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Optical electron spin pumping in n-doped quantum wells

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Abstract

A theoretical model for optical spin pumping of electrons in a quantum well with low intrinsic electron density is presented. A system of electrons under continuous-wave illumination by circularly polarized light tuned to the electron–trion resonance is considered. The simultaneous off-resonant creation of excitons is also taken into account. The spin flip of trions and their radiative decay as the basic processes which allow the electronic spin pumping, as well as other processes, such as the formation of trions from excitons and electrons, are accounted for in the appropriate kinetic equations. The results obtained for CdTe and GaAs quantum wells indicate that significant electron spin polarization can be achieved in a time range of a few nanoseconds.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

The exciting possibility of using the spin of an electron rather than electron charge as an information carrier in electronics has renewed interest in studies of active manipulation of spin degree of freedom in solid-state systems. A sufficient initialization of the electron spin polarization is one of the basic requirements for such studies and for the functionality of present and future spintronic devices [1]. One of the known methods of the spin polarization is the injection of electrons into a semiconductor media through a spin aligner which is usually the magnetic or semimagnetic contact [2–6] (recently users of this technique claim 85% of spin polarization [2, 3]). Another way to obtain the spin polarization is the optical excitation of the semiconducting material with circularly polarized light which creates spin polarized electrons and holes [1, 7]. In direct gap semiconductors, for example in bulk GaAs, the ratio of excited electrons having opposite spin polarizations can reach 3:1 due to the selection rules of the optical transitions from the heavy and light hole subbands. In low-dimensional structures, because of the split-off of the heavy and light hole subbands, the electron spin polarization can in principle reach 100%. However, the spin polarization of electrons excited from the valence band is transient. Its persistence is limited mainly by the lifetime of recombining carriers. In order to extend the lifetime of the spin polarization, the separation of electrons and holes is needed.

In this paper we discuss another possibility to obtain a net spin polarization in semiconductors. It is based on electron spin pumping with the use of a circularly polarized laser light creating trions X^- as intermediate states. This method can be an alternative to those described above having several possible advantages. It makes it possible to create electronic polarization without the need of building heterostructures with ferromagnets or semimagnetic semiconductors. Unlike the optical direct excitation of electron and hole spin polarized pairs it does not require the use of high electric fields in order to prevent the particles' recombination. The light can be addressed locally in the places where the spin polarization is needed in the device.

In our previous work [8] we proposed a scheme of electron spin pumping in bulk CuCl with a small homogeneous concentration of confined electrons in the sample. Taking into account the large binding energy of the trion in CuCl we assumed the possibility of intense continuous coherent excitation of electrons to X^- trions by circularly polarized light. Due to the selection rules, trions X^- decay spontaneously into the electrons in whichever spin state. Since circularly polarized light interacts only with electrons in one of the two possible spin states, a significant increase of the population of electrons with opposite spin may be achieved.

The use of trions as the intermediate states in electron spin pumping has already been proposed by other authors for electrons confined in quantum structures. The binding energy

of charged excitons is much higher in small dimensional structures than in bulk materials and for certain concentrations of confined electrons the negatively charged exciton lines can dominate optical absorption and emission spectra [9, 10]. Shabayev *et al* [11] suggested the use of the heavy hole trion X^- as an intermediate state for feasible initialization of the electronic spin in the case of a single electron in a quantum dot. To obtain well-defined spin polarization, regardless of its initial state, they suggested using a circularly polarized optical π pulse combined with a π pulse of transverse magnetic field and successive spontaneous trion decay. Recently Gerardot *et al* [12] have demonstrated high fidelity (99%) optical pumping of the heavy hole spin in a quantum dot with the use of circularly polarized light via the charged exciton state X^+. The use of the magnetic π pulse is not needed in this case. The coherent rotation of the trion's electron spin in the magnetic filed is replaced by its incoherent evolution due to the hyperfine interaction with spins of the nuclei.

In the case of quantum wells, the interplay between electrons and trions as well as spin pumping via trions was considered by Hoffmann *et al* [13] and Tribollet *et al* [14]. Since they used small laser power density or short laser impulses they observed small polarization. In order to study the possibility of gaining high spin polarization in a quantum well (QW) we assume in our model continuous laser illumination with relatively large power density. Consequently, besides the resonant electron to trion transition we have to take into account the resonant trion to electron transition as well as the off-resonant creation of excitons.

In part 2 of our paper we discuss the general scheme of the spin pumping. It is different from that in the bulk CuCl case where the main channel was given by the spontaneous trion decay into the inverted electron spin state. In the two-dimensional case the trions are built with heavy holes and the channel described above is closed because of the selection rules. Thus the spin polarization in the two-dimensional (2D) case can be guaranteed only by spin flip of trions followed by their decay. To estimate the possible final polarization of the system we built an appropriate set of kinetic equations. The needed rates of induced transitions are deduced in part 3 and the phenomenological lifetimes of decay and spin-flip processes are discussed in part 4. We solve our equations and discuss the results in part 5. For the electron spin relaxation times $\tau_s \simeq 5$ ns in CdTe QW [13] and $\tau_s \simeq 10$ ns in GaAs QW [9], we predict the possibility of getting the significant electron spin polarization up to 80%.

2. Scheme of spin pumping

We consider a 2D quantum well with a small concentration of confined excess electrons. Let us consider a circularly polarized σ^+ plane-wave continuous light signal entering the sample in the positive z direction. Let us assume the light frequency is in resonance with the energy of the electron to heavy hole trion transition. The heavy hole trion has a total angular momentum projection onto the z direction equal to $+3/2$ or $-3/2$ (we will denote these two trion states by $X^-_{\pm 3/2}$) and is made of two electrons and a heavy hole. The

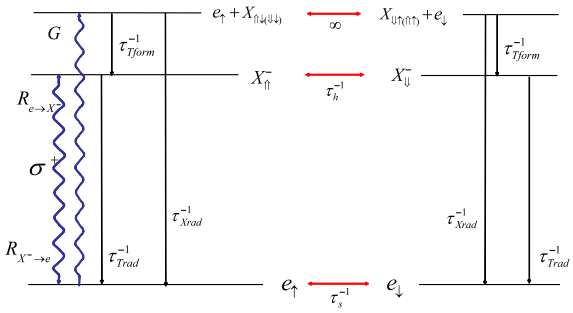

Figure 1. Scheme of transitions between electron, exciton and trion states. Wavy lines denote transitions induced by σ^+ circularly polarized laser light. Spontaneous radiative transitions are denoted by the arrows and are accompanied by the rates of decay, while the spin-flip transitions are denoted by horizontal arrows and are accompanied by the spin-flip rates. The arrows $\uparrow$ and $\downarrow$ denote the electron spin projections $+(1/2)$ and $-(1/2)$, $\Uparrow$ and $\Downarrow$ denote the hole spin projections $+(3/2)$ and $-(3/2)$.

electrons are in a spin singlet so the total angular momentum of the trion is defined by the hole spin state. For σ^+ light polarization the only possible induced optical transitions occur between electrons with spin $+1/2$ and the trions $X^-_{+3/2}$. The created trions $X^-_{+3/2}$ can decay (it can occur spontaneously or be induced by light) into the initial electron spin state $+1/2$. However, if the spin of the trion's hole flips first ($X^-_{+3/2} \to X^-_{-3/2}$) then trions can also decay spontaneously into electrons $e_{-1/2}$. Since a σ^+ light signal does not affect the electrons $e_{-1/2}$, the population of electrons with spin $-1/2$ increases, thus producing the net electron spin polarization.

Unfortunately in addition to the process described above there is also another one which can disturb the spin pumping. The proximity of neutral and charged exciton lines in the absorption spectra makes possible the creation of a neutral exciton (X) accompanying the electron–trion transition. A circularly polarized σ^+ photon can produce a neutral exciton X_{+1} with the total angular momentum $J = 1$ parallel to the light propagation. The exciton spin-flip processes can cause transitions to the exciton states X_{-1} with antiparallel angular momentum projection as well as to the $J = 2$ states $X_{\pm 2}$. Excitons usually recombine within a time of the order of 100 ps. However, before they decay they can collide with electrons. Excitons X_{+1} and X_{-2} can absorb an electron $e_{+1/2}$ forming trions $X^-_{+3/2}$ or $X^-_{-3/2}$, which thereafter can take part in the pumping process as described before. Excitons X_{-1} and X_{+2} can capture electrons $e_{-1/2}$, which in turn is an undesirable process for the described spin pumping. It is usually assumed (and we do the same in our model) that the spin-flip processes within the four exciton states ($J = 1$ and 2) are faster than the exciton radiative decay and faster than the time needed for collision with an electron [10, 15]. We assume that the rate of the trion formation from an exciton is strictly proportional to the concentration $n_{e\uparrow}$ of electrons with spin $+1/2$ when trion $X^-_{+3/2}$ is formed, and to the concentration $n_{e\downarrow}$ in the opposite case.

We present in figure 1. the scheme of electron spin pumping with all the mentioned processes accounted for. The induced transitions between electrons $e_{+1/2}$ and trions $X_{+3/2}$

are indicated by $R_{e\to X^-}$ and $R_{X^-\to e}$, respectively. The rate of generation of neutral excitons is indicated by G, while the induced exciton recombination is not taken into account which will be explained in section 3. The rates of spontaneous processes are characterized by the following time constants: the radiative lifetimes τ_{Trad} of the trion and τ_{Xrad} of the exciton, the formation time τ_{Tform} of the trion, the spin relaxation time τ_s of the electron and the spin relaxation time τ_h of the hole being the part of the trion.

Here we are interested in the electronic spin polarization in a long timescale, much longer than the characteristic times of incoherent dumping processes such as spontaneous trion recombination and other lifetime broadening mechanisms. Therefore, we describe the spin pumping by kinetic equations neglecting coherent effects. In our description of the optical spin pumping we use the following set of equations:

$$\begin{aligned}
\dot{e}_\uparrow &= -e_\uparrow \cdot R_{e\to X^-} + T_\Uparrow \cdot (R_{X^-\to e} + \tau_{Trad}^{-1}) \\
&\quad + (e_\downarrow - e_\uparrow)\cdot \tau_s^{-1} - X\cdot e_\uparrow \cdot \tau_{Tform}^{-1} \\
\dot{T}_\Uparrow &= e_\uparrow \cdot R_{e\to X^-} - T_\Uparrow \cdot (R_{X^-\to e} + \tau_{Trad}^{-1}) \\
&\quad + (T_\Downarrow - T_\Uparrow)\cdot \tau_h^{-1} + X\cdot e_\uparrow \cdot \tau_{Tform}^{-1} \qquad (1) \\
\dot{e}_\downarrow &= T_\Downarrow \cdot \tau_{Trad}^{-1} + (e_\uparrow - e_\downarrow)\cdot \tau_s^{-1} - X\cdot e_\downarrow \cdot \tau_{Tform}^{-1} \\
\dot{T}_\Downarrow &= -T_\Downarrow \cdot \tau_{Trad}^{-1} + (T_\Uparrow - T_\Downarrow)\cdot \tau_h^{-1} + X\cdot e_\downarrow \cdot \tau_{Tform}^{-1} \\
\dot{X} &= G\cdot n_0^{-1} - X\cdot \tau_{Xrad}^{-1} - X\cdot (e_\uparrow + e_\downarrow)\cdot \tau_{Tform}^{-1},
\end{aligned}$$

where we use $e_{\uparrow(\downarrow)}$, $T_{\Uparrow(\Downarrow)}$ and X to denote the concentrations, relative to the confined electron concentration n_0, of electrons, trions and neutral excitons, respectively (we will simply call them concentrations, having in mind that they are given in n_0 units). The arrows $\uparrow$ and $\downarrow$ denote the electron spin projections $+(1/2)$ and $-(1/2)$, $\Uparrow$ and $\Downarrow$ denote the hole spin projections $+(3/2)$ and $-(3/2)$. X is the total concentration of the neutral excitons $X_{\pm1}$ and $X_{\pm2}$. We assume all these four exciton states to be equally populated according to the assumption of the fast spin-flip processes within excitons and relatively slow rate of exciton creation. We would like to note that from the above definitions and from equation (1) the conservation condition $e_\uparrow + e_\downarrow + T_\Uparrow + T_\Downarrow = 1$ arises.

The required rates R of induced electron–trion transitions as well as the rate G of induced exciton generation are calculated in the next section and the estimates for the time constants are given in section 4.

3. Induced optical transitions

The circularly polarized (σ^+) plane-wave light of frequency ω, which enters and traverses a thin sample in the positive direction z is represented by the electric field $\mathbf{E}(t) = (\sqrt{2}\mathcal{E})[\hat{\mathbf{x}}\cos(\omega t - qz) + \hat{\mathbf{y}}\sin(\omega t - qz)]$, where $q = n\omega/c$ and n denotes the refractive index of a medium.

The electronic properties of a medium are described in the frame of a two band model with band extremes at $\mathbf{k} = 0$. The conduction band electrons in the vicinity of $\mathbf{k} = 0$ are described by the Bloch wavefunctions

$$c_{\pm1/2} = S|\pm 1/2\rangle,$$

and the heavy hole with angular momentum $J = 3/2$, by the valence Bloch function

$$v_{\pm3/2} = \frac{1}{\sqrt{2}}(X \pm iY)|\pm 1/2\rangle,$$

where $|1/2\rangle$ and $|-1/2\rangle$ are the eigenstates of the z-component of the spin operator $\hat{\mathbf{s}}$. The wavefunctions S, X, Y have symmetry of s, p_x and p_y orbitals, respectively.

In the quantum mechanical description of optical transitions we consider only a two-level wavefunction $\psi = a_1(t)\psi_1 + a_2(t)\psi_2$, with time dependent population coefficients $a_1(t)$, $a_2(t)$ and functions ψ_1, ψ_2 assigned to initial (1) and final (2) states, respectively. Assuming that the initial state is completely populated before dipole transition is induced we have $a_1(0) = 1$, $a_2(0) = 0$. The probability of transition $1 \to 2$ is then given by

$$|a_2(t)|^2 = \frac{\Omega^2}{\Omega^2 + (\Delta\omega)^2}\sin^2\frac{\sqrt{\Omega^2 + (\Delta\omega)^2}}{2}t, \qquad (2)$$

where $\Omega = 2\mathcal{E}d_{12}/\hbar$ is the characteristic Rabi frequency, d_{12} is the magnitude of electric dipole transition momentum and $\Delta\omega = \omega - \omega_{12}$ where ω_{12} is the absolute value of a frequency of absorbed or emitted photons. For a weak electric field $\mathcal{E}$ the rate of transition from level 1 to level 2 $w_{12} = \mathrm{d}|a_2(t)|^2/\mathrm{d}t$ has a significant value only for $\Delta\omega \approx 0$ and in the limit $t \to \infty$ it becomes

$$w_{12} = (\pi/2)\Omega^2\delta(\omega - \omega_{12}). \qquad (3)$$

3.1. Electron–trion transition rates

The only possible optical electron–trion transitions induced by σ^+ polarized light are between the electron state $|e_{\mathbf{k},+1/2}\rangle$ with spin projection $+1/2$ and the trion state $|X^-_{\mathbf{k},+3/2}\rangle$ with the total angular momentum projection $+3/2$ onto the positive z axis. Because of the orthogonality of the photon wavevector $\mathbf{q}$ to the in-plane momenta of carriers, the wavevector $\mathbf{k}$ of the electron as well as the wavevector of the trion coincide. In the occupation number formalism the electron state can be presented as $|e_{\mathbf{k},+1/2}\rangle = a^+_{\mathbf{k},+1/2}|g\rangle$, while the trion state (in the effective mass approximation) is assumed to be the following linear combination

$$\begin{aligned}
|X^-_{\mathbf{k},+3/2}\rangle &= \frac{1}{\sqrt{2}}\sum_{\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_h} C(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_h) \\
&\quad \times (a^+_{\mathbf{k}_1,+1/2}a^+_{\mathbf{k}_2,-1/2} - a^+_{\mathbf{k}_1,-1/2}a^+_{\mathbf{k}_2,+1/2})d^+_{\mathbf{k}_h,+3/2}|g\rangle. \qquad (4)
\end{aligned}$$

Here $|g\rangle$ denotes the electronic state corresponding to the empty conduction and fully occupied valence band, and $a^+_{\mathbf{k},m_j}$ ($d^+_{\mathbf{k},m_j}$) denote the creation operator of an electron (hole) in the Bloch state with the wavevector $\mathbf{k}$ and projection of the total angular momentum m_j. The linear coefficients $C(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_h)$ are the Fourier transforms of the trion X^- envelope. They do not vanish only for $\mathbf{k}_1 + \mathbf{k}_2 + \mathbf{k}_h = \mathbf{k}$. The electric dipole moment of the electron–trion transition can be expressed as $d_{\mathbf{k}} = p_{cv}\mathcal{M}(\mathbf{k})e/m_0\omega_{\mathbf{k}}$, where $p_{cv} = \langle X|p_x|S\rangle$ and $\hbar\omega_{\mathbf{k}}$ is the energy difference between the electron and trion $\mathbf{k}$-states. The optical matrix element $\mathcal{M}(\mathbf{k}) = \sum_{\mathbf{k}'} C(\mathbf{k},\mathbf{k}',\mathbf{k}')$ [16] is well approximated by $|\mathcal{M}(\mathbf{k})|^2 = c_1\exp(-\epsilon/\epsilon_1)$ [10], where

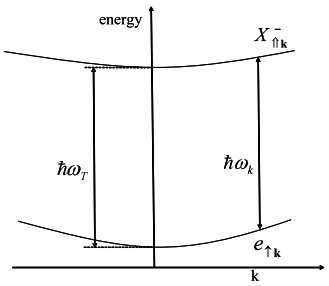

Figure 2. Energy dispersion of the conduction electrons and of the trions X^-.

constants c_1 and ϵ_1 depend on the properties of the sample. The energy $\epsilon = \hbar(\omega_T - \omega_{\mathbf{k}}) = (\hbar^2\mathbf{k}^2/2m_e)(M_X/M_T)$, where $M_X = m_e + m_h$ and $M_T = 2m_e + m_h$ are assigned to exciton and trion masses. The trion threshold transition energy $\hbar\omega_T$ corresponds to the energy difference $\hbar\omega_{\mathbf{k}=0}$, see figure 2. For the energy gap E_g and for exciton and trion binding energies B_X and B_T, respectively, the trion threshold transition energy equals $\hbar\omega_T = E_g - B_X - B_T$. In our model we consider low electron concentration, hence we treat the electron gas as nondegenerate with the Boltzmann distribution of the electron $\mathbf{k}$ pseudomomentum. Defining the electron–trion transition rate $w_{\mathbf{k}}$ by the equation (2), with $\omega_{12} = \omega_{\mathbf{k}}$ and $\Omega = \Omega_{\mathbf{k}} = 2\mathcal{E}d_{\mathbf{k}}/\hbar$, we may express the total number of transitions per unit time from the electron to the trion states by the equation

$$n_{e\uparrow} \cdot R_{e\to X^-} = \int_{\mathbf{k}} w_{\mathbf{k}}\, \mathrm{d}n_{e\uparrow\mathbf{k}}, \tag{5}$$

where $R_{e\to X^-}$ is the transition rate per one electron. The number of thermalized electrons in partition $\mathrm{d}^2\mathbf{k}$ is $\mathrm{d}n_{e\uparrow\mathbf{k}} = n_{e\uparrow} \cdot \mathcal{N}_e(k)\,\mathrm{d}^2\mathbf{k}$, where the normalized distribution function $\mathcal{N}_e(k) = (\hbar^2/2\pi m_e k_B T)\exp(-\hbar^2k^2/2m_e k_B T)$. Thus the transition rate can be expressed as

$$R_{e\to X^-} = \mathcal{P}\cdot C\frac{M_T}{M_X k_B T}\exp\left[-\hbar(\omega_T-\omega)\left(\frac{1}{\epsilon_1}+\frac{M_T}{M_X k_B T}\right)\right], \tag{6}$$

where $\mathcal{P} = c\mathcal{E}^2/2\pi$ is the laser power density and $C = c_1(2\pi p_{cv}/m_0\omega)^2(e^2/\hbar c)$.

Assuming the Boltzmann distribution also for trions [10, 15], the induced transition rate from trion to electron state $R_{X^-\to e}$ can be obtained by inserting electron mass m_e in the place of the mass of the trion M_T in equation (6). Thus the ratio of the transition rates can be expressed as

$$\frac{R_{X^-\to e}}{R_{e\to X^-}} = \frac{m_e}{M_T}\exp[\hbar(\omega_T-\omega)/k_B T]. \tag{7}$$

3.2. Exciton generation rate

The σ^+ polarized light can only create excitons with total angular momentum projection $+1$ onto the positive z axis. The probability of the exciton creation can be deduced from equation (2), with $\Omega = \Omega_{mol} = 2\mathcal{E}d_{mol}/\hbar$ and the electric dipole matrix element $d_{mol} = p_{cv}\Phi_X(0)\sqrt{\mathcal{A}_{mol}}e/m_0\omega_X$, where $\Phi_X(0)$ is the 2D 1 s-exciton envelope taken for the electron and hole at the same site, $\mathcal{A}_{mol}$ is the area of the sample per one CdTe (GaAs) molecule in the QW and $\omega_X = (E_g - B_X)/\hbar$.

In order to estimate the exciton generation rate for the light frequency beyond the resonance with exciton energy we assume the simplest model in which the dipole oscillations at frequency ω are randomly disturbed. In this case the duration of free oscillations is not fixed, but randomly distributed according to the distribution function $\rho(t) = \tau^{-1}\exp(-t/\tau)$. The rate of direct optical exciton generation, counted per one molecule, is then given by $G_{mol} = \tau^{-1}\int_0^\infty \rho(t)|a_2(t)|^2\mathrm{d}t$ [17]. For ω tuned to ω_T the frequency difference $\Delta\omega$ is equal to $\omega_T - \omega_X = B_T/\hbar$ and the exciton generation rate counted per unit of the illuminated area, $G = G_{mol}/\mathcal{A}_{mol}$ may be expressed as

$$G = \mathcal{P}\cdot\frac{2\pi\gamma(d_{mol}^2/\mathcal{A}_{mol})}{c(B_T^2+\hbar^2\gamma^2/4)}, \tag{8}$$

where we have introduced γ as the width of the exciton lineshape at half maximum, which is equal to double the reciprocal decay-time, $\gamma = 2\tau^{-1}$. Moreover, we have neglected the Rabi frequency Ω_{mol} in the denominator of equation (8), as it is small in comparison to γ as well as to $\Delta\omega$. We note that the lifetime τ does not coincide with τ_{Xrad} used in the kinetic equations (1). This is because only the excitons with wavevectors $\mathbf{k}$ from the light cone (see below) contribute to the shape of the exciton line, while τ_{Xrad} is the lifetime, which is averaged over all excitons [10].

In order to estimate the induced exciton recombination rate we note that the Boltzmann distribution of exciton $\mathbf{k}$-state occupation is reached within a few picoseconds, which is much shorter than Ω_{mol}^{-1}. Because of that we can assume a very small probability of finding excitons with $\mathbf{k}$ close to zero, which could send a photon in a perfect light propagation direction parallel to the z axis. However, the perfectly plane-wave laser pulse does not exist. For the laser beam divergence, say 10^{-3} rad, only excitons with momentum $k < 10^{-3}q$ can be stimulated to recombine by the light pulse. So, the induced transition rate from the exciton state to the crystal ground state is the product of the rate $G\mathcal{A}$ ($\mathcal{A}$ is the illuminated area of the sample) and of the probability of the occupation of exciton $\mathbf{k}$-states with $k < 10^{-3}q$. For the Boltzmann distribution of excitons the averaged probability of occupation of these states is well approximated by $f \cdot N_X$, where $f = (4\pi^2/\mathcal{A})\mathcal{N}_X(0)$, $\mathcal{N}_X(k)$ is the exciton Boltzmann distribution function and N_X is the actual number of excitons in the illuminated sample. For $T = 10$ K and $\mathcal{A} = 1$ cm^2 we have $f \approx 1.9\times10^{-11}$ in the CdTe QW and $f \approx 1.2\times10^{-11}$ in the GaAs QW. For the laser power densities actually used, the induced exciton recombination rate fG is negligibly small when compared to the rate of spontaneous radiative exciton decay which is of the order of 0.01 ps^{-1} [15, 18]. Hence we neglect the induced exciton recombination in our calculations.

4. Phenomenological time constants

Besides the calculated rates given in part 3, several time constants are needed in order to solve the kinetic equations. The values taken from the literature are used and their physical origin is not discussed.

The radiative lifetimes of thermalized excitons τ_{Xrad} have been calculated by Andreani *et al* [19], and later for excitons and trions τ_{Trad} by Esser *et al* [10]. They have obtained a linear temperature dependence for the exciton as well as for the trion lifetime. Ciulin *et al* [20] measured the trion lifetimes in the range from $T = 0$ K (40 ps) to $T = 32$ K (200 ps) confirming Esser's [10] prediction. However, in the case of excitons, there is no agreement between calculated [10] and measured [15, 18] exciton decay-times, particularly for $T \approx 0$. At low temperatures the theoretically predicted decay-time should be close to zero, while the measured decay-time exceeds 100 ps. This divergence is interpreted as the effect of exciton localization in the QW.

The formation time of the trions X^- decreases with increasing electron concentration n_0. For the lowest concentration of the order of 10^{10} cm^{-2} it was found to be 500 ps at $T = 2$ K in CdTe based QWs, while for $n_0 = 5 \times 10^{10}$ cm^{-2} this time decreases to less than 40 ps. The spin relaxation time of the hole τ_{h} contained in the trion X^- is equal to about 35 ps for the excitation energy near the $\hbar\omega_{\mathrm{T}}$ threshold. The above times were given by Kossacki for the CdTe QW [21] and they correspond to a 100 Å-wide CdTe/$Cd_{0.75}Mg_{0.25}$ Te heterostructure with a remote donor layer of iodine located 10 nm from the QW. In GaAs based QWs (250 Å-wide GaAs/$Al_{0.3}Ga_{0.7}As$ QW) X^- formation time has been observed by Finkelstein *et al* [22] for $n_0 = 2 \times 10^{11}$ cm^{-2}. It was found to be 150 ps at $T = 5$ K and ≃400 ps at $T = 10$ K. Because of the lack of experimental data for τ_{h} in GaAs we have used in this case the same value as that given for CdTe. In our kinetic description of the spin pumping we simply assume the linear dependence of the formation time on the electron concentration, in the range of available experimental data.

As we have stated in section 1, we use for the electron spin relaxation time $\tau_{\mathrm{s}} = 5$ ns which was measured by Hoffmann *et al* [13] in a 200 Å-wide CdTe/$Cd_{0.78}Mg_{0.22}$Te QW. We would like to mention that longer times have been reported, for example 14 and 19 ns, in the paper of Astakhov *et al* [23]. However, we use the shorter time to show that even in this case a significant spin polarization can be achieved. For GaAs QWs we use $\tau_{\mathrm{s}} = 10$ ns measured by Dzhioev *et al* [9] in a 100 nm-thick GaAs layer capped by a 25 nm-thick $Al_{0.3}Ga_{0.7}As$. The exciton spin-flip times were found by Tribollet *et al* [14] for CdTe QWs (18–36 ps) and Le Jeune *et al* [24] for GaAs QWs (40 ps). They are, according to the assumptions of our model, much shorter than the radiative lifetimes. They also are very much shorter than the reciprocal of the excitons' generation rate, which is for example for $\mathcal{P} = 50$ W cm^{-2} and $\omega = \omega_{\mathrm{T}}$ equal to $(G/n_0)^{-1} \approx 1.2 \times 10^3$ ps in the CdTe QW case with $n_0 = 10^{10}$ cm^{-2} and $(G/n_0)^{-1} \approx 6.7 \times 10^3$ ps in the GaAs QW case with $n_0 = 10^{11}$ cm^{-2}. The assumption of equal populations in different exciton states, which is used in our model, is then justified.

All the above-given phenomenological time constants are typical for QW widths 100–300 Å and barrier heights 200–300 meV. The electron spin relaxation time τ_{s} strongly depends on the quality of the interfaces. The abrupt QW interfaces assure longer τ_{s}.

5. Results and conclusions

In our calculations we assume a low concentration of the confined electrons, thus the electron gas can be treated as nondegenerate. To make the spin pumping process effective we choose a sufficiently low temperature to assure a significant trion excitation rate $R_{\mathrm{e}\to\mathrm{X}^-}$ for moderate laser power density. At the same time we must keep the significant predominance of the trion excitation rate over the induced transition rate $R_{\mathrm{X}^-\to\mathrm{e}}$. These demands, together with the relation given by equation (7), imply the inequality $T > \hbar(\omega_{\mathrm{T}} - \omega)/k_{\mathrm{B}} \ln(M_{\mathrm{T}}/m_{\mathrm{e}})$. In consequence, for the mass ratio $m_{\mathrm{e}}/M_{\mathrm{T}} \simeq 0.25$–$0.30$ and for a finite dispersion energy of the laser pulse (assumed to be $\Delta\hbar\omega \simeq 0.1$ meV) we must keep $T > 1$ K in our equations. On the other hand the upper temperature limit is determined by the trion dissociation energy.

The material parameters used in the estimations of the induced transitions rates are the following for a CdTe (GaAs) QW: $m_{\mathrm{e}} = 0.099m_0$ ($0.067m_0$), $m_{\mathrm{e}}/m_{\mathrm{h}} = 0.45$ (0.29), $E_{\mathrm{g}} \simeq 1.645$ eV (1.528 eV), refractive index $n = 3.26$ (3.54), Kane's matrix element $2p_{\mathrm{cv}}^2/m_0 = 21$ eV (22.7 eV), $\epsilon_1 = 1.95$ meV (1.1 meV), $c_1 = 18.6$ (15.), exciton binding energy (used to estimate the exciton Bohr radius $a_{\mathrm{B}} = \hbar\sqrt{2}/\sqrt{\mu B_{\mathrm{X}}}$ which determines $\Phi_{\mathrm{X}}(0)$) $B_{\mathrm{X}} \simeq 20$ meV (≃7 meV), trion X^- binding energy $B_{\mathrm{T}} = 2.9$ meV (≃1 meV) and the exciton line-width $\hbar\gamma = 0.7$ meV [20] (0.3 meV [10]).

The electron spin polarization for the CdTe QW as a function of illumination time is presented in figure 3. The obtained results correspond to the temperatures 12 and 24 K and to the electron concentration $n_0 = 10^{10}$ cm^{-2}. As an initial condition at $t = 0$ equal populations of spin electron states, i.e. the concentrations $\mathrm{e}_\uparrow = \mathrm{e}_\downarrow = 1/2$ have been assumed. After some time a steady state of the spin polarization is reached independently of laser power density $\mathcal{P}$. Because of a risk of sample heating, $\mathcal{P}$ should be limited to about 50 W cm^{-2}. In the range of $\mathcal{P}$ from zero to 50 W cm^{-2} we observe an increase of the steady polarization. The final polarization depends on temperature and for $T = 12$ K and $\mathcal{P} = 50$ W cm^{-2} we obtain the concentration of electrons with antiparallel spin $\mathrm{e}_\downarrow = 82\%$ while $\mathrm{e}_\uparrow = 10\%$. In the same time the rest (8%) of the excess electrons are confined in trions. This corresponds to the spin polarization $P = (\mathrm{e}_\downarrow - \mathrm{e}_\uparrow)/(\mathrm{e}_\downarrow + \mathrm{e}_\uparrow) \approx 78\%$. At higher temperature ($T = 24$ K in figure 3, bottom line) the final polarization is lower, and corresponds to 72% electrons with antiparallel spin ($P \approx 62\%$). The diminishing of spin polarization with rising temperature is mainly connected with the temperature dependence of the transition rate $R_{\mathrm{e}\to\mathrm{X}^-}$ given by equation (6).

As we have mentioned above, the use of power higher than 50 W cm^{-2} can lead to heating of the sample. In spite of this, we study the solutions of our equations also in the range exceeding 50 W cm^{-2}. We observe that increasing the power would not necessarily lead to a higher final steady polarization. This can be explained as follows: In a steady state the exciton concentration X is proportional to the rate $G(\sim\mathcal{P})$ (see the fifth equation in the set of equations (1)). Neglecting, for simplicity, the small terms $(\mathrm{e}_\downarrow - \mathrm{e}_\uparrow)\tau_{\mathrm{s}}^{-1}$ in

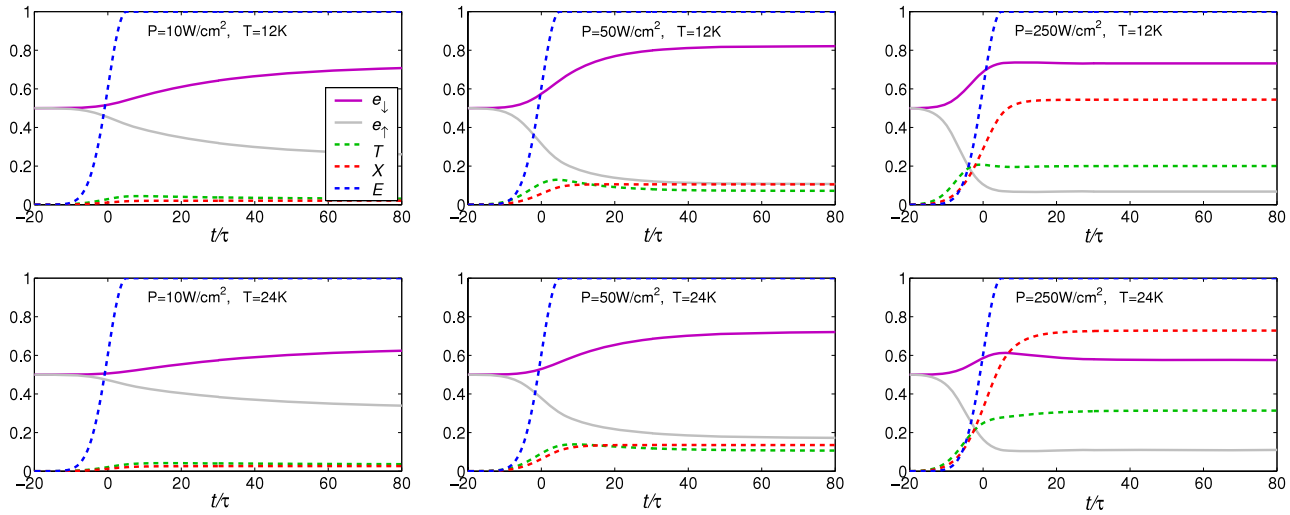

Figure 3. The electron spin pumping in the CdTe QW with electron concentration $n_0 = 10^{10}$ cm^{-2} and varying illumination power density. The time unit $\tau = 40$ ps is chosen to be equal to the radiative lifetime of the trion at $T = 0$ K. The electric field modulus $\mathcal{E}$, normalized to its maximum value, is the blue (dashed) curve. Other colour curves correspond to $e_\downarrow$ (violet), $e_\uparrow$ (grey), $T = T_\Uparrow + T_\Downarrow$ (dashed green), X (dashed red denote total concentration of excitons). The presented results are obtained for $T = 12$ K (24 K), $\tau_s = 5$ ns, $\tau_{Trad} = 105$ ps (167 ps), $\tau_{Xrad} = 165$ ps (250 ps), $\tau_h = 35$ ps and $\tau_{Tform} = 500$ ps.

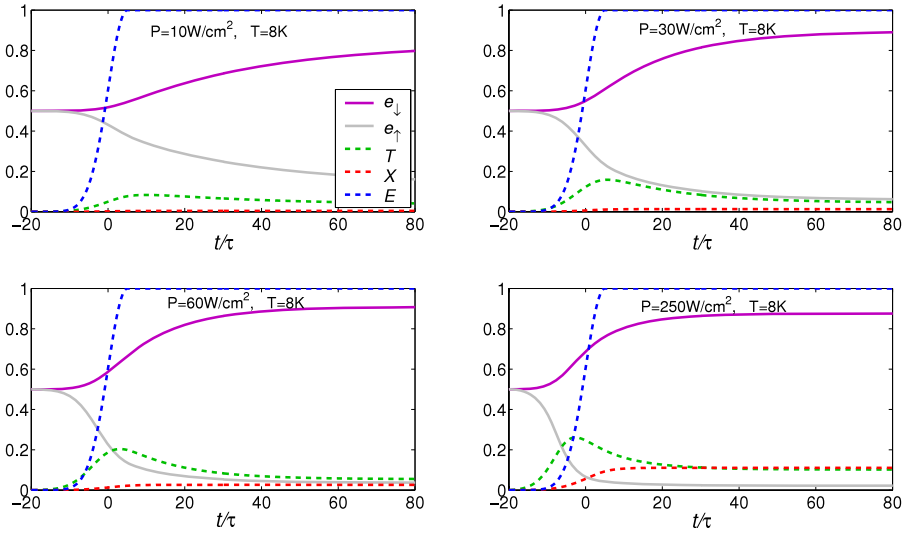

Figure 4. The electron spin pumping in the GaAs QW for different illumination power densities (time unit $\tau = 40$ ps). The results are obtained for the following data: $T = 8$ K, the electron concentration $n_0 = 10^{11}$ cm^{-2}, $\tau_s = 10$ ns, $\tau_{Trad} = 200$ ps, $\tau_{Xrad} = 200$ ps, $\tau_h = 35$ ps and $\tau_{Tform} = 600$ ps.

equation (1) and consequently assuming that $T_\Uparrow = T_\Downarrow = T$, we get $T/e_\downarrow = X\tau_{Trad}/\tau_{Tform}$. This means that higher concentration of excitons implies a higher share of the trions in the conservation condition $e_\uparrow + e_\downarrow + T_\Uparrow + T_\Downarrow = 1$. Thus the presence of excitons limits the polarized electron concentration to $e_\downarrow < 1/(2X\tau_{Trad}/\tau_{Tform} + 1)$. To illustrate this mechanism we present in figure 3 the results also for $\mathcal{P} = 250$ W cm^{-2} where we can observe that the rise of exciton concentration accompanies the reduction of $e_\downarrow$. Studying the solutions in the broad range of $\mathcal{P}$ we have found that the final polarization does not noticeably rise above that obtained for $\mathcal{P} = 50$ W cm^{-2}.

We have also performed calculations for other values of electron concentration n_0, up to 5×10^{10} cm^{-2}, and we have found that in all cases the polarization corresponding to the laser power density $\mathcal{P} \simeq 50$ W cm^{-2} is close to its maximum value. In particular, when $n_0 = 5 \times 10^{10}$ cm^{-2} we have the highest $e_\downarrow \approx 84\%$ ($P \approx 79\%$) for $T = 12$ K and 76% ($P \approx 65\%$) for $T = 24$ K. Comparing these results with the best result which we achieved for $n_0 = 10^{10}$ cm^{-2} we can see that $e_\downarrow$ is larger for higher concentration n_0. It results from the smaller relative generation rate G/n_0 of excitons.

In figure 4 we present the results for the GaAs QW. Similarly to the case of CdTe, we observe that the steady polarization is not a monotonic function of the power density $\mathcal{P}$. The highest concentration $e_\downarrow \approx 92\%$ (spin polarization $P \approx 91\%$) is reached now for the laser power density $\mathcal{P} \simeq$

60 W cm^{-2}. The higher polarization (comparing to CdTe) is connected mainly with a twice as long electronic spin lifetime τ_s. As we have mentioned before, we have used in our calculations the same value of τ_h as for the CdTe QW. We have checked that this is not a bad choice because the calculated spin polarization is practically insensitive to τ_h in a broad range around this value (only for $\tau_h > 200$ ps does the efficiency of electron spin polarization depend strongly on this spin-flip time).

After the laser illumination is switched off, the polarization P depends mainly on the electron spin relaxation time and decays exponentially as $P(t) \approx P(0)\exp(-2t/\tau_s)$. Thus, for example, for $P(0) = 80\%$ and after $t = 1$ ns we still can expect the polarization $P(1\text{ns}) \approx 65\%$ (provided $\tau_s = 10$ ns).

In conclusion, the electron spin pumping by laser illumination seems possible in a relatively broad range of low temperatures. For the laser power density $\simeq$50–60 W cm^{-2} the concentration of spin polarized electrons may exceed 80% of n_0. For the higher laser power, when exciton concentration dominates that of trions, the electron spin polarization is damped because of the destructive processes of the trion formation from electrons and neutral excitons. The general requirements of the presented model (concerning material properties) are the following: (a) long spin relaxation time of the conduction electron, (b) relatively short spin relaxation time of the hole in the trion X^-, (c) relatively short time of the spontaneous radiative decay of the trion. The remaining conditions are: (a) low temperature assuring a sufficiently high rate of the electron to trion X^- transitions and (b) suitably high excess electron concentration ($n_0 \geqslant 10^{10}$ cm^{-2}) to limit the destructive role of neutral excitons in the spin pumping process.

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