize this idea [38–41]. Usually, spin-polarized carriers are injected from the bound state (ferromagnetic δ layer) into the semiconductor QW; the magnetic properties and spin polarization of the carriers can be controlled via the spacer thickness, shape, and the δ-layer parameters [42–45]. This method allows us to obtain spin polarization, however, with no control on its time evolution. Another mechanism of dynamic spin polarization of electrons due to the spin-dependent tunneling from a semiconductor QW into the bound state spin-split by exchange interaction was proposed and realized experimentally [46,47]. Linearly polarized laser pulse creates nonpolarized electrons in the QW. Spin-dependent tunneling into the bound state results in accumulation of the electron spin polarization in the QW detected by the circular polarization of the PL.

In this paper we analyze the dynamic spin polarization theoretically. We extend the formalism considered in Refs. [48–54]. It was first applied to the correlated QD coupled to a reservoir to describe the nonstationary spin polarized currents due to the time evolution of a magnetic moment in the QD under applied bias and external magnetic field. In Refs. [48,49] the spin generation due to the spin-dependent tunneling in the hybrid QW-bound state system was explained. In the present paper we generalize the nonstationary approach to the case when initial nonequilibrium carriers distribution in the QW is tuned by a laser pulse. We show that the spin polarization in QW and circular polarized PL change their signs when the pulse frequency is tuned to match one of the bound state spin sublevels. Our results open

the possibility to control the sign of electrons spin polarization in the QW.

II. THEORETICAL MODEL

We consider nonstationary processes in the system formed by the QW coupled to a spin-split correlated bound state with the energy ε_1 separated from the QW by a tunnel barrier [see Fig. 1(a)]. At the initial time QW is optically excited with linearly polarized light generating unpolarized nonequilibrium electrons with energies ε_k, where k is the in-plane vector. The barrier is characterized by the tunneling rate Γ_{wi}. The electron-hole recombination processes in the QW are described by the relaxation rate γ_w; a separate relaxation channel at the bound state allows electrons to disappear with the rate γ_i. The suggested model gives the possibility to analyze dynamic spin injection processes caused by the spin-dependent tunneling between the QW and bound state considering exactly high order correlation functions for the bound state electrons. The Hamiltonian of the system consists of the QW part, the bound state part, which includes the Hubbard term corresponding to the on-site Coulomb repulsion, and the tunneling part describing electrons transfer between the QW and the bound state:

$$\hat{H} = \sum_{\sigma,k} \varepsilon_k \hat{c}^+_{k\sigma}\hat{c}_{k\sigma} + \sum_{\sigma}\left(\varepsilon_1 \hat{n}^{\sigma}_1 + U\hat{n}^{\sigma}_1\hat{n}^{-\sigma}_1\right) + \sum_{k\sigma} t_k(\hat{c}^+_{k\sigma}\hat{c}_{1\sigma} + \hat{c}^+_{1\sigma}\hat{c}_{k\sigma}). \quad (1)$$

Here index k labels continuous spectrum states in the QW; t_k is the tunneling amplitude between a QW state k and the bound state. The bound state energy level ε_1 can be split by an exchange interaction or a weak external magnetic field into two spin sublevels: $\varepsilon_\sigma = \varepsilon_1 + \sigma\Delta$, where $\sigma = \pm 1/2$ is the electron spin projection and Δ is the splitting energy. Operators $\hat{c}^+_{k\sigma}$ ($\hat{c}_{k\sigma}$) are the creation (annihilation) operators for the QW states. $\hat{n}^{\sigma}_1 = \hat{c}^+_{1\sigma}\hat{c}_{1\sigma}$ is the bound state electron occupation number, where operator $\hat{c}_{1\sigma}$ destroys the electron with spin projection σ. U is the on-site Coulomb repulsion for the double occupation of the bound state.

We neglect the tunneling of holes between the QW and the bound state as it is usually less efficient than for electrons due to the difference in the effective mass. The holes contribution to the resulting nonequilibrium spin polarization is negligible as their spin relaxation is much faster than for the electrons [46].

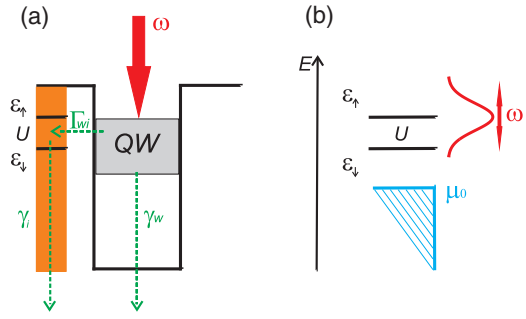

FIG. 1. (a) Scheme of the model structure under external laser excitation with frequency ω. (b) Schematic energy diagram showing the bound state split energy levels, initial and equilibrium electrons distribution in the QW.

III. NONSTATIONARY ELECTRONIC TRANSPORT FORMALISM

Let us further consider $\hbar = 1$ and $e = 1$ elsewhere and assume the low temperature regime. The equations of motion for the electron operators products $\hat{n}^{\sigma}_1$, $\hat{n}^{\sigma}_{1k} = \hat{c}^+_{1\sigma}\hat{c}_{k\sigma}$, and $\hat{n}^{\sigma}_{k'k} = \hat{c}^+_{k'\sigma}\hat{c}_{k\sigma}$ can be written as:

$$i\frac{\partial \hat{n}^{\sigma}_1}{\partial t} = -\sum_{k,\sigma} t_k \cdot (\hat{n}^{\sigma}_{k1} - \hat{n}^{\sigma}_{1k}), \quad (2)$$

$$i\frac{\partial \hat{n}^{\sigma}_{1k}}{\partial t} = -(\varepsilon^{\sigma}_1 - \varepsilon_k)\cdot \hat{n}^{\sigma}_{1k} - U\cdot \hat{n}^{-\sigma}_1 \hat{n}^{\sigma}_{1k} + t_k \cdot (\hat{n}^{\sigma}_1 - \hat{n}^{\sigma}_k) - \sum_{k'\neq k} t_{k'}\cdot \hat{n}^{\sigma}_{k'k}, \quad (3)$$

$$i\frac{\partial \hat{n}^{\sigma}_{k'k}}{\partial t} = -(\varepsilon_{k'} - \varepsilon_k)\cdot \hat{n}^{\sigma}_{k'k} - t_{k'}\cdot \hat{n}^{\sigma}_{1k} + t_k \cdot \hat{n}^{\sigma}_{k'1}. \quad (4)$$

Substituting the solution of Eq. (4) into Eq. (3) reveals the relaxation term $i\Gamma_{wi}\hat{n}^{\sigma}_{1k}$ due to the tunneling with the rate $\Gamma_{wi} = \pi\nu_0 t_k^2$ determined by the unperturbed density of states ν_0 and the tunneling amplitude t_k [48]. Further we assume ν_0 constant for 2D electrons and t_k. independent of k. The equations for the bound state occupation numbers $n^{\pm\sigma}_1$ are further obtained by averaging operator equations (2)–(4) and by decoupling QW electrons occupation numbers from the bound state occupation numbers [48]. Within the decoupling procedure the operators $\hat{n}^{\sigma}_k$ are replaced with the distribution function f^{σ}_k. Assuming that equilibrium state corresponds to the empty bound state and equilibrium Fermi distribution of electrons in the QW the following equations can be obtained [48]:

$$\frac{\partial n^{\sigma}_1}{\partial t} = -2\cdot\Gamma_{wi}\cdot I^{\sigma}_k - \gamma_i \cdot n^{\sigma}_1,$$
$$\frac{\partial f^{\sigma}_k}{\partial t} = 2\cdot\Gamma_{wi}\cdot J^{\sigma}_k - \gamma_w\cdot(f^{\sigma}_k - f^0_k), \quad (5)$$

where

$$I^{\sigma}_k = n^{\sigma}_1 - (1 - n^{-\sigma}_1)\cdot\Xi(\varepsilon_\sigma) - n^{-\sigma}_1\cdot\Xi(\varepsilon_\sigma + U)$$
$$J^{\sigma}_k = \frac{1}{\nu_0\pi}\cdot\left[(1-n^{-\sigma}_1)(n^{\sigma}_1 - f^{\sigma}_k)\cdot\frac{\Upsilon}{(\varepsilon_\sigma - \varepsilon_k)^2 + \Upsilon^2} + \frac{n^{-\sigma}_1(n^{\sigma}_1 - f^{\sigma}_k)\Upsilon}{(\varepsilon_\sigma + U - \varepsilon_k)^2 + \Upsilon^2}\right] \quad (6)$$

and QW occupation function $\Xi(x)$ with $x = \varepsilon_\sigma, \varepsilon_\sigma + U$ reads:

$$\Xi(x) = \int d\varepsilon_k \cdot f^{\sigma}_k(\varepsilon_k)\cdot\frac{1}{\pi}\frac{\Upsilon}{(x-\varepsilon_k)^2 + \Upsilon^2}. \quad (7)$$

In Eqs. (5) we have introduced relaxation rates γ_w and γ_i describing relaxation processes in the QW and at the bound state, respectively. The relaxation rate $\Upsilon = \Gamma_{wi} + \gamma_i$ is introduced to describe correctly the bound state structure; it accounts for its broadening due to both tunneling and relaxation [48].

In the absence of the tunnel coupling the electrons in the QW are described by Fermi distribution f^0_k with a chemical

potential μ_0 and a temperature T_0. Let us consider an optical excitation by a short laser pulse with Gaussian spectral distribution. By tuning the laser wavelength the peak of the excited nonequilibrium electron distribution in the QW could be put in a resonance with one of the bound state spin sublevels as shown in Fig. 1(b). For the bound state the initial conditions are: $n_1^\sigma = n_1^{-\sigma} = 0$. The tunneling of the QW electrons into the bound state leads to the renormalization of the stationary distribution function in the QW. Solving Eqs. (5)–(7) in the stationary case ($\frac{\partial n_1^\sigma}{\partial t} = \frac{\partial \tilde{n}_k^\sigma}{\partial t} = 0$) one can get stationary bound state occupation numbers:

$$n_1^{\sigma\, st} = \frac{\Phi(\varepsilon_\sigma) - \Delta\Phi^\sigma \cdot \Phi(\varepsilon_{-\sigma})}{1 - \Delta\Phi^\sigma \cdot \Delta\Phi^{-\sigma}}, \tag{8}$$

where

$$\Phi(\varepsilon_\sigma) = \frac{2\Gamma_{wi}}{2\Gamma_{wi} + \gamma_i} \cdot \Xi(\varepsilon_\sigma),$$
$$\Delta\Phi^\sigma = \Phi(\varepsilon_\sigma) - \Phi(\varepsilon_\sigma + U). \tag{9}$$

Functions $\Phi(\varepsilon_\sigma)$ are determined with stationary distribution functions f_k^{st}, which can be found from Eqs. (5)–(7). Solution of Eqs. (5)–(7) in the stationary case reveals the presence of residual spin polarization for electrons in the QW for $\Gamma_{wi}/\gamma_w \ll 1$. The spin polarization given by $\rho_s = N_\uparrow - N_\downarrow$, where $N_\sigma = \int f_\sigma(\varepsilon_k) d\varepsilon_k$, results in the circular polarization of the PL from the QW:

$$P_{PL} \sim \frac{N_\uparrow - N_\downarrow}{N_\uparrow + N_\downarrow}. \tag{10}$$

The polarization degree P_{PL} is proportional to the spin polarization of the electrons in the QW. The coefficient depends on the radiative recombination details, in particular, on the occupation of heavy and light hole subbands [2]. In our model spin polarization of the electrons in the QW appears due to the tunnel leakage. So, for the considered effect we neglect the influence of the valence band structure on the QW electrons spin polarization. This assumption is valid for not too wide QWs with separated heavy and light holes subbands.

IV. RESULTS AND DISCUSSION

Kinetics of the photoexcited electrons in the QW is characterized by the recombination processes with a typical time γ_w^{-1} and tunneling between the QW and the bound state with a time Γ_{wi}^{-1}. Figure 2 shows the kinetics of the spin polarization in the QW. At a small time the spin-dependent tunneling leads to a linear increase of electron spin polarization [48,49], it further approaches its stationary value given by the equilibrium carries distribution in the QW.

The Coulomb correlations strongly influence the carriers dynamics when the carrier lifetime at the bound state exceeds the tunneling time, which is in its turn smaller than the relaxation time in the QW:

$$\gamma_i^{-1} \gg \Gamma_{wi}^{-1}; \quad \Gamma_{wi}^{-1} \leqslant \gamma_w^{-1}. \tag{11}$$

The role of Coulomb correlations and bound state energy in the nonequilibrium spin polarization of the photoexcited electrons is shown in Fig. 2. We assume the initial Fermi distribution of the photoexcited electrons in the QW with a chemical potential μ^*. In the following calculations we

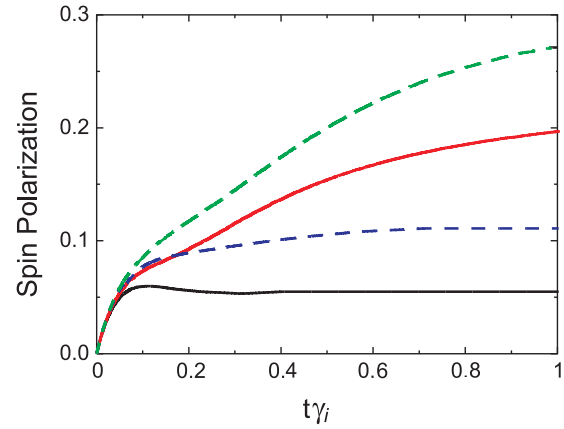

FIG. 2. Time evolution of the spin polarization in the QW. Solid black ($U = 0$) and red ($U = 1$) lines: $\varepsilon_\uparrow = 0.35$, $\varepsilon_\downarrow = 0.25$, $\mu^* = 0.3$. Dashed blue ($U = 0$) and green ($U = 1$) lines: $\varepsilon_\uparrow = 0.2$, $\varepsilon_\downarrow = 0.1$, $\mu^* = 0.15$. Parameters are $\mu_0 = 0$, $W = 2$, $\gamma_i = 0.005$, $\Gamma_{wi} = 6\gamma_i$, $\gamma_w = 5\gamma_i$.

neglect thermal broadening of the distribution and consider the energy relaxation via the hole recombination processes. The thermal broadening and energy relaxation via electron-phonon interaction affects the tunneling rate. However, it is less important than the electron-hole recombination, as the latter directly changes the number of carriers in the QW. The calculations were performed following the Eqs. (5)–(7). Two main effects can be clearly seen in Fig. 2. Firstly, the presence of Coulomb correlations increases spin polarization. Secondly, spin polarization of photoexcited electrons (and, thus PL circular polarization) is sensitive to the relative position of the equilibrium distribution chemical potential $\mu_0 = 0$ and the bound state spin split levels. It substantially increases when the bound state energy levels are located closer to the equilibrium chemical potential μ_0.

Our theory predicts an effect, the ultrafast switching of spin polarization of electrons in a QW with a laser pulse. By tuning the excitation laser frequency the nonequilibrium distribution maximum of the excited electrons can be shifted along the energy scale matching one of the spin-split bound state energy levels [see Fig. 1(b)].

Figure 3 shows calculation results for the Gaussian energy distribution of photoexcited electrons in the QW. Consequently, the tunneling of the electrons with the corresponding spin projection into the bound state becomes more effective and causes spin polarization of the resident electrons in the QW. Changing the maximum of the electron energy distribution between the two bound state spin sublevels results in the switching of the spin polarization and, subsequently the reversal of the circular PL polarization sign [Fig. 3(a)]. This finding opens a possibility to generate spin polarized train pulses with opposite polarization as illustrated in Fig. 3(b). Here the nonpolarized electrons in the QW are generated by laser train pulses with the peak of the laser spectrum alternating between the two spin sublevels of the bound state. Consequently, spin polarization of the electrons in the QW changes its sign from one pulse to another. We do not consider the process of the nonequilibrium electrons generation assuming they are created instantly by the laser pulse. The increasing part of

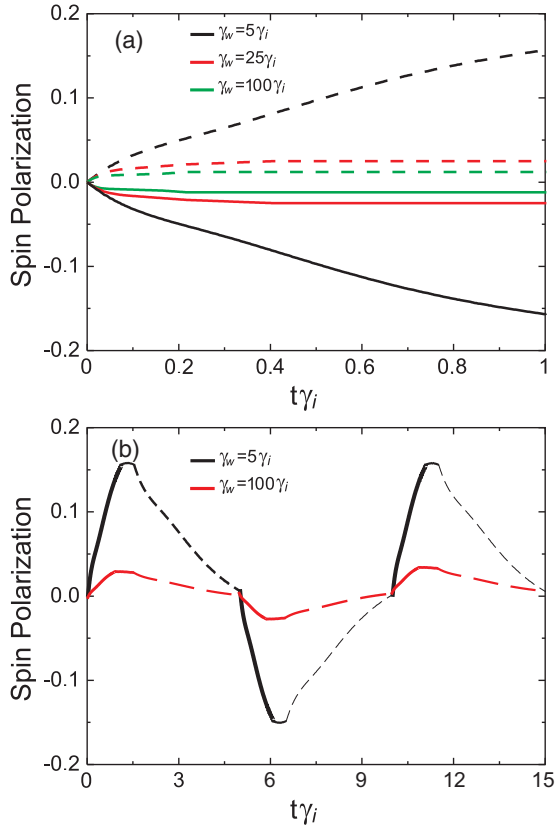

FIG. 3. (a) Time evolution of the spin polarization in the QW. Dashed lines: $\omega = \varepsilon_\uparrow = 0.35$. Solid lines: $\omega = \varepsilon_\downarrow = 0.25$. (b) Switching of the spin polarization sign by tuning the laser frequency between $\omega = \varepsilon_\uparrow = 0.35$ and $\omega = \varepsilon_\downarrow = 0.25$. Parameters are $\mu_0 = 0$, $U = 1$, $W = 2$, $\gamma_i = 0.005$, $\Gamma_{wi} = 6\gamma_i$.

the spin polarization response pulse indicated by the solid lines in Fig. 3(b) is determined by the spin inertia time [55], which is in our system the inverse tunneling rate Γ_{wi}^{-1}. The full decay of the spin polarization is due to spin relaxation; this time is assumed to be the longest on the problem timescale ($\sim$10 ns [46]). The spin relaxation is not accounted for in our theory, so the decay of the spin polarization pulse is shown schematically by the dashed line representing an exponent with the characteristic time $t \approx \gamma_i$. So, Fig. 3(b) describes the case of the the interval between the laser pulses exceeding spin relaxation time. The red line illustrates the case of a very fast recombination in the QW exceeding the tunneling rate. The amplitude of the polarization appears to be small as the carriers in the QW relax to equilibrium faster than the polarization develops.

The requirements to observe the predicted spin polarization switching are (i) possibility to create nonequilibrium distribution of photoexcited electrons and (ii) inverse tunneling rate should not exceed the time of the electron distribution thermalization. Promising candidates for experimental observation are, for example, Mn-doped colloidal core/multishell nanoplatelets [56,57] or semiconductor heterostructures formed by several QWs separated by the barriers with different width and height [46,47]. In the latter case one of the QWs is doped with Mn. In both cases the pump-probe technique (with both pump and probe laser pulses linearly polarized) should be used to reveal the PL circular polarization. Linearly polarized pump pulse creates nonequilibrium distribution of the electrons in the core of the nanoplatelet (for the first configuration) or in the QW without Mn dopant atoms (for the second configuration). The spin-dependent tunneling of the electrons to the Mn impurity states leads to the spin polarization of the electrons in the core or QW and can be detected by the emergent polarization of the probe pulse.

To observe the polarization sign switching the inequalities Eqs. (11) should be fulfilled. The radiative recombination rate in the QW is of the order of 1 ns. The bound state relaxation time γ_i^{-1} is determined by recombination processes; its value can vary from 1 ns down to 10 ps [49,58]. The tunneling rate extracted from experiments on dynamic spin injection in semiconductor heterostructures [46,47,59,60] $\Gamma_{wi} \sim$ 1–100 ps^{-1} depending on the barrier thickness [61], which can be also tuned by an external bias. Typical relaxation rate of the nonequilibrium electron distribution excited by the laser pulse is about a few picoseconds, for example in CdSe thin films it was reported to be 1–5 ps [62]. So, inequalities Eqs. (11) can be realized experimentally.

Another important parameter is the magnitude of the bound state spin splitting. As the typical width of the nonequilibrium electron distribution excited by the 1 ps laser pulse is about 0.5 meV, for the effective spin-dependent tunneling the spin splitting should exceed this value. The appropriate values of the spin splitting are typical for the semiconductor superstructures with magnetic impurities due to exchange interaction. For example, in (Ga,Mn)As heterostructures the magnitude of the exchange induced splitting is about 2.5 meV [47,58]. It can be also tuned by an external magnetic field.

V. CONCLUSION

We considered time-dependent processes in the system formed by a QW coupled to a remote spin-split correlated bound state. It was shown that Coulomb interaction at the bound state leads to the significant increase of the spin polarization in the QW. The residual spin depends on the equilibrium Fermi level in the QW. We propose a mechanism for ultrafast switching of the spin polarization in the QW. As energy distribution of photoexcited electrons depends on the excitation laser pulse spectrum, the spin-dependent tunneling efficiency can be controlled by matching of the laser pulse frequency to the spin split bound state energy. The time-dependent electron spin polarization in the QW and, consequently, the circular polarized PL could reverse the sign depending on the laser pulse frequency. We suggested possible hybrid low-dimensional structures as candidates to probe the predicted effect and discussed the conditions necessary for the experimental observation. We believe that these results open a possibility for spin polarization control in nanoscale systems.

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